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Additional Information

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14	Abstract
15	Several advanced combustion techniques have been developed to reduce the harmful
16	emissions from diesel engines. Among these, the low-temperature combustion (LTC)
17	strategies are very important. These LTC strategies, such as homogeneous charge
18	compression ignition (HCCI), premixed charge compression ignition (PCCI), reactivity-
19	controlled compression ignition (RCCI), are able to reduce engine-out nitrogen oxides
20	(NOx) and soot emissions simultaneously. LTC investigations exhibit several limitations
21	of HCCI and PCCI combustion modes, such as lack of combustion control and other
22	operational issues at higher engine loads, making their application in production-grade
23	engines challenging. RCCI combustion combustion modeexhibited promising results in

terms of combustion control, engine performance, and applicability at higher engine loads. 24 25 The potential of the RCCI concept was demonstrated on different engine platforms, 26 showing engine-out NOx levels below the limits proposed by the emissions regulations, together with ultra-low soot emissions, eliminating the need for using after-treatment 27 devices. However, the RCCI combustion mode has several challenges, such as excessive 28 29 hydrocarbons (HC) and carbon monoxide (CO) emissions at low loads and excessive 30 maximum pressure rise rates (MPRR) at high loads, which limit its effective operating range and practical applications. This review article includes recent advancements in 31 32 RCCI combustion mode, its potential for using alternative fuels, effects of different 33 parameters on RCCI combustion mode and its optimization, and the ability of RCCI mode 34 to extend the engine operating limit to cater to higher loads, which prevents the application of this concept in commercial applications. The findings of different optical 35 diagnostics have been also included, which have been performed to understand the 36 detailed chemical kinetics of the fuel-air mixtures and the effect of fuel reactivities on the 37 38 RCCI combustion combustion mode. The first part of this article focuses on these studies, 39 which provide important outcomes that can be used for the practical implementation of 40 RCCI combustion combustion modein production-grade engines. The second part of this article covers different RCCI combustion combustion modestrategies that can be used to 41 eliminate the restrictions of RCCI combustion modeat high loads. Among the different 42 43 techniques, dual-mode concepts are being extensively investigated in recent years. The 44 dual-mode concept is based on switching between two different combustion modes, typically an LTC mode and conventional compression ignition (CI) combustion mode, to 45 cover the entire operational landscape of the engine. Many studies showed that the NOx 46 47 and soot emissions from stationary engines with dual-mode RCCI/CI combustion had substantially improved versus a single-fueled CI combustion mode engine. Results related 48 to the measurements of emissions and performance in transient conditions and driving 49

50 cycles have also been included, which exhibit promising results for RCCI mode. A 51 comprehensive review on overcoming the challenges and real-world applicability of RCCI 52 combustion mode is unavailable in the open literature. To fill this research gap, this article also includes the results of relevant RCCI combustion mode investigations carried 53 54 out in single-cylinder and multi-cylinder engines. Finally, the results from alternative 55 RCCI combustion mode concepts such as the dual-mode, hybrid-RCCI, simulations, and 56 experiments in transient conditions using various driving cycles make this article uniquely relevant for researchers. 57

58 Keywords: Reactivity-controlled compression ignition; Alternative fuels; Optical
59 diagnostics; Simulations; Reactivity gradient.

60 1. Introduction

61 The internal combustion engines (ICE) were conceptualized in 1876 [1], more than 145 years ago, and they have played an extremely important role in human development by 62 63 catering to power generation and transportation. Fossil fuels have traditionally been the 64 main energy source to power the ICEs; however, it is well documented that their 65 production, exploitation, and use a generate negative environmental impact [2-3] and 66 several health issues associated to the air pollution generated [4]. The energy landscape 67 has exploded in recent years, giving rise to newer options such as synthetic fuels, fuel 68 cells, electric and hybrid vehicles, and renewable fuels. These are aimed to reduce the 69 negative impact of pollutants from the transport sector, which currently account for 70 ~16.2% of the global greenhouse gas (GHG) emissions [5]. Additionally, to prevent the dire effects of GHG emissions and achieve the goal of a climate-neutral economy by 2050, 71 72 emissions regulators have imposed stricter emission limits in the transport sector [6-8], 73 thus accelerating the demand from the automotive manufacturers for a cleaner engineout exhaust from vehicles. Hence, an accelerated optimization of ICEs and introduction 74 75 of efficient combustion concepts are vital for reducing emissions in the foreseeable future.

This section deals with these aspects related to the automotive sector and presents thecurrent scenario and possible solutions to deal with these burning issues.

78 1.1 Current Automotive Scenario and Challenges

79 Knowing that in the short-to-medium term, ICEs would continue to move the world and 80 remain an important power plant for vehicles in the transport industry, active research 81 and development for the improvement in fuel consumption and reduction in pollutants 82 (namely carbon monoxide (CO), hydrocarbon (HC), oxides of nitrogen (NOx), and 83 soot/particle matter (PM)) would continue to be important. In particular, the evolution of compression ignition (CI)engines with direct injection (DI) of high cetane fuels would be 84 85 important since they are highly efficient, widely used, and produce relatively lower CO 86 and HC emissions than their spark ignition (SI) engine counterparts. On the other hand, 87 one of the main challenges for these direct injection compression ignition (DICI) engines 88 would be higher NOx and PM emissions due to their higher combustion temperatures [9-89 10] and fuel-rich regions [11-12], respectively. A close relationship among these pollutant 90 species is another challenge because of a trade-off between them. Preventive measures to 91 control one of them promote the formation of the other [13]. Several routes have been 92 discussed to control the emissions of these pollutants, such as the use of alternative fuels 93 [14], exhaust after-treatment systems [15], improving combustion, and using advanced 94 combustion concepts [16]. The advanced combustion concepts have potential among these options since they provide several benefits over conventional diesel combustion (CDC) 95 96 without using after-treatment devices in the exhaust. The use of after-treatment systems 97 increases vehicle production and operational costs, fuel consumption, engine complexity, 98 and requires frequent maintenance [17-18]. Among several advanced combustion 99 techniques, the reactivity controlled compression ignition (RCCI) is a dual-fuel 100 combustion concept aiming to simultaneously reduce the engine-out soot and NOx

101 emissions while maintaining the engine performance and efficiency comparable to the102 CDC [19].

103 1.2 Need for Alternative Combustion Concepts

Between 1990 to 2017, global GHG emissions were reduced by ~21.7% (4483 megatons of 104 105 CO_2 equivalent (CO₂e)) [6]. In 2018, the energy sector reduced CO₂e emissions by 22.2% 106 with relation to 1990; however, the transport subsector (the category that provides 107 movement of humans, animals and goods from one location to another) GHG emissions 108 increased~20% with relation to1990 [6] (Figure 1). Among the GHG emissions from the 109 transport sector, road transport contributed 73.45% [5] (Figure 2). A clear impact of the activities in this sector underscores the need for emission regulations such as Euro-VI for 110 111 light-duty and heavy-duty vehicles and subsequent strategies to develop efficient ICEs that can achieve the target GHG emissions by cutting down on carbon dioxide (CO_2) 112 emissions. Hence, a combination of newly developed combustion strategies such as low 113 temperature combustion (LTC) and alternative fuels will be hugely beneficial since 114 115 electrification on its own does not guarantee decarbonization if the main sources of electricity generation are non-renewables [20-21]. 116





Figure 1: GHG emission trends, projections, and targets [6].

CDC has excellent efficiency and engine performance, indicating that its fuel-to-energy 119 120 conversion is effective [22]. The problem lies in the engine-out emissions. Reducing 121 emissions will then get the ICE to an acceptable emission footprint. The emissions control 122 techniques have traditionally been deployed on two fronts: (i) active control strategies using after-treatment systems and (ii) passive control strategies to control pollutant 123 124 formation inside the engine's combustion chamber. On the after-treatment side, diesel 125 oxidation catalysts (DOC), diesel particulate filters (DPF), and selective catalytic 126 reduction (SCR) have been deployed to comply with prevailing emission standards. 127 However, they have some disadvantages, which have already been mentioned in section 128 1.1. It is worth restating that reducing emissions during the in-cylinder formation stage 129 would reduce the after-treatment requirement, thus minimizing overall system complexity and engine cost. Traditionally, a large number of pollutant formation control 130 131 strategies – such as optimized fuel injection strategies including the use of high fuel 132 injection pressures (FIP), injection timing optimization, multiple fuel injections, use of 133 exhaust gas recirculation (EGR), increased in-cylinder turbulence (increased in-cylinder 134 motion and turbocharging), and redesign of the combustion chamber and injection system 135 -have been deployed with varying degrees of success for controlling the NOx and the soot emissions. A combined in-cylinder combustion optimization and after-treatment device 136 approach has also been explored to control the emissions to meet the regulatory 137 138 requirements [23-24]. On their own, however, these methods struggle primarily because 139 of the intrinsic trade-off between the soot and the NOx in diesel engines since, more often 140 than not, managing to reduce one pollutant increases the other. Primarily, NOx is formed 141 through a thermal mechanism (although other mechanism also exist and can be prevalent 142 during some combustion modes) where nitrogen and oxygen inside the cylinder mixture combine with one another in presence of high combustion temperatures and enough 143 residence time. Because of this, reducing the combustion temperatures and duration can 144

reduce NOx. Nonetheless this has a counter effect on soot, as the temperatures and 145 146 residence times are not high enough to burn off and reduce soot particles. Advanced 147 combustion strategies that deal with the soot-NOx trade-off have been developed over time and can also integrate the use of more advanced fuels with properties to mitigate 148 these emissions. In particular, many LTC strategies have been developed, which improve 149 150 the fuel-to-work conversion efficiency while providing low soot and NOx emissions [25]. It 151 is worth noticing that, to avoid severe global warming effects, CO₂ equivalent emissions need to be reduced by more than half the current levels by 2050 [26]. To reach this 152 153 emission goal a single pathway is not enough, thus it is important to address more than 154 one strategy at the time. Another path that has been explored from more than two decades 155 is the use of oxygenated fuels to reach practically soot-free emissions [27-28], however the increase in the fuel's oxygen content (as with some alcohols) can lead to increase NOx 156 157 emissions and peak pressures if strategies that address those issues are not utilized [29-158 30]. For other alternative fuels, as their characteristic properties also differ from the 159 commercially available fossil fuels, dedicated calibrations and strategies also need to be 160 developed. However, research has shown that combining both the LTC and alternative 161 fuels in the same system can provide advantages in both emissions and engine efficiency.





Figure 2: Global greenhouse gas emissions, sector-wise [5].

164 **1.3 Evolution of Different LTC Concepts**

In the past few decades, LTC modes have gained significant attention from researchers 165 (correlated to the increasing number of publications in the field) due to their excellent 166 capabilities in terms of emission reduction, especially NOx and PM. LTC modes cover 167 several advanced combustion strategies, including homogeneous charge compression 168 ignition (HCCI), partially premixed charge compression ignition (PPCCI), premixed 169 170 charge compression ignition (PCCI), which have the potential to reduce NOx and PM emissions simultaneously without a "PM-NOx dilemma," that remains a critical issue in 171 CDC engines [31-33]. In all LTC modes, relatively lower in-cylinder combustion 172 temperatures are common, which is the main reason for extremely low NOx emissions. To 173 explore other features of LTC, Akihama et al. [34] applied ultra-high exhaust gas 174 recirculation (EGR) in a conventional diesel engine. The objective of this study was to 175

explore the potential of EGR for the simultaneous reduction of NOx and PM. They
reported a 'soot-NOx dilemma' in conventional diesel engines, which could be avoided only
by achieving the combustion outside the NOx and soot zones in Figure 3.





180

Figure 3: ϕ -T diagram for identification of soot and NOx emission zones [34].

The authors indicated that smokeless engine combustion could be achieved for any 181 equivalence ratio at in-cylinder temperatures below 1500K. However, a more sensitive 182 behavior of soot emissions was observed at in-cylinder temperatures above 2000K. They 183 184 reported that soot formation increased rapidly at excessively richer mixtures. The NOx 185 formation increased significantly at relatively lower loads (1< ϕ <2). In this regard, Beatrice et al. [27] reported an important finding that oxygenated fuels exhibit a different 186 soot-NOx characteristics compared to conventional diesel. They reported that the use of 187 oxygenated fuels along with high EGR can result in a drastic reduction of both soot as 188 well as NOx. Various LTC methodologies were extensively explored by researchers 189 worldwide to meet stringent emission norms. LTC strategies, as the name indicates, 190 191 employ combustion at lower temperatures. Therefore, it involves reducing flame temperature and allowing sufficient air-to-fuel mixing to increase the fuel-air mixture 192 homogeneity [35]. As mentioned earlier, this method theoretically reduces NOx and soot, 193

because these pollutants are strongly influenced by the flame temperature and the equivalence ratio, [36] while preserving the thermal efficiency. This concept virtually combines the benefits of CI and SI engines. The necessary in-cylinder conditions can be achieved by early fuel injections [37], improved fuel atomization [38], or even using fuels with lower cetane number [39-40], alternative fuels, or a combination of diesel- and gasoline-like fuels [41]. This allows more time for fuel-air mixing before the start of combustion (SoC).

Similarly, EGR-controlled combustion can trigger this combustion mode with injections 201 202 close to the top-dead-center (TDC) [42]. LTC addresses the conventionally inferior 203 capacity of diesel-fueled engines to prepare a premixed fuel-air mixture before the SoC, 204 caused by the higher fuel viscosity and lower fuel volatility [25]. Similar to other combustion strategies, the LTC can use a wide variety of fuels providing an additional 205 degree of freedom when controlling the combustion. Alternative fuels reduce dependence 206 207 on petroleum reserves [43] and open up the usage of new fuels with special properties 208 such as higher fuel oxygen or the absence of carbon, which by having extremely low soot 209 emissions can increase the tolerance of the fuel-air mixture to higher proportions of EGR 210 to also reduce NOx. Two key concepts emerge while referring to this advanced combustion method: (i) reduced in-cylinder temperature and (ii) improved in-cylinder fuel-air mixture 211 formation. NOx-soot trade-off associated with the conventional CI combustion mode is 212 213 another important aspect, which promotes the development of LTC. Even though the LTC strategies address the NOx-soot trade-off issues and maintain conventional diesel-like 214 215 efficiencies, they have challenges as well, such as higher CO and HC emissions [44], lack 216 of ignition timing control over the high and low load extremes [45], and increased 217 combustion noise [46].

A broad range of technologies is covered in the ambit of LTC, including HCCI, PCCI,
partially premixed combustion (PPC), premixed compression ignition (PCI), gasoline

compression ignition (GCI), and RCCI. Among these, RCCI combustion mode is the
specific strategy that is extensively reviewed in this article. Besides these combustionspecific methods, integration of LTC in hybrid powertrains has also been explored [47].
Among many LTC strategies, few are described here, which have assisted in the evolution
of the RCCI combustion mode concept, focusing on their capabilities, the drawbacks
encountered, and how they have helped the evolution of the RCCI combustion mode.

226 1.3.1 HCCI Combustion Concept

227 HCCI is the most common technique for achieving LTC by combining the premixing of 228 fuel (similar to SI engines) and then compression ignition (similar to diesel engines) [48]. 229 Originally coined by Najt and Foster [49], it consists basically of homogeneous mixtures 230 of fuel, air and EGR. An early injection of fuel to form a well-mixed charge is required to achieve HCCI combustion in gasoline engines. In diesel-engines, early direct injection of 231 fuel in the intake stroke can be implemented, which provides enough time for fuel-air 232 233 mixing in the combustion chamber. The fuel-air mixture is compressed in the compression 234 stroke, increasing its temperature up to the autoignition temperature. Volumetric 235 combustion is initiated at multiple sites in this homogeneous fuel-air mixture. The 236 ignition is controlled by the composition of the fuel-air mixture and the in-cylinder temperature. A lean and homogeneous fuel-air mixture auto-ignites without a spark 237 238 because of the increased in-cylinder pressure at the end of the compression stroke. Local 239 temperatures are kept at low levels, and there is no high-temperature flame front [50]. It 240 experiences lower throttling losses, favoring higher thermal efficiency [51], fuel versatility [52], and lower emissions of CO₂ [53], NOx, and PM [54]. Due to these attributes, HCCI 241 242 combustion mode ensures smoother LTC, leading to significantly lower NOx and PM emissions [55]. The gasoline HCCI concept was first demonstrated by Onishi et al. [56] in 243 244 1979, who named it 'Active Thermo-Atmosphere Combustion (ATAC).' They successfully achieved stable combustion in a gasoline-fueled 2-stroke engine and reported significant 245

improvement in emission characteristics, engine noise, and engine vibrations. They 246 247 suggested that relatively leaner mixture combustion was the main reason for these 248 observations. However, the applicability of this novel combustion technique only up to 249 part-load was a major limitation. After this pioneering work, several researchers, including Thring [57] (1989), Christensen et al. [58] (1997), Stanglmaier et al. [59] (1999), 250 251 and Maurya et al. [60] (2011), successfully demonstrated this combustion concept in gasoline-fueled engines. In most of these research studies, gasoline-like fuels were used 252 to achieve HCCI combustion, demonstrating superior emission characteristics to the 253 254 conventional combustion modes. Researchers also explored HCCI combustion using 255 diesel-like fuels to resolve the NOx-PM trade-off observed in CDC engines. Some of the 256 most influential factors were the injection strategies [61] and other characteristics of the fuel injection system, such as an injector and the injection pressure [62]. The main 257 challenges in achieving diesel-fueled HCCI combustion were achieving a homogeneous 258 259 mixture, fuel wall-wetting, controlling the auto-ignition, and excessive pressure rise rates 260 (PRR). In particular, the PRR was crucial to avoid potential damage to the engine 261 components and control the noise and vibration issues [63]. The combustion control brings 262 another set of issues. Combustion can be affected by small variations in the charge composition or system temperature [64]. Singh et al. [65] reported combustion 263 characteristics of diesel-fueled HCCI combustion in which they used an external mixture 264 265 preparation device for preparing a homogeneous charge. They reported that combustion characteristics of HCCI combustion mode were dominantly affected by the charge 266 267 homogeneity. They also explored the performance and emission characteristics of diesel-268 fueled HCCI combustion engines and reported superior emissions characteristics but slightly degraded engine performance. A detailed comparison of particulate emission 269 characteristics of diesel-fueled HCCI combustion engine and CDC engine exhibited lower 270 PM and trace metals from the HCCI combustion mode [66]. In another study carried by 271

Singh et al. [67], biodiesel blends were used to achieve the HCCI combustion mode. They 272 273 reported almost similar HCCI combustion characteristics of biodiesel blends as that of 274 mineral diesel. However, relatively inferior biodiesel properties affected the engine performance and emissions adversely. Several test fuel blends were also used to explore 275 the effects of fuel properties on the HCCI combustion engine, including mineral diesel-276 277 alcohol, mineral diesel-gasoline, mineral diesel-kerosene, and mineral diesel-biodiesel. 278 They reported that fuel properties, especially volatility, affected the HCCI combustion, 279 resulting in superior engine performance and emission with increased fuel volatility. Nonetheless, HCCI is one of the few LTC concepts that has been commercially 280 281 implemented, as is the case with the Mazda Skyactiv X engine, which uses a strategy 282 denominated spark controlled compression ignition (SPCCI), which uses a lean burn compression ignition combustion with gasoline, a spark-ignited local combustion (to 283 284 increase in-cylinder temperature an propitiate auto-ignition) and a high compression ratio, which develops in a HCCI combustion that is stable and improves fuel economy by 285 286 around 5% [68].

287 Although the HCCI combustion exhibited a sharp reduction in both NOx and PM 288 emissions, relatively higher HC and CO emissions due to lower in-cylinder temperatures, the use of high EGR adversely impacts the popularity of HCCI combustion for deployment 289 290 in production-grade engines. This also leads to an overall reduction in the thermal efficiency of the HCCI combustion engines. Lack of direct combustion control and 291 excessive PRR are two critical challenges of HCCI combustion, limiting its application at 292 293 higher engine loads. This issue assumes a more serious dimension in the mineral diesel-294 fueled HCCI engine due to its relatively lower volatility and lower auto-ignition 295 temperature. It has been reported in several studies that mineral diesel-fueled HCCI 296 combustion results in either too advanced or too retarded combustion phasing, leading to

297 lower thermal efficiency. These drawbacks motivated researchers to develop a new LTC298 strategy, known as PCCI combustion mode.

299 1.3.2 PCCI Combustion Concept

PCCI combustion mode evolved from the HCCI mode to address the difficulties and 300 301 challenges of HCCI. Like its predecessor, PCCI mode reduces soot and NOx, but not as much as the HCCI mode [69]. HC and CO emissions can be lower under PCCI combustion 302 mode than with the HCCI mode [70]. Unlike HCCI mode, the PCCI combustion mode is 303 not fully homogeneous. Additionally, control of the SOC and combustion duration 304 305 improves because of charge dilution by higher EGR rates to delay the ignition and 306 increase the mixing duration [71]. Higher EGR levels affect combustion stability at high 307 engine loads, which could be addressed by early injections strategies. However, this has 308 a counter-effect of reducing the thermal efficiency [72]. Another negative effect of using high EGR rates is the increased fuel consumption and higher CO and HC emissions [73] 309 and PM emissions due to the reduced availability of in-cylinder oxygen [16]. Musculus et 310 311 al. [74] proposed conceptual models for LTC that describe the spray formation, mixing, 312 ignition and pollutant formation within the concept. From their analysis, they stated that 313 both HC and CO are highly dependent on the mixture distribution formed, and that a narrow range of equivalence ratios providing a low CO yield can make this emission more 314 problematic than the HC emissions in systems that have early-injection PCCI. Solutions 315 such as increased boost pressure [70, 75], high fuel-injection pressure, multiple injections 316 317 to reduce the maximum heat release rate (HRR), and enhanced mixing to reduce combustion noise and soot [76] have been proposed to extend the engine operating range. 318 319 The effect of EGR and variations in injection parameters on the PCCI combustion had been extensively studied by various researchers worldwide [77-80], with a common 320 321 observation of a simultaneous reduction in NOx and PM by early injection timings. This led to a slight increase in brake-specific fuel consumption (BSFC). Torregrosa et al. [79] 322

reported that a reduction in the combustion noise could be attained by using a pilot injection in the PCCI engine; however, brake mean effective pressure (BMEP) decreased significantly as the pilot injection quantity was increased. Emission reduction by variable valve timing (VVT)and early start of injection (SoI) timing as 46 and 30°bTDC were achieved by Murata et al. [69] and Torregrosa et al. [79], respectively, at the cost of a significant reduction in the engine torque.

329 Combustion in PCCI can be realized using a variety of fuels [81], such as fuel blends with lower cetane numbers (CN), e.g., gasoline, to enhance the charge mixing before the SoC 330 331 [82] and to extend the engine operating range [83]. Fuels with high CN are favorably 332 combined with the extended ignition delay of the PCCI strategy. Lilik and Boehman [84] 333 tested two synthetic diesel fuels produced by a low and high temperature Fischer-Tropsch process, with 81 and 51 CN respectively. They found that HC and CO emissions could be 334 decreased by 32% and 31% respectively to the CN 51 fuel compared to diesel and 80% and 335 74% respectively to the CN 81 fuel. CN 81 fuel also maintained PM and NOx emissions 336 337 at the same level as the conventional diesel due to the shorter and less intense premixed 338 combustion phase that preceded a mixing controlled combustion phase. Alternative fuels 339 such as alcohol blending seem feasible for achieving more efficient and controllable PCCI combustion. Some researchers used ethanol-diesel blends in CI engines to attain low-340 temperatures during combustion. Mohammadi et al. [78] explored the possibility of very 341 clean combustion using ethanol in a partial PCCI combustion engine. They realized that 342 343 the utilization of cooled EGR with early pilot injection leads to a significant PM-NOx 344 trade-off. Park et al. [80] investigated bioethanol blended diesel with a narrow spray angle 345 injection strategy in a conventional diesel engine. They reported that premixed combustion phasing decreased with increased bioethanol content in the fuel. A significant 346 347 reduction in HC and CO emissions was also observed. More recent works have evaluated bio-origin fuels such as hydrotreated vegetable oil (HVO) [85] and diesel/biodiesel blends 348

under the combustion mode [86]. In [85], the sensitivity to the mixture dilution and 349 350 conditions is evaluated by varying the boost pressure and EGR levels, showing that higher 351 EGR rates increases (0% to 39%) can reduce NOx emissions by almost 7 g/kWh with a penalty increase of PM of around 0.3 g/kWh, and providing an optimized condition that 352 balances both pollutants near 0.5 g/kWh of PM and 1.5 g/kWh of NOx, which is lower than 353 354 diesel fuel in both emissions. Specifically, they emphasized that HVO responds well to 355 higher EGRs (25%) and lower boost pressures (130 kPa). Reductions of similar trend were found in [86] for both NOx and PM emissions when increasing the percentage of cook oil 356 357 biodiesel. Additionally, they observed some reducing effect in CO and HC emissions 358 ($\sim 0.05\%$ and ~ 25 ppm respectively). Biofuels, like HVO, free from cycloalkanes and 359 aromatics and with presence of oxygen in their molecule play a significant role in diminishing soot production, making easier the balance with NOx emissions. 360

361 Many research studies also explored another version of LTC, namely PPC. In PPC, a stratified fuel-air mixture is used. The degree of fuel stratification can be controlled by 362 363 varying the SoI timing and other fuel injection parameters such as multiple fuel injections 364 [87]. In PPC, both premixed and diffusion flames are present after auto-ignition in several 365 locations in the combustion chamber [88-89]. PPC can be applied to both gasoline and diesel-fueled engines. Kalghatgi et al. [90-91] applied PPC in light- and heavy-duty 366 engines and achieved superior engine efficiency and relatively lower exhaust emissions 367 368 simultaneously. It was suggested that gasoline with the research octane number (RON) of around 70 is highly recommended for the optimum PPC. Using this fuel, PPC results 369 370 in a trade-off between combustion stability at low loads and HRR at high loads [92-94]. 371 Han et al. [95-96] performed PPC experiments using diesel/gasoline blends. They reported 372 that PPC exhibited relatively higher engine efficiency and lower exhaust emissions than 373 CDC. However, PPC's two major concerns were relatively inferior combustion stability at low loads and the high soot emissions at the high loads. Liu et al. [97] also performed PPC 374

375 investigations to resolve these issues. They used a blend of polyoxymethylene dimethyl 376 ethers (PODEn) and gasoline in a heavy-duty diesel engine for achieving PPC. They 377 reported significantly improved NOx-soot trade-off in the PPC using gasoline/PODEn blends without any fuel efficiency penalty. Due to the addition of PODEn, the combustion 378 379 efficiency and combustion stability also exhibited a significant improvement, especially at low loads. Although preliminary LTC approaches demonstrated significant potential in 380 381 reducing NOx and PM, these combustion strategies have many issues related to engine 382 performance compared to baseline CDC and, more specifically, their applicability in production-grade engines. Several techniques such as EGR [98], VVT [99-100], variable 383 384 compression ratio (VCR) [101], and intake air temperature variations [60] have been 385 investigated extensively to overcome these challenges. These interventions resolved some of the issues related to the engine performance and emission characteristics; however, 386 these techniques could not resolve their applicability at higher engine loads. In most LTC 387 388 studies involving mineral diesel as test fuel, it was seen that the high reactivity of mineral 389 diesel was the main hurdle for extending the extreme load limits (low and high loads) of 390 the LTC concept. In LTC, many researchers advocated using low octane fuels (diesel-like 391 fuels) at lower engine loads. However, high octane fuels (gasoline-like fuels) could be used at high loads to achieve stable combustion, superior engine performance, and lower 392 393 emissions [102]. Lu et al. [103] explored the effect of addition of gasoline like fuels in PCCI 394 combustion mode. They carried out experiments using the blends of butanol and gasoline vis-à-vis pure butanol fuelled PCCI combustion mode. They reported that addition of 395 396 small amount of gasoline in butanol resulted in superior PCCI combustion mode by 397 increasing the maximum pressure and temperature, which led to lower HC and CO emissions from PCCI combustion mode. Following the preliminary research efforts of 398 Bessonete et al. [102], Inagaki et al. [104] demonstrated yet another version of LTC, 399 namely dual-fuel PCI combustion. Two fuels with different reactivity were injected into 400

401 the combustion chamber using two separate injectors in the PCI combustion. The fuel 402 quantities were manipulated independently as per the engine load to control the overall 403 mixture reactivity in the combustion chamber. The results exhibited an excellent control 404 over various combustion parameters, which was not possible in the previous LTC 405 strategies. PCI combustion yielded extremely low NOx and PM emissions and also showed 406 a significant potential for further improvement. Shim et al. [105] also carried out a detailed investigation of dual-fuel PCCI combustion mode using a combination of 407 408 compressed natural gas (CNG) and mineral diesel. They compared the combustion, 409 performance and emission characteristics of dual-fuel PCCI combustion mode with single-410 fuel PCCI combustion mode and reported that combustion controlling was much easier in 411 dual-fuel PCCI combustion mode. It can be done by adjusting the premixed amount of second fuel. They also reported that dual-fuel PCCI combustion mode exhibited superior 412 413 engine performance along with lower CO and HC emissions compared to single-fuel PCCI 414 combustion mode. Hence, many researchers explored this combustion strategy, further 415 developed as RCCI combustion strategy. The next sub-section provides insights into the 416 fundamentals of RCCI combustion mode and its evolution with time.

417

1.4 RCCI Combustion Concept

Previous efforts to develop a commercial LTC engine failed due to certain limitations, 418 419 especially for high-load applications. The high chemical reactivity of mineral diesel is the 420 major concern for diesel-fueled LTC engine development, resulting in inferior control over 421 combustion. Bessonette et al. [102] suggested that the in-cylinder reactivity of the fuel-422 air mixture should be varied to achieve stable LTC at different engine loads. Based on 423 these recommendations, Kokjohn et al. [106] proposed the concept of RCCI, wherein gasoline was the premixed low reactivity fuel (LRF), and diesel was the directly injected 424 425 high reactivity fuel (HRF) for achieving reactivity stratification in the cylinder. In their 426 work, 50% thermal efficiency could be achieved along with near-zero NOx and soot emissions using this configuration. In RCCI combustion mode, an LRF was supplied in
the intake manifold, mixed with the intake air to form a fully premixed intake charge.
The HRF was directly injected to ignite the premixed intake charge; however, the directly
injected fuel did not completely mix and remained stratified before the SoC.

In the RCCI combustion combustion mode, the combustion phasing is determined by the 431 global reactivity of the mixture, given by the LRF to HRF ratio, while the combustion 432 433 timing (duration and phasing) is controlled by the stratification of the ignition delay. 434 Initiation of combustion takes place at HRF-air mixture locations and combustion progress from high-to-low reactivity, which allows a controlled sequential ignition. It 435 436 provides superior combustion control than other LTC strategies. RCCI combustion 437 combustion modeexhibits very low soot and NOx emissions with enhanced engine efficiency than CDC for a wide range of speeds and loads. This combustion strategy 438 439 requires highly premixed fuels, which can be achieved by using two injections of HRF in the combustion chamber. The first injection is used to increase the in-cylinder reactivity. 440 441 The second injection improves the mixing and acts as an ignition source, resulting in lower 442 soot formation [107]. By altering the global charge reactivity in combination with one or 443 more direct injections, a complete control over the combustion phasing and HRR can be accomplished [106, 108-113]. This allows RCCI combustion engine operation with 444 445 combustion efficiencies over 97% across a wide load range (from 4 bar to 20 bar BMEP) 446 [114].

Additionally, gross indicated thermal efficiencies (ITEg) approaching 60% have been demonstrated experimentally. The dual-fuel strategy allows easier control of the combustion phasing, which is regulated by the local concentration of the HRF and the injection timing of the HRF. The combustion duration is controlled by the mixture reactivity gradient, which can be tailored to reduce pressure rise rates and reduce the

452 combustion noise. With proper feedback, cycle-to-cycle control would be relatively easier453 to implement in production-grade engines.

454 RCCI engine operation has been expanded by the direct dual-fuel stratification (DDFS) 455 combustion strategy, which utilizes two direct injectors, each dedicated to injecting either 456 the LRF or the HRF. These are centrally mounted in the combustion chamber to achieve 457 clean and efficient RCCI combustion mode in a heavy-duty engine [115-116]. RCCI 458 combustion mode relies heavily on the fuel reactivity stratification gradient to achieve 459 clean and efficient combustion, and a broad range of fuels may be used. RCCI is essentially 460 an inherently fuel-flexible advanced combustion strategy. Research shows that RCCI 461 combustion mode offers relatively flexible control over combustion since fuel concentration 462 and reactivity stratifications regulate heat release [117]. Due to wide availability, gasoline and diesel have been used as the LRF and HRF, respectively, in most previous RCCI 463 research [108, 113, 108-121]. In many studies, the primary reference fuels (PRF) as iso-464 octane and n-heptane have also been studied as the LRF and HRF, respectively [104, 122-465 466 125]. Researchers also explored the potential of alcohol blends as LRF [120, 122, 126-130] 467 and biodiesel as HRF for RCCI combustion mode [131-132]. However, mobile engine 468 applications may preclude the ability to carry large quantities of two separate fuels. As a result, cetane improvers, such as di-tert-butyl peroxide (DTBP) and 2-ethylhexyl nitrate 469 470 (EHN), have been studied as low quantity additives to condition the LRF to perform as an HRF to approximate single-fuel RCCI operation [120, 127, 130-131, 133]. Splitter et 471 al. [133] carried out RCCI investigations using two fuels having large reactivity 472 473 differences and optimized the in-cylinder fuel stratification. They observed a 474 simultaneous reduction in NOx and PM emissions and achieved ~60% indicated thermal efficiency (ITE). Liu et al. [134] explored the ignition and flame development in RCCI 475 476 combustion mode. They used several fuel supply strategies to achieve different fuel 477 stratifications. They reported that the auto-ignition and flame front propagation could be

478 controlled by regulating the degree of fuel stratification. Kokjohn et al. [135-136] carried 479 out detailed investigations of RCCI combustion mode using optical diagnostic and 480 simulation. They reported that the ignition and combustion events are dominantly 481 controlled by fuel-air mixture reactivity stratification. However, the role of mixture concentration stratification and thermal stratification was less influential. This is why 482 the reactivity and concentration stratification focus on RCCI combustion mode to extend 483 the engine load range. The mixture reactivity gradient can be enhanced by enlarging the 484 485 reactivity difference between the premixed and DI fuels [137-140]. Many studies have 486 reported that RCCI combustion mode exhibits higher thermal efficiency due to shorter 487 combustion duration, increased specific heat ratio, and decreased heat transfer losses 488 than conventional CI combustion [19, 141]. RCCI combustion combustion modehas higher 489 thermal efficiency than CDC, mainly attributing to its shortened combustion duration, 490 increased specific heat ratio, and decreased heat transfer losses [19, 141].

491 Olmeda et al. [142] suggested that the primary reason for the relatively higher efficiency 492 of all LTC modes is common in which mixture homogeneity plays an important role 493 because this leads to rapid combustion events. Under optimized combustion phasing, this 494 results in higher fuel-to-work efficiency. The presence of uniform charge inside the combustion chamber also results in similar global and local temperature distribution. 495 496 This is useful in reducing the localized effect of temperature distribution in heat transfer 497 from cylinder walls. Advanced combustion strategies have arisen as pathways for 498 achieving high engine efficiency and relatively cleaner combustion than conventional 499 combustion engines in the last few decades. In most advanced combustion techniques, the 500 thermodynamic cycle efficiency can be increased by idealizing the heat release during 501 combustion towards constant-volume energy conversion. In addition to the constant-502 volume approach, LTC strategies also result in relatively lower heat loss from the cylinder 503 walls due to significantly lower peak of in-cylinder temperatures. This feature of LTC is also reflected in NOx emissions. By improving mixture formation and spray performance,
reductions in peak equivalence ratios during combustion reduce PM formation. Numerous
strategies have been developed to achieve clean and efficient combustion, as summarized
in Figure 4 [143].



508

Figure 4: Comparison of various conventional and advanced combustion strategies with
respect to fuel stratification [143].

511 Despite the great potential of RCCI combustion mode to operate at high engine loads, it 512 required a high degree of fuel stratification to avoid excessive HRR. In the absence of fuel 513 stratification at higher engine loads, the fraction of diffusion phase combustion becomes 514 dominant in RCCI combustion mode, leading to higher soot emissions [125].

515 2. Evolution of RCCI Combustion Concept

516 RCCI combustion mode can be considered special dual-fuel combustion in which an 517 appropriate fuel pair can be used under optimized operating conditions. The dual-fuel 518 combustion strategy is not new. It was introduced in the 1970s as the ATAC. The basic 519 difference between two dual-fuel combustion modes is the methodology of fuel 520 introduction. In ATAC, the LRF is directly injected into the combustion chamber, and a 521 two-stage ignition assists combustion. In the ATAC concept, a hot (thermal) atmosphere 522 with (active) combustion products and radicals is created by igniting a small amount of premixed HRF, which dominantly affects the combustion of LRF. In comparison, in the 523 524 RCCI combustion mode, an LRF is supplied through the intake port, forming a premixed charge. This premixed fuel-air mixture is then ignited by using the directly injected HRF 525 526 [144-145]. The HRF is delivered through the intake port to form a premixed charge [146-149]. 527

528 In ATAC, the combustion parameters at varying engine loads can be controlled effectively 529 by the fuel injection parameters such as fuel injection timing of the LRF. However, the 530 RCCI combustion mode is more popular due to its engine performance improvement and emission reduction potential. Hence, RCCI combustion mode is extensively explored 531 experimentally and numerically [150-153]. Recent studies focused on extending the 532 engine load window from low to high load and then to full load [154-156] in RCCI mode. 533 534 Cooper-Bessamer applied the dual-fuel concept, and they demonstrated a gas-diesel dualfuel engine in 1927 for the first time [157]. However, a successful dual-fuel concept was 535 536 first demonstrated commercially in 1940 by the National Gas & Oil Engine Company Limited, England [158]. Their dual-fuel engine model used a dual-fuel combustion 537 538 strategy in which a direct-injected diesel ignited a premixed natural gas-air mixture. In this concept, the pilot injection of mineral diesel acted as a "spark plug," allowing 539 540 sufficient ignition energy at multiple ignition sites in the premixed charge. This resulted 541 in flame propagation, which combined the ignition mechanisms of both conventional SI 542 and CI engines.

543 Most dual-fuel engines use gasoline as the premixed fuel [159], and, in most of the 544 industry-standard dual-fuel modern engine combustion strategies, a diesel pilot ignition 545 (DPI) is used with gaseous fuels [160-162]. In turn, the basic concept of RCCI combustion

mode has become popular among researchers because of its potential for achieving 546 547 superior engine performance and significantly lower emissions, especially NOx and PM. 548 Previous studies have shown that RCCI combustion combustion modecan effectively utilize many renewable fuels, such as biofuel [163-167], PODEn [168-169], and methanol 549 550 [170-171]. Olmeda et al. [142] performed RCCI combustion mode experiments using 551 different LRF (gasoline and E85) and compared the heat transfer characteristics with 552 baseline CI combustion engine (Figure 5). These experiments were performed in a singlecylinder light-duty research engine equipped with several thermocouples in the cylinder 553 554 head and liner (Figure 6). They reported that the heat transfer characteristics of both 555 LRFs were almost similar; however, both test fuels exhibited relatively lower heat 556 transfer compared to baseline CI combustion. The exhaust losses were slightly higher for 557 the E85 as a result of the longer combustion duration. This effect was balanced by higher combustion efficiency, proving that both LRFs can be applied in RCCI combustion mode 558 559 and would deliver similar energy use and efficiency results. The authors also compared 560 the energy analysis of gasoline-diesel fueled RCCI mode and conventional CI combustion mode engines for energy balance. The RCCI combustion modeengine exhibited superior 561 562 energy capacity from the combustion. Relatively shorter combustion duration was responsible for this trend, which reduced 13% heat loss than conventional CI combustion 563 engine. Relatively lower enthalpy of exhaust gases from the RCCI combustion combustion 564 565 modewas another important factor, resulting in lower exhaust losses. These lower heat 566 losses are also positively affected combustion efficiency.



Figure 5: In-cylinder temperature, and apparent heat release (left), and energy
distribution (right) for RCCI mode gasoline engine vis-à-vis CDC engine [142].



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573

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Figure 6: Measured cylinder head temperature surfaces at 7 mm (upper graphs) and 4 mm (middle graphs) from the firedeck and liner (lower graphs) temperatures for RCCI mode gasoline engine (left) and RCCI mode E85 engine (right) [126].

575 Jia and Denbratt [172] investigated combustion characteristics of mineral diesel-576 methanol fueled RCCI combustion mode at higher engine loads. They reported ultra-low 577 soot and NOx emissions. Singh et al. [173] performed comparative investigations of RCCI 578 and PCCI combustion modes with baseline CI combustion mode using critical parametric analyses. They reported that RCCI combustion mode resulted in comparable performance
to CI combustion mode at lower loads. However, the RCCI combustion combustion
modeexhibited superior engine performance at higher engine loads and significantly lower
NOx and PM emissions than PCCI and baseline CI combustion combustion mode(Figure
7).





Figure 7: Comparison of baseline CI, PCCI, and RCCI combustion modes at (a) low
engine load (1 bar BMEP) and (b) high engine load (4 bar BMEP) [173].

In a similar investigation by Han et al. [174], different LTC modes, namely PCCI, HCCI, and RCCI, were compared to baseline CI combustion mode. They used n-butanol and mineral diesel as test fuels in different combustion modes. They reported that all LTC modes emitted significantly lower NOx and soot than baseline CI combustion combustion mode. RCCI combustion combustion modeexhibited relatively higher efficiency and superior combustion control compared to other LTC modes.

593 The potential of using single-fuel in RCCI combustion mode to reduce the system 594 complexity was explored. In this strategy, an LRF can also be used as HRF with a small 595 amount of cetane improver such as DTBP, 2-EHN, etc. [122]. Splitter et al. [133] used 596 gasoline as LRF and gasoline with DTPB as HRF. They compared single-fuel RCCI 597 combustion mode results with dual-fuel (gasoline/ mineral diesel) RCCI combustion mode. 598 They reported slightly improved engine performance of the single-fuel RCCI combustion 599 mode. This might be due to the higher reactivity gradient generated by DTPB, which led 600 to relatively lower low-temperature heat release (LTHR). A lower LTHR results in lower 601 compression work, eventually leading to higher engine efficiency. In another study by 602 Hanson et al. [175], single-fuel gasoline-gasoline + 3.5% 2-EHN fueled RCCI combustion mode was compared with gasoline-diesel-fueled RCCI combustion mode. They reported 603 604 that adding a cetane improver resulted in relatively faster high-temperature heat release 605 (HTHR) of single-fuel RCCI combustion mode, leading to relatively shorter combustion 606 duration than dual-fuel RCCI combustion mode. Mohammadian et al. [176] explored this strategy using isobutanol as LRF and isobutanol+20% DTPB as HRF and reported similar 607 608 RCCI combustion mode characteristics to dual-fuel RCCI combustion mode. Wang et al. [161] also used isobutanol/ isobutanol doped with DTPB fuel-pair in a heavy-duty RCCI 609 610 combustion mode engine. They reported that isobutanol required more DTPB for generating a reactivity gradient. Although the combustion characteristics of both alcohol 611 612 and gasoline were similar in RCCI combustion mode, alcohols required higher doping with 613 cetane improver due to their lower cetane number of alcohols, especially butanol [122, 614 178]. A reverse single-fuel strategy was also explored in few studies, in which HRF was used as the LRF [179-180]. 615

Although PPC and RCCI combustion modes have significantly improved combustion, 616 617 performance, and emission characteristics than baseline CDC and other LTC modes, 618 RCCI combustion mode has several technical challenges. Results of previous studies 619 showed that the RON of the fuel should be changed, depending on the engine load. This 620 is because the RON indicates how much compression the fuel can withstand before autoigniting; the higher the RON the more pressure needed for ignition. For effective RCCI 621 622 combustion mode at low loads, RON should be reduced to enhance combustion stability. 623 Similarly, a higher RON should be used to accelerate the combustion at medium and high

loads. In RCCI combustion mode, a fraction of premixed LRF cannot be completely 624 625 oxidized due to the dominant charge cooling effect near the cylinder walls and crevice 626 regions. This results in relatively higher HC and CO emissions, promoting incomplete combustion. A new combustion strategy, 'reverse RCCI' (R-RCCI) combustion, is gaining 627 significant attention in recent years. In the R-RCCI combustion mode, reverse reactivity 628 629 stratification is used to achieve superior performance and emissions than conventional RCCI combustion mode. In R-RCCI, a small amount of HRF is premixed in the intake 630 manifold to ignite the LRF, which is directly injected into the cylinder during the 631 compression stroke (Figure 8). 632



633

Figure 8: PPC, RCCI, and R-RCCI modes of combustion at low-to-medium loads [87].

In R-RCCI combustion mode, a relatively smaller quantity of HRF than conventional
RCCI is premixed, resulting in lesser fuel trapped in the squish and crevice regions.
Higher oxidization tendency of the trapped premixed HRF is another important aspect of
the R-RCCI combustion mode, enhancing the combustion efficiency. A lower combustion
rate of R-RCCI compared to PPC makes it more suitable due to more reactivity

640 stratification. Many researchers have carried out detailed experimental and simulation 641 studies and reported that the R-RCCI combustion mode results in relatively lower heat 642 transfer, leading to higher thermal efficiency. Previous studies showed that R-RCCI balanced the crucial parameters required for soot oxidation, namely the degree of 643 premixed combustion and combustion temperature. This led to very low soot emissions at 644 645 all engine loads. Huiquanet al. [87] conducted an experimental investigation in RCCI, PPC, and R-RCCI combustion modes. They reported that R-RCCI combustion mode 646 resulted in lower NOx and soot emissions than PPC (Figure 9). The authors focused on 647 the RCCI combustion mode exhaust losses caused by its lower combustion rate, which 648 649 resulted in lower ITE. They suggested that the R-RCCI configuration effectively resolved 650 this issue by using PODE injection in the port and gasoline injection directly in the 651 combustion chamber (Figure 10).



652

Figure 9: Fuel induction strategies corresponding to the electrical signal to the injectors
for PPC, RCCI, and R-RCCI modes of combustion (DI: direct injection; PFI: port fuel
injection; PR: premixed ratio) [87].

Liu et al. [131] used PRFs (n-heptane and iso-octane) as test fuels in the R-RCCI combustion mode to investigate the effects of fuel concentration and reactivity stratifications. They reported a significant increase in the thermal efficiency of the engine and the combustion efficiency. Ji et al. [181] and Lu et al. [146] also used the same test 660 fuels (n-heptane as the premixed HRF and iso-octane as DI fuel) in their investigations.
661 They focused on the fuel injection timing of the LRF and reported a weak effect of DI
662 timing of the LRF on the R-RCCI combustion mode. The SoI sweep exhibited the best
663 ignition characteristics at the DI timing of - 25°CA aTDC, leading to the highest thermal
664 efficiency.



665

Figure 10: Comparison of NOx and soot emissions among PPC, RCCI, and R-RCCI
modes of combustion [87].

Yao et al. [149] and Chen et al. [182] investigated the RCCI combustion mode using the reverse fuel reactivity combination. They used a modified single-cylinder diesel engine with premixed DME as HRF and injected methanol as LRF directly into the combustion chamber. They utilized the basic concept of ATAC, in which the heat released by premixed DME was enhanced by the in-cylinder thermal atmosphere. They reported that this 673 method extends the load limit due to higher reactivity stratification achieved by direct 674 methanol injection. They found that early direct injection of fuel results in homogeneous 675 combustion, similar to HCCI; however, the late direct injection can be used for the threestage combustion. Lu et al. [146, 183-184] investigated a similar type of combustion using 676 677 different LRFs, namely iso-octane, n-butanol, and ethanol, along with premixed n-678 heptane as HRF. Based on the heat release pattern, they categorized the combustion of 679 two fuels having different reactivities in three ways:(i) a two-stage HCCI-like heat release 680 process dominated by the thermal atmosphere, (ii) a three-stage heat release process 681 dominated by the active atmosphere combustion, and (iii) a heat release process lying in 682 between the above two categories dominated by both the active and thermal atmospheres 683 [146]. Cui et al. [185] performed an optical investigation of the R-RCCI combustion mode 684 to understand the combustion better. They reported that the premixed ratio was the most 685 critical parameter in controlling the combustion phasing.

Lu et al. [183] also used premixed n-heptane and directly injected methanol in the R-RCCI combustion mode and reported significantly lower NOx and soot emissions. They suggested that combining optimum combustion characteristics and fuel properties promoted huge reductions in soot and NOx emissions.





Figure 11: Experimental and predicted in-cylinder pressure and HRR profiles at three
SoI timings [186]

693 Tang et al. [186] also investigated the R-RCCI combustion mode using a combination of 694 n-heptane and iso-octane as test fuels. They analyzed the effect of injection timings of isooctane on the combustion characteristics of dual-fuel combustion. They divided the 695 696 combustion into three regimes, namely LTHR, homogeneous combustion heat release (HCHR), and premixed combustion heat release (PCHR), where HCHR and PCHR were 697 the two sub-parts of the HTHR (Figure 11). They observed that the SoI timings of iso-698 octane had little effect on the LTHR; however, retarding the SoI timing of iso-octane 699 700 resulted in relatively dominant HTHR. This was mainly attributed to HCHR and PCHR 701 as the two HTHR regimes, in which HCHR became more dominant at retarded SoI 702 timings. Recently, a new version of RCCI combustion mode namely "Intelligent Charge 703 Compression Ignition" (ICCI) has been introduced in which both LRF and HRF can be directly injected to achieve a better control [187-188]. Zhao et al. [188] carried out a 704 705 detailed investigation using both ICCI and RCCI modes of combustion and compared their 706 performance and emission characteristics with conventional combustion using blends of HRF and LRF. They reported that blending technique resulted in superior combustion and performance characteristics at lower engine loads, however, at higher engines, RCCI and ICCI modes of combustion exhibited superior combustion control, leading to higher engine efficiency. Among both LTC modes, RCCI combustion mode was found superior especially up to medium engine loads, however, ICCI mode of combustion showed more potential to be adopted at higher engine loads. This was mainly due to precise control on injection parameters of both LRF as well as HRF.

This section summarizes the evolution of RCCI combustion mode as an LTC strategy, 714 715 wherein an LRF is introduced into the engine through the intake port, forming a premixed 716 charge, which is ignited by the use of a directly injected HRF. The RCCI combustion mode 717 performance using numerous test fuels, including alternative fuels, is analyzed. Some of these alternative fuels aided in reducing soot and NOx emissions. RCCI combustion mode 718 719 was compared with other combustion concepts, including the ATAC. The injection 720 mechanisms/ strategies were different among these combustion modes, e.g., the ATAC 721 inducted the LRF through direct injection and the HRF through the intake port. This 722 section also underscores some of the issues faced by RCCI, including relatively higher HC 723 and CO emissions. In the RCCI combustion mode evolution, reverse RCCI was explored as a new strategy consisting of reverse reactivity stratification, i.e., injecting a smaller 724 725 quantity of HRF into the intake manifold to ignite the LRF, directly injected into the cylinder. A smaller quantity of HRF injected in the R-RCCI mode leads to lesser fuel 726 727 entrapment in the crevices. Finally, some fundamental studies on the effect of boundary 728 conditions of R-RCCI are listed to explore how the combustion is affected.

729 2.1 Fuel Kinetics of LTC

Most of the ICEs are operated by gasoline- and diesel-like fuels, which contain primarily
straight chain alkanes, branched alkanes and aromatics. Previous research studies using
primary reference fuels (PRFs) exhibited that the combustion kinetics of these fuels and

733 their auto-ignition characteristics are significantly affected by the composition, molecular 734 size and structure of fuel. The combustion of gasoline and diesel in traditional combustion 735 modes are mainly affected by transport-controlled high-temperature premixed flames and non-premixed flames, respectively [189]. However, in LTC modes, autoignition and cool 736 737 flames play an important role in overall combustion characteristics due to higher degree 738 of fuel-air premixing. Ju et al. [189] reviewed a number of studies carried out for detailed 739 investigation of cool flames in LTC and reported that cool flames are affected by a number 740 of parameters such as EGR, fuel structure, etc. in case of LTC modes. In all LTC modes, 741 EGR is used to reduce the flame temperature because EGR has a dominant role in 742 suppressing hot flames. This results in dominant cool flames in LTC modes because in 743 LTC modes, a transition between cool flames and hot flames during the combustion affects the engine performance and emission characteristics [190]. Agarwal et al. [191] reviewed 744 745 the fuel combustion chemistry and reported that PRF (n-heptane and iso-octane) behave 746 differently during auto-ignition in LTC engines. N-heptane is reactive straight-chain 747 paraffin and has a low octane number, which means that it has a relatively lower 748 resistance to auto-ignition. In comparison to n-heptane, iso-octane is a less reactive 749 branched-chain paraffin, and has a higher octane number [192]. They explained the combustion characteristics of LTC and reported a two stage combustion in which the first 750 751 stage is related to low temperature heat release (LTHR) and the second or main 752 combustion is associated with the high temperature heat release (HTHR) (Figure 12). The 753 first stage ignition is also regarded as 'cool flame', with negative temperature coefficient 754 (NTC) [193].




Figure 12: Typical HRR for two-stage combustion in LTC [193]

757 The analysis of chemical reactions during the combustion of hydrocarbons in IC engines 758 shows three main routes of reactions, which mainly depends on temperatures. Below 750 759 K, reactions are dominated by chain-propagating steps including oxygen molecules and 760 generation of partially oxidized species [194]. Between 800 K and 950 K, the chain 761 propagating steps involving O molecules yield conjugate alkenes and HO_2 radicals [194]. 762 Above 1000 K, the main fuel radical reactions are thermal decomposition by C-C bond 763 breakage, forming alkenes and smaller radicals [195]. In previous studies, two different 764 regions of chain reactions have been defined as, (i) a low-temperature kinetics region or ignition process, below 950 K, and (ii) the high-temperature kinetics region, where the 765 bulk of chemical energy is released [193]. However, the introduction of different 766 767 alternative fuels including alcohols, ethers, biodiesel, etc. have shown slightly different LTC kinetics due to their different molecular sizes, structures, bond energies and 768 769 functional groups [196-200].

The cool flame in combustion has been investigated extensively, however, after the introduction of LTC modes, this become more relevant due to a relative dominance of low temperature reactions (LTR) and high temperature reactions (HTR) during LTC. Pease [201-202] reported that in LTC, radical formation is sensitively affected by the

774 temperature, which decreases with increasing temperature due to relatively faster 775 dissociation of chain-branching intermediates. In this context, fuel reactively also plays 776 an important role because, at low-temperatures, the rate of chain branching for straight-777 chain paraffin's (like n-heptane) are much more intense compared to as that of branched-778 chain paraffin (like iso-octane). This is due to the structure of n-heptane radicals, which 779 lead to higher rates of alkylperoxy radical (RO₂) isomerization from keto-hydro-peroxide 780 decomposition [192]. In a very popular research carried out by Barusch et al. [203], it has 781 been reported that cool flame chemistry of LTC is significantly affected by RO₂ and 782 hydroperoxyl alkyl radical (QOOH). On the other hand, large number of less reactive 783 methyl groups and presence of tertiary and quaternary C atoms in its structure promotes 784 the lower reactivity of branched-chain paraffins (iso-octane). For compact and highly 785 branched fuels, ignition is inhibited due to presence of a large fraction of strongly bonded 786 H atoms [204]. The combustion of iso-octane is controlled through the OH pool, which is 787 mainly derived from the n-heptane low temperature pathway.

788 The fundamentals of cool flames and its application in combustion models have been 789 already reviewed in many previous studies [205-207]. In most of these studies, it has been 790 reported that the temperature and the rate of radical production affect the oxidation of 791 fuels with low-temperature reactivity [205-206]. Ignition of hydrocarbons is also affected 792 by the chain branching processes, which occur in two stages. In first stage, partial 793 oxidation of fuel takes place, resulting in intermediate species like C₂H₄, CH₂O, and 794 $CH_{3}CHO$. First stage of ignition process is mainly controlled by low-temperature peroxy 795 chemistry [196,206, 208] and it does not contribute much in temperature rise. The first 796 stage ignition processes further progress by oxidation of intermediate species, which 797 raises the temperature up to a critical limit, where second stage ignition starts. In this 798 stage, complete oxidation of fuel results in formation of CO₂ and H₂O along with a 799 significant temperature rise. This is the major reason why the second stage of ignition is

800 also known as high temperature ignition (HTI). In conventional combustion modes, 801 presence of significantly higher initial temperatures results in only HTI, however, in LTC 802 modes, both LTI as well as HTI contribute and their relative dominance depends on 803 temperature and pressure conditions. In most of the studies related to cool flame behavior, 804 depending upon the initial temperature, chain branching chemistry has been divided into 805 three categories as low-temperature, intermediate-temperature, and high-temperature 806 routes. At the lower temperatures (up to 750K), OH, O or HO_2 radical starts the fuel 807 oxidation by H-abstraction from the fuel molecule (RH) and generates fuel radicals (R). This fuel radical reacts with O₂ and forms a RO₂, which undergoes internal isomerization 808 809 and results in QOOH. Decomposition of QOOH leads to formation of many OH radicals, 810 which depends on low-temperature pathway. Due to formation of these large number of radicals, chain branching becomes quite rapid, which adds more energy and OH radicals 811 812 to the system [209]. The low-temperature reactions are exothermic, which further increase OH radical formation due to increasing temperature of the system [192]. 813

814 $R \rightarrow RO_2 \rightarrow QOOH \rightarrow O_2QOOH \rightarrow OQ'O + 2OH$

815 With increasing temperature, chain-propagating pathway suppresses the chain-816 branching pathway in which peroxy hydroperoxyl alkyl radical (O₂QOOH) is replaced 817 with cyclic ether through following reaction.

818

$$R \rightarrow RO_2 \rightarrow QOOH \rightarrow OH + cyclic ether$$

However, NTC effect also affects the above reactions due to reverse reactions, which significantly reduces the formation of radicals. Pilling et al. [210] reported another feature of low-temperature reactions that the chain-branching reactions do not act as actual chain-branching because of the formation of either one or no radials at low-temperatures. They suggested that this behavior of chain branching at low-temperatures is common for all fuels. However, few oxygenated fuels including diethyl ether, dibutyl ether, etc. exhibit differently with two NTC regions. The dominance between second O₂ addition and QOOH 826 decomposition is the main reason for the first NTC; however, competition between first 827 O₂ addition and direct beta scission for the fuel radicals is the main cause of second NTC 828 [211]. The tendency of dual NTC effect is mainly controlled by C-O bond energy. Rodriguez et al. [212] explored the low-temperature behavior of dimethyl ether (DME) and reported 829 that NTC effect for DME starts at ~550K, however, it become significantly noticeable in 830 831 the temperature range of 600 to 750 K. In this temperature region, fuel oxidation becomes slow due to dominant contribution of O₂QOOH, QOOH, and RO₂ decomposition reactions. 832 At intermediate-temperatures (900 to 1100K), high-temperature chemical reactions 833 dominate the low-temperature reactions resulting in chain branching, where fuel 834 835 oxidation is thought to be governed by a branching process involving HO_2 [193]. 836 $RH+HO_2 \rightarrow H_2O_2+R$ $H_2O_2+M\rightarrow OH+OH+M$ 837 838 Here, M is any non-reactive third-body. For initiating these reactions, HO_2 is mainly 839 formed from following reactions. $RO_2 \rightarrow alkene + HO_2$ 840 $QOOH \rightarrow cyclic$ ether + $OH \rightarrow alkene + HO_2$ 841 842 The reaction progresses with formation of OH radicals by decomposition of RO_2 and QOOH in addition to reaction between aldehyde radical and O₂. Most of the time, the 843 formation of these lower aldehydes is the possible reason of faint blue luminescence of cool 844 845 flames [201-202]. At intermediate-temperatures, chain-branching reaction produces multiple OH radicals; however, these reactions are highly sensitive to heat loss and 846 oxygen concentration. 847 $CH_2O \rightarrow HCO \rightarrow HO_2 \rightarrow H_2O_2 \rightarrow 2OH$ 848

In some cases (at relatively higher temperatures), this intermediate chain-branching reactions are surpassed by a multi-stage warm flame, where both cool flame and hot flame can exist [213]. Mostly the LTC lies in the H₂O₂ concentration history, which is produced 852 ~1000K by low and intermediate temperature reactions. Li et al. [214] suggested for a 853 prominent pathway of H_2O_2 formation, as given below.

 $HO_2 + HO_2 \rightarrow H_2O_2 + O_2$

 H_2O_2 concentration increases steadily, with H_2O_2 decomposition much slower than its 855 production. H_2O_2 decomposes rapidly at temperatures around 1000K, yielding large 856 857 numbers of OH radicals. Increase in OH radicals consumes any remaining fuel, resulting 858 in ignition. The important requirement for cool flames to induce in-cylinder combustion is the energy requirement so that low temperature reactions can increase the chamber 859 temperature to the high temperature region. When the reactions become slower during 860 861 the NTC period, high cylinder temperature can be increased sufficiently by external 862 factors such as external heating or increasing CR, which forces the engine operation towards high temperature regime by increasing compression work during compression 863 864 stroke.

At extremely high-temperatures (above 1100 K), a different chain-branching reactionoccurs as shown below.

867

$$H + O_2 \rightarrow OH + O$$

868 This reaction dominates the overall reactions leading to main ignition, which is identical for all test fuels. This is the unique chemical kinetics of all test fuels at high-temperatures, 869 870 where chain branching reaction become fuel independent. Since molecular oxygen participates in this reaction, lean fuel mixtures are more reactive in this high-871 temperature regime, whereas rich fuel-air mixtures are oxidized quickly at low-872 873 temperatures due to chain branching, which depends on radical species formed directly 874 from the parent fuel [215]. The consumption and production pathways of O_2 and OH(radical pool), revealing that the major O_2 consumption reactions related to the LTC 875 876 chemistries of hydrocarbons contribute to the heat release directly. Heat release reactions are related to O_2 , implying the significance of the fuel mixing process for the LTC 877

878 characteristics. These radical-branching reactions are very important for the high-879 temperature regime because the products contain more radicals than the reactants. 880 Similar to previous experiments, Mancaruso and Vaglieco [216] performed UV-Visible imaging and spectroscopic measurements and they detected the chemical species involved 881 882 in LTC. They injected bio-ethanol in the intake manifold and mineral diesel directly in the cylinder similar to RCCI combustion mode. They reported that the SoC of 883 homogeneous fuel-air mixture in LTC was mainly controlled by OH radicals, which were 884 885 generated during the intermediate temperature chain branching pathways of LTC. The 886 concentration of OH radicals was found to be directly controlled by the temperature and 887 type of reactions as low, intermediate or high temperature. Liu et al. [217] explored the 888 fuel kinetics of RCCI combustion mode and reported that CH₂O and HCO (formyl radical) 889 are two most effective species, which play an important role in conducting the reaction in 890 the HTHR stage. Even in diesel engines, it has been revealed by the stabilization mechanism and overall ignition process that LTHR pattern is similar to LTC engines, 891 892 which progresses towards a dominant HTHR. Krisman et al. [218] experimentally showed 893 this behavior, where thermal and molecular diffusion results in a gradual transition from LTHR to flame stabilized autoignition (HTHR). 894



Figure 13: Evolution of characteristic events and associated time scales for ignition in high-pressure spray flames [219]

Dahms et al. [219] related these combustion regimes and suggested that LTC reactions and cool flames cannot be neglected in case of diesel engines also because the ignition process in diesel engines is also associated with these reactions (Figure 13). Overall, this sub-section shows the general fuel combustion kinetics followed in LTC and importance of cool flames and NTC in different LTC modes. More details about these reaction mechanisms are discussed in next sub-sections, where effects of different features of LTC kinetics on combustion, performance, and emissions are explained.

905 2.2 Emissions from RCCI Combustion mode

Researchers have explored several techniques to resolve the issue of higher emissions of
CO and HC. This section summarized the studies wherein the performance and emission
characteristics of the RCCI combustion mode engine are the primary focus.

909 Wai et al. [221] investigated a diesel-methanol dual-fuel (DMDF) combustion strategy to 910 control the engine-out emissions. They conducted experiments using a pilot injection to 911 achieve stable combustion at a high methanol substitution ratio (MSR). They examined 912 the effects of pilot injection parameters, such as the start of pilot injection timing, pilot 913 injection quantity, etc. They reported that these parameters could effectively control the 914 DMDF combustion, resulting in superior fuel economy, especially at higher MSR. Zhang et al. [222] performed a similar experiment on a mineral diesel-methanol dual-fuel engine 915 916 and reported that methanol fumigation exhibited a significant reduction of NOx and PM 917 emissions; however, the charge cooling effect of methanol enhanced incomplete 918 combustion, leading to relatively higher HC, CO and formaldehyde emissions.

Although RCCI combustion mode emits significantly lower PM than other conventional
combustion modes, the PM emitted by RCCI combustion mode cannot be neglected.
Therefore, PM emissions from RCCI combustion mode engines were routinely

922 investigated. Splitter et al. [223] reported that RCCI combustion mode engines emitted 923 approximately two orders of magnitude lower PM mass emissions than conventional CI 924 engines. Singh et al. [171] performed RCCI combustion mode investigations intending to explore the permissible limits of the premixed ratio of methanol at different engine loads. 925 926 They reported that the increasing premixed ratio of methanol first improved the engine 927 performance; however, it exhibited relatively inferior performance at higher premixed 928 ratios of methanol (>75%). To investigate the RCCI combustion mode and emission 929 characteristics further at extreme limits (combining engine load and premixed ratios), 930 they measured the particle number (PN) concentrations emitted by the RCCI mode engine 931 vis-à-vis baseline CI mode engine (Figure 14). They reported that the number of particles 932 emitted by RCCI combustion mode was significantly lower than baseline CI combustion 933 mode. RCCI combustion mode emitted more nucleation mode particles (NMP) size range 934 [171]. The relative dominance of particles in various size ranges, namely nanoparticles (NP), NMP, and accumulation mode particles (AMP) in the two combustion modes, 935 936 showed an interesting trend. Figure 14 shows that higher engine loads exhibited higher 937 AMP concentrations in both combustion modes, and NMP concentration decreased when 938 increasing engine load. Variations in the number of NP followed a mixed trend at different engine loads, and maximum NP concentration was found at the maximum engine load 939 (BMEP of 4 bar). Higher peak in-cylinder temperature at maximum engine load might be 940 941 a possible reason for the emission of a higher number of NPs. A significant fraction of NPs was generated due to the pyrolysis of lubricating oil. 942



Figure 14: (a) Number-size distributions of particulates, (b) Correlation between the
total particulate mass and NOx emitted in RCCI combustion mode at different engine
loads and premixed ratios [171]

In another study [224], the results showed a very low soot emission (<0.01 g/kW-hr) from a light-duty engine operating in RCCI mode. Prikhodko et al. [225] compared the effectiveness of the DOC on different advanced combustion techniques. They concluded that DOCs could reduce the emission of small-sized particles effectively. They collected the PM samples (on filter paper) from the CI, PCCI, and RCCI combustion mode engines operated at identical speed and load conditions. The filter papers exhibited the PM

953 sampling with the lightest color from the RCCI mode engine; however, the PM mass
954 collected on the filter paper was higher than that collected on the PCCI engine filter
955 (Figure 15).







Figure 15: (a) Comparison of PM samples collected on the filter papers from conventional CI, diesel PCCI, and dual-fuel RCCI combustion modes [225], and (b)

961

Gravimetric analysis of PM mass collected on the filter papers from CI, PCCI, and RCCI

modes [16]

In a similar study by Agarwal et al. [16], a comparison of the physical appearance of the particulate-laden filter papers and PM mass emitted by PCCI, RCCI, and CI combustion modes was done. They reported that the RCCI combustion mode resulted in a slightly higher PM mass on the filter paper, in which a significant fraction of the PM included volatile substances. The physical appearance of these filters also exhibited the presence of black carbon (BC) to be the maximum in the CI combustion mode, followed by the PCCI combustion mode.

970 Jiang et al. [226] performed RCCI experiments to investigate the particulate emissions 971 by gravimetric measurements and reported slightly lower total particle mass (TPM) than 972 the baseline CI combustion. They reported that PM mass emitted by the RCCI combustion mode engine was significantly lower than the baseline CI combustion mode engine. At 973 higher engine loads, TPM emitted by RCCI combustion mode engine reduced 974 975 substantially (by ~57%) due to higher premixed ratio of LRF because of improved fuel-air 976 mixing. Due to emissions of condensable HC species, PM emitted by RCCI combustion 977 mode engine contained higher soluble organic fraction (SOF) than baseline CI combustion mode. The relatively lower in-cylinder temperature was the main reason for the higher 978 979 SOF of the PM emitted by the RCCI combustion mode engine, which remained the same even at higher engine loads; however, SOF in the PM emitted by the baseline CI 980 981 combustion mode reduced significantly at higher engine loads.

Morphological analysis of the particulates using TEM was done by Storey et al. [227], which revealed the presence of condensed hydrocarbon droplets in the PM samples from the RCCI combustion mode engine. This analysis also indicated that the PM from the RCCI combustion mode engine had lower carbonaceous material than the soot emitted by the baseline CI combustion mode engine. The absence of graphitic structure in the PM

987 emitted by the RCCI combustion mode engine indicated a different PM formation 988 mechanism than baseline CI combustion mode. For investigating the effect of fuel pairs 989 on the particulate characteristics, chemical characterization (organic carbon (OC)/total carbon (TC) ratio) of the PM emitted from CI and RCCI combustion mode engines fueled 990 with different fuel pairs was performed. It was concluded that the fuel chemistry did not 991 992 affect the RCCI combustion mode because all fuel pairs showed similar OC/TC ratios. 993 Smokemeter measurements showed a consistent and significant reduction in BC content 994 of the PM emitted by the RCCI combustion mode engine. However, this does not 995 accurately represent the TPM since the smoke meter only assesses the BC. Kolodziej et 996 al. [228] investigated RCCI combustion mode when both fuels (LRF and HRF) were 997 directly injected into the combustion chamber. Their results showed that PM emitted by the RCCI combustion mode had a bimodal particle size distribution (PSD), which was 998 999 sensitive to the SoI timings of gasoline and gasoline-diesel ratio. They also concluded that advancing the SoI timing of gasoline resulted in lower NMP concentration; however, AMP 1000 1001 concentration increased. Increasing the gasoline-diesel ratio decreased the TPN 1002 emissions. Storey et al. [227] also investigated the PSD sensitivity to the fuels used in the 1003 RCCI combustion mode by employing three fuels pairs: diesel-gasoline, diesel-E85 (85% 1004 ethanol + 15% gasoline), and B20 (20% biodiesel + 80% diesel)-gasoline. There was no 1005 significant difference in the PSD among these fuel pairs, indicating that PSDs from the 1006 RCCI combustion mode were largely insensitive to the test fuel properties. Agarwal et al. [16] compared the particulate characteristics of different LTC modes vis-à-vis baseline CI 1007 1008 combustion mode. They used mineral diesel to achieve PCCI and CI combustion modes and a combination of methanol and mineral diesel as HRF and LRF to achieve the RCCI 1009 1010 combustion mode. They reported a significantly lower particulate number emissions from 1011 both LTC modes than baseline CI combustion mode; however, the RCCI combustion mode emitted relatively larger particles than the other two combustion modes (Figure 16 a). 1012

1013 They suggested that fuel-rich zones in the combustion chamber and a dominant diffusion 1014 combustion phase in the baseline CI combustion mode were the two prime factors for 1015 higher particle concentration emission from the RCCI combustion mode than baseline CI 1016 combustion mode. However, homogeneous fuel-air mixing in RCCI combustion mode 1017 resulted in fewer fuel-rich zones, leading to dominant premixed combustion.





1020 Figure 16: (a) Number-size distribution of particulates and (b) Qualitative correlation

1021 between the number-size and mass-size distribution of particles and (c) Particulate

1022 1023

loads [16]

bound trace metals emitted in CI, PCCI, and RCCI combustion modes at varying engine

Statistical analysis of particulate results exhibited that the number concentration of 1024 particles was higher in CI combustion mode; however, the RCCI combustion mode emitted 1025 1026 relatively higher particulate mass (Figure 16 b). The particulate number-mass 1027 distribution showed that the PCCI combustion mode emitted lower particle numbers and 1028 mass than the RCCI combustion mode. Relatively superior in-cylinder conditions in the 1029 PCCI combustion mode than RCCI mode might be a possible reason for this behavior, 1030 promoting homogeneous fuel-air mixing, leading to lesser soot nuclei formation. Their 1031 study concluded that all LTC modes could reduce both PM and NOx emissions; however, 1032 the RCCI combustion mode was more dominant in NOx and PM reduction due to the 1033 combined effects of LTC and the use of oxygenated fuels. Particulate bound trace metal 1034 analysis was another important aspect related to particulate toxicity. Agarwal et al. [16] 1035 compared the particulate bound trace metal concentrations emitted in the PCCI, RCCI, 1036 and baseline CI combustion modes and reported significantly lower trace metals from the 1037 PCCI and RCCI combustion modes than the baseline CI combustion mode (Figure 16 c). 1038 They suggested that relatively lower peak in-cylinder temperature was a major factor 1039 responsible for lesser trace metals such as Ni, Ar, Pb, Cr, etc., in both these LTC modes. 1040 In summary, this section covers the findings of the experimental investigations of the 1041 RCCI combustion mode, particularly emissions. Attention was given to higher HC and CO 1042 emissions than other combustion modes such as baseline CI combustion mode, even when using fuel combinations such as diesel and methanol (formaldehyde was also detected). 1043 1044 Additionally, detailed studies on the emission characterization, particularly PM emission, were summarized, concluding that RCCI combustion mode emitted lesser BC but might 1045

emit a higher mass of non-BC particles. It also emerged that the chemical characterization
of particulate matter in terms of OC/TC ratio was not affected by the fuel chemistry.
Finally, RCCI combustion mode exhibited fewer trace metals than baseline CI combustion
mode.

1050 2.3 Alternative Fuels for RCCI Combustion mode

1051 Alternative fuel utilization potential is an important feature of the RCCI combustion 1052 mode. RCCI mode can utilize many alternative fuels, ranging from high-cetane to low-1053 cetane fuels. The RCCI combustion mode with alternative fuels exhibits higher efficiency 1054 and cleaner combustion than CDC. This feature makes RCCI combustion mode suitable in the current scenario when the transport sector is battling energy security and 1055 emissions issues. A wide range of alternative fuels has been investigated in RCCI 1056 1057 combustion mode in the last few years. These are divided into two main categories: 1058 alternative fuel usage as (i) HRFs, and (ii) LRFs.

1059 2.3.1 Alternative Fuels as HRFs in RCCI Combustion mode

1060 Most RCCI combustion mode investigations were carried out using mineral diesel as HRF. To explore the effects of HRF properties on RCCI combustion mode, Ryskamp et al. [229] 1061 1062 carried out RCCI combustion mode experiments using nine different compositions of 1063 mineral diesel, having different cetane numbers, aromatic content, and distillation 1064 temperatures. They reported that the cetane number of HRF was dominant compared to 1065 other fuel properties. They concluded that a lower cetane number of HRF resulted in 1066 higher NOx and PRR; however, higher cetane fuels showed greater potential for higher 1067 engine load operation. Therefore, in RCCI combustion mode, higher cetane alternative fuels are preferred [230-236] to compensate for the combustion delay that is typical in the 1068 1069 mode due to the elevated proportions of LRF. Many alternative fuels such as biodiesel, 1070 dimethyl ether (DME), etc., have been explored as alternatives to mineral diesel [230-244]. Recent studies have focused on biodiesel types, achieving a superior reactivity 1071

1072 stratification due to the higher cetane number of biodiesel than baseline mineral diesel. 1073 The cetane number of biodiesels depends on the saturation and molecular structure of the 1074 constituent hydrocarbons from the oils or facts from which the fuels are produced, which 1075 in turn affect the oxygen the molecule can contain. In the case of biodiesel, the oxygenation 1076 of the molecule is generally higher, however there is high variability in the determination 1077 of this metric even with tow fuels coming from the same feedstock [245]. Wang et al. [235] used polyoxymethylene dimethyl ethers PODEn as HRF and gasoline as LRF to achieve 1078 1079 RCCI combustion mode at higher engine loads. They reported that relatively higher 1080 cetane number of biodiesel, it created a more reactivity stratification compared to mineral 1081 diesel. This helped in achieving superior combustion control, especially at higher engine 1082 loads. Relatively higher PM reduction potential of PODEn than mineral diesel is another 1083 reason due to which PODEn was extensively explored as HRF in the RCCI combustion mode. Tong et al. [154] compared the RCCI combustion mode and performance 1084 characteristics of a heavy-duty engine fueled with gasoline-PODEn and gasoline-diesel. 1085 1086 They concluded that gasoline-PODEn RCCI combustion mode exhibited more stable 1087 combustion characteristics. Near-zero PM emissions from the gasoline-PODEn-fueled 1088 RCCI combustion mode were another important finding of their study; however, they did not discuss these characteristics elaborately. To further explain the reasons for superior 1089 1090 emission characteristics of PODEn as the directly injected HRF in the RCCI combustion 1091 mode, Wang et al. [236] performed detailed simulations of the in-cylinder fuel distribution of gasoline-PODE₃ fueled RCCI combustion mode. They reported relatively superior 1092 1093 management of the in-cylinder equivalence ratio and reactivity distribution of PODE₃ for 1094 improving the RCCI combustion mode engine performance more flexibly than baseline mineral diesel. García et al. [237] explored the potential of PODEn in the RCCI 1095 1096 combustion mode wherein they used diesel-PODEn blends as HRF and gasoline as the premixed LRF in different premixed ratios. They concluded that the higher blends of 1097

1098 PODEn with mineral diesel complied with the Euro VI emission regulations even at 1099 higher engine loads (up to 80% load). In a few other studies, this technique was also 1100 explored, where lower blends of PODEn with mineral diesel resulted in superior 1101 combustion efficiency for the same NOx levels. This increase in combustion efficiency 1102 showed higher brake thermal efficiency [240].

Song et al. [238] evaluated the suitability of PODEn in the RCCI combustion mode by 1103 comparing emissions from natural gas-diesel and natural gas-PODEn-fueled RCCI 1104 1105 combustion mode engines. They demonstrated that a combination of natural gas-PODEn resulted in significantly lower HC, CO, NOx, and PM emissions along with superior 1106 1107 engine performance. The higher difference in reactivities of natural gas and PODEn might 1108 be a possible explanation for this trend. Hararia et al. [231] explored a new strategy, in 1109 which they used a mixture of CNG and compressed biogas (CBG) as the LRF and different blends of Thevetiaperuviana methyl ester (TPME) were used as the HRF. The main 1110 objective of this research was to enhance the load limit of RCCI combustion mode. They 1111 1112 reported that this fuel combination resulted in relatively higher BTE and NOx emissions 1113 than the reference RCCI combustion mode fueled with CNG-mineral diesel; however, CO 1114 and HC emissions were marginally lower. Aydin [246] also used CNG as LRF in RCCI combustion mode along with biodiesel as HRF. They used a higher fraction of safflower 1115 1116 biodiesel (up to 95%) to achieve RCCI combustion mode and compared the results with conventional CI combustion mode. They reported that the RCCI combustion mode with 1117 higher fraction of biodiesel as HRF in RCCI combustion mode resulted in shorter 1118 1119 combustion duration compared to conventional CI combustion mode.

Due to the significant effect of fuel properties of HRF on the RCCI combustion mode,
researchers also used DME as HRF in the RCCI combustion mode. Kakoee et al. [232]
investigated the RCCI combustion mode using natural gas as LRF and DME as HRF.
They added hydrogen to the natural gas to improve the control over the global reactivity

and reactivity stratification. They reported that DME decomposition during the RCCI 1124 1125 combustion mode was an important phenomenon that affected the fuel mixture's cetane 1126 number and the HRR. A higher cetane number of DME becomes less effective in such 1127 conditions due to these changes in the mixture properties before the combustion, leading to lower combustion quality, higher emissions, and lower power output than baseline 1128 1129 diesel [232]. Jin et al. [233] performed detailed experimental investigations to understand 1130 the ignition dynamics of the DME-methane mixture, which were explored as a potential 1131 fuel pair in the RCCI combustion mode. They explained the ignition characteristics of the 1132 DME-methane mixture and reported that the initial ignition events were similar in all 1133 cases of RCCI combustion mode. However, for the DME-methane case under turbulent 1134 conditions, the formation of high-temperature autoignition kernels was relatively 1135 advanced compared to a homogeneous mixture. They indicated the existence of typical 1136 tetrabranchial flames, such as cool flames, fuel-rich premixed flames, diffusion flames, 1137 and fuel-lean premixed flames, where the fuel-lean premixed flame branches finally 1138 trigger the premixed methane-air flames. Krisman et al. [247] also performed similar 1139 investigations using DME as HRF and reported that DME exhibited a two-stage ignition 1140 in a turbulent mixing layer. They reported that cooler regions in the combustion chamber affect the timings and location of ignition in the second stage auto-ignition. 1141

Park et al. [234] explored the RCCI combustion mode characteristics using DME and 1142 ethanol as HRF and LRF, respectively. Few other researchers used DME in R-RCCI 1143 1144 combustion mode to achieve superior combustion control than the RCCI combustion mode. 1145 They reported that the DME-ethanol fueled RCCI combustion mode resulted in lower 1146 emissions than biodiesel-ethanol and diesel-ethanol dual-fuel combustion. They reported a significant reduction in ISNO_x in the DME-ethanol fuel pair without deterioration of 1147 1148 IS_{soot}. Yao et al. [149] and Chen et al. [182] assessed DME as HRF in a modified singlecylinder diesel engine, in which methanol was injected directly in the combustion chamber 1149

1150 as LRF. Yao et al. [149] reported that premixed DME enhanced the in-cylinder thermal 1151 atmosphere, which resulted in superior combustion than conventional RCCI combustion 1152 mode. Chen et al. [182] performed a detailed investigation of methanol-DME fueled RCCI combustion mode by varying fuel injection parameters such as fuel injection timings of 1153 1154 methanol, FIP, etc. They reported that varying SoI timings of methanol exhibited a weak effect of the LTHR in the DME as HRF; however, it significantly affected the HTHR. They 1155 suggested that the overall combustion duration of methanol-DME fueled RCCI 1156 1157 combustion mode could be reduced by increasing the FIP of methanol [182].

In summary, RCCI combustion mode can be enhanced by using HRF, other than mineral diesel. HRF such as PODEn has fuel properties that aid in making the combustion more controllable, thus allowing to cater to higher load operating points in the LTC mode. With the use of biodiesel, higher combustion efficiency and brake thermal efficiency can be achieved. Additionally, these alternate fuels enhance the potential of reducing soot emissions while keeping the same level of NOx emissions as the baseline mineral diesel.

1164 2.3.2 Alternative Fuels as LRFs in RCCI Combustion mode

1165 Many alternative fuels have been extensively explored for IC engines in the past five decades. Among those alternative fuels, alcohols have shown significant potential to be 1166 utilized as an alternative to mineral diesel and gasoline. However, for practical 1167 1168 applications, alcohols have been promoted more as an alternative to gasoline. In many 1169 countries, gasoline has been replaced with alcohol blended gasoline, in which 20% (v/v) 1170 alcohols are blended with gasoline. Unlike gasoline engines, the use of alcohol in diesel 1171 engines is challenging. In many studies, alcohols have been explored as a direct 1172 replacement of mineral diesel in CDC; however, igniting alcohols in diesel engines is very 1173 difficult due to their lower cetane number. In the literature, many techniques have been 1174 proposed for alcohol utilization, such as fuel blending [241-244, 247-248], port fumigation 1175 [249-255], and dual-fuel emulsions [221]. The blending of alcohols with mineral diesel has

1176 attracted researchers due to its simplicity. In most blending strategies, the suitability of 1177 alcohol has already been justified [256-258]. However, most studies concluded that 1178 blending strategies can be used up to only a certain fraction of alcohol in diesel engines 1179 as high alcohol proportions are closely related to rust and corrosion and due to the 1180 previously mentioned low cetane number of alcohols ignition becomes more difficult, requiring higher proportions of HRF and not taking advantages of the RCCI combustion 1181 mode as the higher proportions of HRF get the combustion closer to CDC. Moon et al. 1182 1183 [259] conducted experiments using mineral diesel-ethanol blends. They reported that lower ethanol blends with mineral diesel (<30%v/v of ethanol) exhibited stable combustion 1184 1185 without significant variations in fuel spray atomization characteristics. However, higher 1186 blends of alcohol with diesel pose several serious issues such as phase separation 1187 (especially for methanol), inferior fuel spray atomization, poor combustion, engine performance, and serious material compatibility issues with the fuel injection system. 1188 1189 Therefore, alcohol blending with mineral diesel has not been implemented commercially. 1190 Methanol utilization was then explored using diesel-methanol compound combustion (DMCC) [250-251]. Yao et al. [250] performed DMCC experiments and reported a 1191 1192 significant reduction in the NOx and soot emissions simultaneously. Haribabu et al. [252] investigated a similar combustion mode using direct injection of methyl ester and port 1193 1194 injection of methanol via carburetion. They also reported relatively lower NOx and smoke 1195 emissions and improved fuel economy.

The RCCI combustion mode showed tremendous potential to use alternative fuels in the last few years. In RCCI combustion mode, CNG [260-263], syngas [264], alcohols including methanol [122, 253, 265-266], ethanol [267-268], and butanol [177-178, 269] have been injected into the port as LRF. These LRFs helped in extending the load range of engine operation due to their higher resistance to auto-ignition (less knocking), higher reactivity gradient (optimum combustion phasing), and charge-cooling effect (due to their higher 1202 latent heat of vaporization). In most preliminary investigations of the RCCI combustion 1203 mode, methanol was used as the LRF due to its lower reactivity (lower cetane), making it 1204 suitable for achieving a reactivity stratification with mineral diesel. The gasification of coal can be done to produce methanol; however, in the last few years, production of 1205 1206 methanol from black liquor gasification, biomass gasification, and the reaction of 1207 hydrogen and CO_2 (directly from the atmosphere or from the coal-fired power plants, 1208 industrial flue gases, etc.) is attracting significant global attention [270]. The presence of 1209 relatively higher oxygen than the other alcohols also makes it more suitable, promoting 1210 soot oxidation, leading to relatively lower particulate emissions [241].

1211 Excessive LRF leads to incomplete combustion, resulting in higher HC and CO emissions, 1212 especially at lower loads. Sayin et al. [242-244] investigated the RCCI combustion mode 1213 using mineral diesel-methanol fuel-pair and reported a significant reduction in NOx and PM than conventional CI combustion mode. They also emphasized PM reduction due to 1214 the use of methanol. They reported that the absence of a C-C bond was the main reason 1215 1216 for lower PM emissions from mineral diesel-methanol-fueled RCCI combustion mode. A 1217 few studies reported that the lack of a C-C bond also reduces the formation of polycyclic 1218 aromatic hydrocarbons (PAHs), which act as soot precursors.

Like methanol, ethanol was also explored as a potential candidate for LRF in the RCCI 1219 1220 combustion mode. Hanson et al. [271] performed preliminary RCCI combustion mode investigations on a light-duty, multi-cylinder diesel engine using a blend of ethanol and 1221 1222 gasoline (E20) as the LRF. They reported that the lower reactivity of ethanol was suitable 1223 for achieving sufficient reactivity gradient for the RCCI combustion mode. They found 1224 lower PRR in E20-mineral diesel-fueled RCCI combustion mode than gasoline-mineral diesel-fueled RCCI combustion mode. Apart from these findings, they also reported that 1225 1226 E20 also helped in increasing the peak power output because ethanol is less prone to autoignition. Qian et al. [272] conducted RCCI mode experiments using n-heptane as the HRF 1227

and three different LRFs, namely ethanol, n-butanol, and n-amyl alcohol. They reported 1228 1229 that all three LRFs showed similar combustion characteristics at the lower premixed 1230 ratios; however, ethanol-n-heptane fueled RCCI combustion mode resulted in relatively retarded combustion events at higher premixed ratios. This was also visible in the 1231 emissions characteristics, where n-heptane-ethanol-fueled RCCI combustion mode 1232 exhibited a relatively greater reduction in NOx and soot emissions than the other two fuel 1233 pairs. Agarwal et al. [150] performed RCCI combustion mode using n-butanol to explore 1234 1235 its potential as the LRF. They performed RCCI combustion mode experiments at different 1236 premixed ratios of n-butanol at varying engine loads. They reported that n-butanol 1237 exhibited relatively superior combustion characteristics even at higher premixed ratios; 1238 however, they did not observed any significant variation in performance parameters. They 1239 suggested that the relatively higher cetane number of n-butanol than other alcohols, such 1240 as methanol and ethanol, was the main reason for superior engine performance. To further explore the effect of fuel properties on the RCCI combustion mode, Agarwal et al. 1241 1242 [154] performed a detailed experimental study on the RCCI combustion mode using 1243 methanol, ethanol, and n-butanol as the LRF.



Figure 17: (a) In-cylinder pressure and HRR variations w.r.t. CAD, (b) Number-size and 1245 mass-size correlations of particles at different premixed ratios of Methanol, Ethanol, 1246 and Butanol at constant engine speed (1500 rpm) and load (3 bar BMEP) [154] 1247 They performed experiments at constant engine load and the premixed ratio of the LRF. 1248 1249 They found quite a similarity between the RCCI combustion mode fueled with methanol and ethanol as the LRF; however, butanol fueled RCCI combustion mode exhibited closer 1250 1251 similarity with the CI combustion mode (Figure 17 a). This was also remarkably reflected 1252 in the emission characteristics, where methanol and ethanol-fueled RCCI combustion modes showed greater potential for PM reduction than the butanol (Figure 17 b). They 1253 1254 concluded that all three alcohols could be used as the LRF in the RCCI combustion mode 1255 to simultaneously reduce NOx and PM emissions.

The use of biogas in IC engines is not a new area; however, the performance of biogas-1256 1257 fueled engines using conventional combustion mode has not been found satisfactory. 1258 Therefore, biogas has always been used in dual-fuel combustion mode engines to achieve acceptable performance and emissions. Prajapati et al. [273] investigated the potential of 1259 biogas utilization as the LRF in the RCCI combustion mode. They reported that the RCCI 1260 combustion mode engine fueled with mineral diesel-biogas fuel-pair exhibited lower NOx 1261 and CO₂ emissions than the CDC mode. In another study by Bora et al. [274], biogas-rice 1262 1263 bran biodiesel fuel-pair was used in the RCCI combustion mode, delivering superior 1264 engine performance due to superior reactivity stratification between the biogas and the 1265 biodiesel. In a comparative investigation by Verma et al. [275-276], the effectiveness of 1266 biogas, CNG, and hydrogen as the LRF was assessed. In the exergy analysis, it was 1267 observed that mineral diesel-hydrogen-fueled RCCI engines exhibited better engine performance than the ones using biogas. This might be due to higher free radicals such as 1268 1269 O, H, and OH, enhancing the net reaction rate and a higher net HRR [277]. Use of 1270 hydrogen in RCCI combustion mode also beneficial due to its higher laminar burning 1271 velocity, leading to shorter combustion duration. The replacement of mineral diesel with 1272 biodiesel showed slightly inferior engine performance, though. In a similar investigation by Khatri et al. [278], hydrogen addition in biogas in dual-fuel mode improved the engine's 1273 1274 efficiency. Ebrahimi et al. [279] reported that the addition of hydrogen also helped in the dissociation of methane present in biogas, which resulted in a relatively shorter 1275 combustion duration of the RCCI combustion mode engine fueled with biogas-mineral 1276 1277 diesel fuel pair. They also reported about the fuel chemistry related to hydrogen addition 1278 in RCCI combustion mode. They observed that the formation of OH radicals delayed due to increasing fraction of hydrogen, leading to relatively longer ignition delay. 1279

1280 For this reason, in many studies, reformed biogas was also investigated [280-282].
1281 Mahmoodi et al. [283] investigated reformed biogas-mineral diesel-fueled RCCI

combustion mode using a 3D modeling approach. They used higher premixed ratios of 1282 1283 reformed biogas and observed significantly reduced mean combustion temperature in the 1284 RCCI combustion mode than the baseline CI combustion mode. This might be due to the 1285 relatively weaker effect of CO_2 present in biogas on the LTC strategies than the CDC mode [284]. CO_2 present in the biogas also affected the combustion characteristics due to 1286 1287 the dominant role of thermal, reactive, and transport properties of CO_2 on the ignition characteristics. Nieman et al. [260] used the KIVA3V CFD software and CHEMKIN, the 1288 1289 chemical kinetics analysis tools, to investigate the diesel-CNG dual-fuel RCCI combustion 1290 mode. They observed superior performance and lower exhaust emissions in the RCCI 1291 combustion mode at low engine loads without EGR. However, EGR controlled the RCCI 1292 combustion mode at higher engine loads. Aydin [246] also used CNG as LRF in RCCI 1293 combustion mode and reported that CNG is capable of reducing high gas temperature, which is useful for reduction in NOx. They reported that biodiesel/CNF fueled RCCI 1294 1295 combustion mode reduced NOx emissions in proportion to the amount of CNG. Martin et 1296 al. [285] used a blend of propane and DME as LRF to achieve RCCI combustion mode. 1297 They reported that this fuel combination can result an intermediate mode of combustion 1298 namely premixed dual-fuel combustion (PDFC), which lies in between CI and RCCI mode. In this study, they found relatively higher NOx from PDFC compared to RCCI combustion 1299 mode, however, relatively lower PRR makes this combustion technique more suitable at 1300 higher engine loads. Relatively higher BTE of PDFC was another important finding of 1301 1302 their study.

In summary, this section reports the findings on several studies performed on the RCCI combustion mode using LRF other than gasoline. It could be concluded that though alcohols are potentially superior LRF than fossil fuels, their higher concentrations could hamper the combustion stability and cause complications such as fuel blend separation, in addition to other engine operational issues. Since alcohols have a lower cetane number,

1308 their ignition in RCCI combustion mode presents some challenges. Other alternative 1309 LRFs extend the operational range of RCCI combustion mode if they have higher cetane 1310 numbers, higher resistance to auto-ignition, higher reactivity gradient, and charge cooling effect, preventing some of the better-known issues of RCCI combustion mode, such as 1311 probable knocking. Using some of these alternate fuels as the LRF reduced soot emissions, 1312 1313 especially when the C-C bonds are absent in the test fuel. Hydrogen was also tested as 1314 the LRF and helped enhance the RCCI mode engine performance, as reported by some studies due to higher net HRR caused by free radicals such as O, H, and OH. 1315

1316 2.4 Fundamental Investigations of RCCI Combustion mode

1317 Literature shows that LTC strategies have been extensively assessed in various all-metal 1318 engines, including heavy-duty engines. Most experimental studies in metal engines have focused on advanced combustion strategies and combustion, performance, and emission 1319 1320 characterization. However, LTC strategies, especially RCCI combustion mode, require a finer understanding that can only be explored using fundamental experimental 1321 investigations. RCCI combustion mode is a kinetically controlled combustion, 1322 1323 significantly affected by the in-cylinder processes, including charge stratification, 1324 reactivity gradients, ignition, and flame evolution, which need to be explored in detail using advanced optical diagnostic techniques [135, 286-587]. This section deals with the 1325 1326 methodology and results of fundamental optical investigations using different test fuels, 1327 control techniques, engine loads, etc.

Liu et al. [288] performed a comparative investigation of PPC and RCCI combustion modes using different optical diagnostics techniques such as fuel-tracer planar laserinduced fluorescence (PLIF) imaging, high-speed natural flame luminosity (NFL) imaging, and Formaldehyde and -OH PLIF imaging. The researchers used a quartz window-fitted piston engine for the optical studies (Figure 18).



Figure 18: Scheme of the combustion chamber and field of view of the optical diagnostics[288].

Their objective was to focus on the flame development process and assess the distribution 1336 1337 of intermediate combustion products and free radicals. The authors used iso-octane as the LRF and n-heptane as the HRF to achieve the RCCI combustion mode; however, PRF70 1338 was used as the test fuel in the PPC mode. They kept most of the operating conditions 1339 identical in both combustion modes. They reported that RCCI combustion mode exhibited 1340 relatively shorter ignition delay than the PPC mode, where the RCCI case has a higher 1341 1342 minimum equivalence ratio (0.54) and fuel rich regions with higher reactivity where the fuel is more easily auto-ignited (thus reducing the ignition delay). Optical investigations 1343 NFL at 70% premixed ratio (70% LRF) revealed that RCCI combustion mode does not 1344 1345 exhibit flame front propagation (Figure 19). The locally high reactivity regions in the 1346 RCCI combustion form flames in the periphery of the cylinder during the first crank-angle degrees (CAD), when in the central region of the cylinder the reactivity of the mixture is 1347 lower. After the 3.1° CAD the flame reaches the central region. In the case PPC, auto-1348 1349 ignition kernels appear at the 13.9° CAD in local fuel-rich zones which later show flame propagation fronts. The difference between the two modes in terms of flame propagation 1350 lies in the fact that the fuel reactivity stratification of RCCI induces more auto-ignition in 1351 1352 the earlier combustion stages, which translates into a faster combustion rate than PPC

whose auto-ignition depends on the local equivalence ratio (the difference between fuel-1353 1354 rich and fuel-lean-regions). The researchers later increased the premixed ratio of RCCI (only 5% of the input energy coming from the HRF), finding that the rate of combustion 1355 of RCCI combustion mode decreased. As the RCCI combustion with higher premixed 1356 ratios gets closer to the combustion in spark-ignition engines, the increase in premixed 1357 ratio also resulted in a flame front propagation in RCCI combustion mode; however, the 1358 laminar flame speed was lower than the PPC mode. The authors concluded that the degree 1359 1360 of fuel stratification played a crucial role in auto-ignition and flame front propagation in the RCCI combustion mode, and that the degree of stratification can be controlled by 1361 1362 different fuel injection strategies.

1363 combustion mode





Figure 19: Time-resolved single-shot true-color images and flame boundary evolution of
PPC and RCCI modes firing cycles. [Top] PPC [Bottom] RCCI [288].

In RCCI combustion mode, different optical diagnostics techniques are limited by the PRR 1367 1368 and soot emissions. In most optical investigations, experiments are limited to a certain 1369 part-load condition, depending on the strength of optical components. The optical diagnostics of RCCI combustion mode under higher engine loads have higher practical 1370 significance since it provides vital information about the limiting criteria. Kokjohn et al. 1371 [135, 289] performed optical experiments only up to 4.2 bar gross IMEP due to the limited 1372 1373 optical strength of the optical components of the engine using high-speed imaging and 1374 PLIF techniques to investigate the RCCI combustion mode. They concluded that local fuel 1375 reactivity controlled the ignition sites. They found that ignition sites were more dominant 1376 in high reactivity zones; therefore, they appeared downstream of the injected fuel jets. In 1377 such conditions, HRR can be controlled by fuel stratification. They further extended this work and included chemical kinetics modeling [135] to explore the role of fuel reactivity 1378 1379 stratification, equivalence ratio, and temperature on the RCCI combustion mode control. 1380 They concluded that the fuel reactivity was the main parameter, which controlled the 1381 ignition delay, and fuel concentration and temperature stratification exhibited a 1382 relatively weaker effect on the combustion. The combustion images captured by the high-1383 speed camera also showed that RCCI combustion mode was significantly affected by the degree of fuel stratification. Upon comparing high-speed imaging, they reported that PLIF 1384 1385 imaging was more informative and provided valuable information about forming formaldehyde and OH radicals in the two-phase (LTHR and HTHR) ignition of RCCI 1386 combustion mode. However, the high-speed imaging provided only a two-dimensional 1387 1388 natural flame luminosity in a three-dimensional reaction zone. This technique also had 1389 another limitation. It could not distinguish between the LTHR and HTHR phases, an important aspect of RCCI combustion mode. However, this issue of high-speed imaging 1390 was reported only in few studies related to RCCI combustion mode at different degrees of 1391 fuel stratification. 1392

Tang et al. [290] also explored the effects of fuel stratification on the RCCI combustion mode using several optical diagnostics techniques such as PLIF under non-reactive conditions, Time-resolved natural combustion luminosity imaging, and single-shot OH PLIF imaging in a light-duty optical engine. PLIF technique was used to quantify the fuel-air equivalence ratio and PRF number. Experiments were performed at different injection timing (Figure 20).



1399

Figure 20: PLIF images acquired at -5° CA aTDC with (a) PI and (b) DI. The results are
averaged from 10 single-shot PLIF images [290].

The authors reported that higher fuel concentration and reactivity regions were affected 1402 by the SoI timing, which moved downstream to the edge of the combustion chamber with 1403 retarded SoI timing of n-heptane from -90° CA aTDC (RCCI-90 case) to -10° CA aTDC 1404 1405 (RCCI-10 case). They reported that combustion took place in two stages in the case of 1406 RCCI-10. In the first stage, an auto-ignition was observed around the combustion 1407 chamber due to high reactivity regions. In the second stage, auto-ignition occurred in the 1408 low reactivity regions in the central part of the combustion chamber. This two-stage heat 1409 release process was the main reason for relatively lower PRR in the RCCI combustion 1410 mode. Formaldehyde and OH PLIF images exhibited a more stratified fuel distribution in 1411 retarded SoI timing of n-heptane. Another important observation of their investigation

1412 was a relatively slower formaldehyde consumption rate and formation of OH radicals at





1414

Figure 21: True-color natural luminosity (NL) image sequences for the cases of RCCI-90
(top), RCCI-25 (middle) and RCCI-10 (bottom), respectively [290].

In Figure 21, a time-resolved, single-shot NL image sequence for each case was captured from a typical engine cycle. The crank angle position for 50% (CA₅₀) and 90% (CA₉₀) cumulative heat-release (CHR) were marked on the lower right side of each corresponding image. The boundary of the combustion chamber is shown with a white circle at the start of each image sequence. Field of view (FoV) used in the OH and formaldehyde PLIF imaging was shown with a white dashed line at a 6.6° crank angle for the RCCI-10 case.





1424 Figure 22: Fuel-air equivalence ratio (left column) and PRF number (right column)
1425 distribution under different n-heptane direct injection timings [290].

1426 Figure 22 shows that the combustion duration (from CA₁₀to CA₉₀) in RCCI-10 was longer

1427 $\,$ than RCCI-25 case. A higher degree of fuel stratification was the main reason for a

relatively longer combustion duration in RCCI-10, promoting higher soot production and
reduced PRR. OH PLIF images exhibited that the HTHR phase of RCCI combustion mode
could be extended up to the central part of the combustion chamber. However, Musculus
et al. [291] used a low-load LTC conceptual model and suggested that no HTHR occurs in
the central part of the combustion chamber, where the HC forms mainly.





Figure 23: HTHR process shown by single-shot false-color images of OH-LIF for
different direct injection timings (Top row: RCCI-90, Middle row: RCCI-25, Bottom row:
RCCI-10). The white dashed line marked by "b" shows low-reactivity regions not
occupied by OH radicals [290].

1438 Tang et al. [290] investigated the OH PLIF signal using different SoI timings (Figure 23). They reported that OH radicals begin to emerge at 1°, 1°, and 7° CA for RCCI-90, RCCI-1439 1440 25, and RCCI-10, respectively, in the regions where formaldehyde vanishes. In RCCI-90, the most field of view was occupied by the OH radicals after 3° CA, showing a uniform OH 1441 radical distribution. After that, the OH PLIF signal remained uniform for the next several 1442 1443 crank angle degrees; however, the signal intensity became weak after 30° CA. Comparison 1444 of OH PLIF signals of different SoI cases showed relatively slower development in the RCCI-10 case than in the other two cases. At 8° CA, OH radicals first occupied the regions 1445 around the combustion chamber, where n-heptane resides, fuel reactivity was high, and 1446 there were no signs of OH radicals in the regions of lower reactivity, as shown by white 1447

1448 dashed lines marked by 'b' (Figure 23). However, in the following crank angles (9°, 10°, 1449 and 11°), a weak OH PLIF signal appeared gradually in this region. By 11° CA, OH PLIF signal of intense non-uniformity occupied the most field of view. Kokjohn et al. [135] 1450 1451 explored the role of equivalence ratio, temperature, and fuel reactivity stratification on 1452 the HRR using a combination of optical diagnostics and chemical kinetics modeling. They used iso-octane as the LRF, injected during the intake stroke to provide sufficient time 1453 for fuel-air premixing, and then injected n-heptane as the HRF at the end of the 1454 1455 compression stroke. Results showed that the ignition first occurred in the squish region and then expanded towards the center of the combustion chamber. They also used PLIF 1456 to get the quantitative information related to the fuel concentration distributions before 1457 1458 the ignition. PLIF showed that the combustion events followed the direction of reactivity 1459 gradient, which was an important characteristic of the RCCI combustion mode (Figure 1460 24).



Figure 24: Sequence of ensemble-averaged PRF maps at several crank angle positions
during a common-rail injection event [152]. The time in crank angle degrees after TDC
is shown in each image's upper left-hand corner. The PRF maps were generated from
the vapor-fuel concentration measurements with the camera viewing downward through
the cylinder-head window. The relative error in the PRF number images is
1467 approximately 7% of the measured value with a filter size of 0.25 mm and 3.4% of the
1468 measured value with a filter size of 1 mm.

1469

1470 The researchers employed a modeling technique to explore the effects of temperature, 1471 equivalence ratio, and PRF number stratification on the RCCI combustion mode. They 1472 reported that reactivity stratification was the prime factor that controls the combustion 1473 chamber's ignition location and growth rate. Compared to reactivity stratification, a 1474 relatively weaker effect of equivalence ratio and temperature on the RCCI combustion 1475 mode was another important finding of this investigation.

1476 In other studies, the RCCI combustion mode was compared to a diesel-pilot injected 1477 natural gas-fueled engine. A diesel injection near the TDC is a strong ignition source for 1478 an overall lean natural gas-air mixture. In diesel-pilot initiated combustion, the combustion is presumed to take place via deflagration [292]. Still, in RCCI combustion 1479 1480 mode, the role of flame propagation is unclear as only one study has experimentally 1481 explored this question. Kokjohn [289] used laser ignition to initiate flame propagation. It 1482 was found that flame propagation could exist in the RCCI conditions, as high-speed 1483 chemiluminescence showed luminous regions propagated away from the ignition spot. Flames did not propagate in every cycle, but the probability of successful flame kernels 1484 1485 increased with increasing equivalence ratio, suggesting that higher equivalence ratio regions were more likely to support the flame propagation. The flame growth rate was 1486 similar to the autoignition reaction fronts, indicating that flames could propagate from 1487 1488 the auto-ignition sites. Still, the two combustion fronts were not discernable with current 1489 optical measurements. KIVA 3D CFD tools allowed newer insight into the combustion process, combustion of intermediate species, and sources of inefficiency and losses. As 1490 1491 expected, RCCI combustion mode temperatures were much lower, and the peak temperature locations were farther away from the piston and cylinder walls. This reduced 1492

the heat losses to the cooling system, which was one of the reasons for the increasedthermal efficiency of the RCCI combustion mode [144].

1495 Tang et al. [186] performed RCCI combustion mode investigations to extract detailed 1496 information about the RCCI combustion mode and chemical interactions between the LRF 1497 and HRF in different engine operating conditions. They performed experiments in an 1498 optical engine using premixed iso-octane, assisted by the two-stage reactions of n-heptane. 1499 They used high-speed imaging to visualize the NFL and Cantera-based data processing 1500 to understand detailed combustion kinetics. They observed a shifting behavior of 1501 combustion from two-stage ignition to three-stage ignition dominated by the retarding of 1502 the iso-octane injection timing due to the reduction of the local air-fuel mixture reactivity 1503 and weakening of the chemical interaction between the HRF and LRF, which lowers the 1504 HRR. The three stages of the heat release correspond respectively to LTHR, HCHR and 1505 PCHR. The CANTERA analysis of the PCHR showed that varying the HRF injection 1506 timings towards later injections resulted in different combustion reactions with more soot 1507 precursor formation potential (which is also observed in the higher luminosity of the NFL 1508 results), especially in the primary reacting regions. This is correlated with the higher 1509 stratification of both the fuel and flame associated with earlier injections. Results in Figure 25Results exhibited that the advanced injection timings (SoI = -27° and -7°) 1510 1511 resulted in an earlier ignition kernel, primarily near the combustion chamber bowl. In SoI = -27° aTDC, dominant premixed combustion was observed, which was seen as blue 1512 ignition kernels appearing at 3° CA. A significant heat was released rapidly at this stage, 1513 1514 due to which the whole combustion chamber was occupied with the blue flames at 3.7° 1515 CA. After this, a dominant effect of fuel stratification was observed in the combustion images; however, after ~9° CA, the luminosity was dominantly controlled by the soot 1516 1517 oxidation.



1519 Figure 25: Experimentally measured natural flame luminosity images at various crank
1520 angle positions [186].

A combined analysis of n-heptane and iso-octane reaction mechanisms exhibited several 1521 1522 reactive species formations from the premixed n-heptane promoted iso-octane, which further accelerated combustion. The consumption of n-heptane (injected at -360° aTDC) 1523 1524 was similar independently of the SOI of the iso-octane, at was almost depleted before TDC 1525 being consumed in low and intermediate-temperature combustion processes. During the 1526 consumption of n-heptane, CH₂O and CO accumulated while the amount of OH remained at a low level until the rapid consumption of *iso*-octane, which led to the prompt heat 1527 1528 release.



1529

Figure 26: Comparison of the predicted distributions of (a) temperature and (b)
exothermic HRR at various crank angles. The predicted results are on a plane 10 mm
below the cylinder head [186].

Tang et al. [186] investigated the predicted distributions of temperature and recalculated chemical HRR for three SoI cases at the height of 10 mm below the cylinder head (Figure 26). They observed a much wider main heat release region in the space, which approached the central part of the combustion chamber when advancing the SoI timings for the isooctane. The stratified heat release was observed at all SoI timings. At retarded SoI timings, the primary reacting locations moved towards the upstream spray, which are the fuel-rich regions. The PCHR is dominated by the combustion of the iso-octane fuel, as the 1540 heat release contribution from the n-heptane is low in relation. In the premixed 1541 combustion of n-heptane, heat release was predominantly controlled by the HCO + O_2 = $CO + HO_2$ reaction; however, this reaction shifted to hydrogen-oxygen reaction 1542 $(H + O_2(+M) = HO_2(+M))$ in the presence of increased temperatures. Both reactions belong 1543 1544 to the intermediate-temperature reactions and the equivalence ratio of the premixed 1545 mixture was low, thus the HRR remains low in these stages. During the primary combustion stage, the heat release is dominated instead by the $HO_2 + OH = H_2O + O_2$ 1546 reaction, the formation of OH is propitiated by the $H + O_2(+M) = HO_2(+M)$ reaction 1547 surrounding the primary reacting kernels, and furthers the combustion process. 1548



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Figure 27: Comparison of predicted distributions of mass fractions of NC7H16, IC8H18, CH₂O, CO, and OH at TDC at different SoI timings [186] 1551

Figure 27 shows the mass fractions of NC7H16, IC8H18, CH2O, CO, and OH at the TDC at 1552 different SoI timings for iso-octane. In advanced SoI timings (SoI = -27° aTDC), the 1553 availability of more time for premixing resulted in relatively lower in-cylinder 1554 temperature and HRR, especially in the HCHR regions. This also led to greater cooling of 1555

1556 the spray, resulting in higher non-reacting n-heptane and iso-octane. Results also showed 1557 that more CH_2O and CO were produced in advanced SoI timings than the SoI = $3^{\circ}aTDC$ 1558 case. However, the regions of formation for these species were different in all cases. For $SoI = -27^{\circ}aTDC$ case, CH_2O and CO mainly formed in the large fuel regions; however, in 1559 SoI = -7° aTDC, CH₂O and CO mainly formed around the spray periphery. They also 1560 1561 reported the formation mechanism of these species and suggested that reactions C_7H_{15} + $O_2 \rightarrow C_7 H_{15} O_2$ and $AC_8 H_{17} + O_2 => AC_8 H_{17} O_2$ are important in the presence of higher n-1562 1563 heptane and iso-octane. This was mainly due to their close relationship with the low-1564 temperature reaction pathways. However, heat release at a higher temperature was dominated by the reaction $HCO + O_2 = CO + HO_2$, which laid the foundation of the intense 1565 1566 HTHR. The higher reactivity is located in the CH₂O-rich regions, which the authors 1567 establish a correlation to the whole mixture reactivity. The formation of CH_2O is dominated by $C_2H_4 + OH = CH_2O + CH_3$ in the high temperature region (with the delayed 1568 SOI scenario) in the earlier SOI cases the precursors are respectively CH₂OH for the SOI 1569 1570 = -27° aTCD and CH₃O for the SOI = -7° aTCD case. In all cases, it should be highlighted 1571 that the reaction of iso-octane enhances the formation of reacting species that accelerate 1572 the combustion process.

1573 In summary, this section focused on the fundamental investigations of RCCI combustion 1574 mode using optical diagnostics and simulations approach, explaining the combustion 1575 processes in totality, including the kinematic processes, control techniques, flame 1576 evolution, among other descriptive conditions during combustion. Several studies that 1577 implemented optical diagnostics techniques have been included, indicating that the flame 1578 propagation under RCCI combustion mode was highly sensitive to fuel stratification. 1579 Higher premixed ratios promoted higher flame propagation. It also emerged from the 1580 review that local fuel reactivity controls ignition sites, with high reactivity zones being more likely to be ignited first, e.g., downstream of injected HRF jets. Optical techniques 1581

1582 such as PLIF have some limitations in terms of dimensionality and definition of the 1583 combustion phases. Some researchers provided information about the spatial distribution 1584 of formaldehyde and OH radicals during RCCI combustion mode. Also, using optical diagnostics, the effects of the SoI on different stages of the RCCI combustion mode were 1585 1586 investigated, especially when the SoI was delayed more. In this case, the first stage 1587 autoignition occurred at the perimeter region of the cylinder (squish region), representing 1588 higher reactivity gradients. The second autoignition stage occurred in the combustion 1589 chamber's central region, representing lower reactivity gradients. Delayed SoI exhibited 1590 a slower rate of formaldehyde consumption and OH radical formation. A combination of 1591 optical and modeling techniques was also undertaken in some studies, which provided 1592 elaborate new information on the chemical kinetics of the RCCI combustion mode.

1593 2.5 Effect of Control Parameters on RCCI Combustion mode

1594 Several studies investigated various parameters to control the RCCI combustion mode, 1595 among which the LRF quantity was an important one. A few studies indicated that RCCI 1596 combustion mode with optimum LRF quantity complied with steady-state NOx and soot 1597 emission limits imposed by EURO-VI emission regulations without using the aftertreatment systems [151, 156, 294]. The LRF quantity must be low at low loads to increase 1598 the combustion stability and reduce the HC and CO emissions. At medium engine loads, 1599 a relatively higher LRF quantity could be used (which may reach the maximum levels); 1600 1601 however, a moderated LRF quantity should be used to control the maximum in-cylinder pressure and the PRR [295]. Wang et al. [296] investigated the RCCI combustion mode in 1602 1603 different premixed ratios of methanol and reported that high premixed ratios resulted in 1604 unstable combustion, especially at low engine loads. This led to misfire and significant 1605 cyclic variations, resulting in higher HC emissions and reduced fuel economy. Several 1606 interventions, including intake air heating, methanol heating, etc., were explored to avoid such events. Pan et al. [297] performed RCCI combustion mode investigations to explore 1607

1608 the potential of intake air temperature variations as a control parameter. They reported 1609 that increasing intake air temperature resulted in relatively superior combustion 1610 stability. RCCI combustion mode exhibited lower HC, CO, and formaldehyde emissions 1611 at higher intake air temperature, especially while using higher proportions of fumigated 1612 methanol. The overall objective of implementing such techniques was to enhance the 1613 methanol vaporization, which directly affected the degree of completion of combustion. It 1614 has already been demonstrated that mineral diesel fuel injection parameters play an 1615 important role in modern CRDI diesel engines. In a diesel engine, fuel injection 1616 parameters directly affect the spray characteristics. Fuel-droplet size distribution in the 1617 combustion chamber and fuel-air mixture homogeneity are critical for a diesel engine's 1618 good performance and emission characteristics. In diesel engines, pilot injection results 1619 in relatively lower peak HRR, leading to smoother combustion [298]. The pilot injection also results in relatively superior engine performance and lower HC emissions [298]. 1620 Therefore, the effect of fuel injection parameters of the HRF, namely FIP, the SoI timing, 1621 1622 and the number of injections, were also assessed to optimize the RCCI combustion mode 1623 for varying engine load. Liu et al. [299] conducted the DMDF combustion investigations 1624 using mineral diesel and methanol. They explored the effect of FIP of diesel on engine combustion, performance, and emission characteristics. They reported that increasing 1625 1626 FIP of mineral diesel resulted in relatively superior engine performance characteristics due to optimized combustion phasing (near TDC) and shorter combustion duration. 1627 Increasing FIP of mineral diesel led to relatively lower HC, CO, and smoke emissions; 1628 1629 however, NOx emissions increased slightly. Higher CO_2 emissions at higher FIP of 1630 mineral diesel were observed due to improved DMDF combustion. The other important 1631 finding of this study was a relatively more dominant response of varying FIP on the 1632 DMDF combustion mode than the CDC mode.

1633 Singh et al. [152] explored the FIP variations of mineral diesel (as HRF) on the RCCI 1634 combustion mode at different premixed ratios of methanol (as LRF). They reported that increasing FIP led to higher knocking in baseline CI combustion mode; however, RCCI 1635 combustion mode exhibited relatively superior engine performance, especially at higher 1636 1637 FIP and higher premixed ratios. Another important observation was a relatively weaker effect of FIP variations on the HC and CO emissions from the RCCI combustion mode. 1638 However, the PM emissions correlated strongly with the FIP. Due to greater penetration 1639 1640 of the HRF at higher FIPs, increasing FIP resulted in lower PM emissions from the RCCI combustion mode. They also analyzed experimental results to identify a suitable FIP for 1641 the RCCI combustion mode (Figure 28). 1642



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(u)



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- 1646

Figure 28: (a) TPM and TPN emissions, and (b) TPM and NOx emissions from RCCI
combustion mode at different premixed ratios of methanol and FIPs of mineral diesel
w.r.t. baseline CI combustion mode [152].

1650 This optimization exercise exhibited that the engine performance and particulate 1651 characteristics of RCCI combustion mode were significantly affected by the FIP of the 1652 HRF. Contour maps of TPN in figure 28 (a) showed a weak effect of FIP variations on the 1653 TPN emitted by both baseline CI combustion and RCCI combustion mode engines; 1654 however, the premixed ratio exhibited a relatively stronger correlation with the TPN, 1655 which decreased with increasing premixed ratio of methanol. Combined analysis of TPN and TPM contours exhibited that a combination of 750 bar FIP at $0.50 < r_p < 0.75$ of the 1656 LRF was the optimum range for the lowest TPN and TPM emitted by the RCCI 1657 1658 combustion mode. Figure 28(b) showed a strong correlation of NOx emissions with the premixed ratio of methanol; however, increasing FIP did not show a significant reduction 1659 in NOx emissions. Walker et al. [300] also explored the effect of FIP of mineral diesel on 1660 1661 RCCI combustion mode. They reported that increasing FIP of mineral diesel led to 1662 superior control over the combustion phasing. Li et al. [253] explored the effect of the SoI 1663 timings of mineral diesel on the RCCI combustion mode. They reported that advancing 1664 the SoI timings of mineral diesel resulted in relatively more stable combustion. At higher premixed ratios of methanol, advancing the SoI timings of mineral diesel led to superior 1665 1666 fuel economy. Mohammadian et al. [176] explored a single-fuel RCCI combustion mode strategy fueled with isobutanol + 20% DTPB. They performed a detailed investigation to 1667 explore the effect of different DI parameters, namely SoI timing, FIP, and spray cone 1668 1669 angle of the HRF and the premixed ratio of the LRF. They reported that SoI timing was an essential parameter in single-fuel RCCI combustion mode, which exhibited that 1670 advancing the SoI timing of the HRF (from 58° bTDC to 88° bTDC) resulted in superior 1671 1672 engine performance and reduced emissions than the baseline case (SoI timing= 58° 1673 bTDC). The pilot injection was the other important parameter for controlling the RCCI 1674 combustion mode. Results showed that pilot injection and main injection resulted in 1675 relatively higher in-cylinder temperature and pressure than a single injection, which 1676 promoted the combustion at lower engine loads and higher premixed ratios of the LRF. 1677 Suh et al. [301] reported that pilot injection provided favorable conditions for the main 1678 injection, leading to superior ignition. Due to these advantages of pilot injection, several researchers explored the optimum pilot fuel quantity w.r.t. main injection fuel quantity 1679 1680 because too much pilot injection quantity results in lower engine efficiency. Wei et al. 1681 [221] reported that increasing pilot fuel injection quantity resulted in relatively higher peak in-cylinder temperature. The use of pilot injection also changes the HRR pattern in 1682 1683 the RCCI combustion mode, which becomes bimodal for higher pilot fuel injection 1684 quantity. The HRR characteristics also depend on the premixed ratio of methanol. Increasing the premixed ratio of methanol changes the HRR pattern from bimodal to uni-1685 1686 modal for a constant pilot fuel injection quantity. Wei et al. [221] suggested that the 1687 charge cooling effect of methanol (due to higher latent heat of vaporization) was the main

reason for this trend, leading to dominant premixed phase combustion, resulting in a 1688 1689 single peak in the HRR curve. SoI timing of pilot injection was also explored by many 1690 researchers, who reported that advancing pilot injection timing resulted in higher peak in-cylinder temperature. Wei et al. [221] concluded that a relatively larger pilot fuel 1691 1692 injection quantity at advanced pilot injection timing was suitable for achieving higher fuel 1693 efficiency from the RCCI combustion mode at a higher premixed ratio of methanol (M50). 1694 Their study also reported that most emissions, including CO, HC, and unregulated 1695 species, decreased with increasing pilot fuel injection quantity and advancing pilot 1696 injection timings. Jia and Denbratt [172] explored the effect of different methanol 1697 injection strategies in RCCI combustion mode. They compared the performance and 1698 emission characteristics of RCCI combustion mode achieved by port injected methanol 1699 and direct injection of methanol. They reported that port injection of methanol resulted in lower CO and HC emissions along with higher BTE. They also varied different control 1700 parameters to check the suitability of direct injection of methanol and observed that port 1701 1702 injection of methanol was superior in most of the conditions. Zhao et al. [187] also explored the potential of direct injection of LRF in RCCI combustion mode by using a combination 1703 1704 of butanol and biodiesel. They performed the experiments using different biodiesel 1705 injection timing, butanol energy ratio and butanol injection pressure. They reported that 1706 direct injection of both LRF and HRF provides better control on combustion due to two-1707 stage heat release including premixed combustion of butanol and biodiesel in first phase 1708 and diffusion combustion of remaining biodiesel in the second phase. These two phases 1709 can be adjusted by controlling the injection parameters of biodiesel and butanol along 1710 with premixed ratio of butanol.

Several studies evaluated boosting and EGR to achieve RCCI combustion mode at higher
engine loads. Wang et al. [302] investigated biodiesel-gasoline-fueled RCCI combustion
mode using boosting, EGR, and late intake valve closing (LIVC) control and reported a

1714 significant improvement in the upper load limit of the RCCI combustion mode. However, 1715 at higher load operation, biodiesel-gasoline-fueled RCCI combustion mode exhibited 1716 inferior engine performance than mineral diesel-gasoline-fueled RCCI combustion mode. For improving RCCI combustion mode performance (lower torque output), the charge 1717 1718 dilution using air and EGR was also explored. Although EGR dilution is preferred for 1719 achieving RCCI combustion mode due to its simplicity, air dilution exhibited higher thermal efficiency. This was mainly due to a relatively higher specific heat ratio with air 1720 1721 dilution than the EGR dilution [235]. Wang et al. [235] performed RCCI combustion mode 1722 investigations using experiments and theoretical thermodynamic modeling to evaluate the effects of air and EGR dilutions at high load conditions on the ITEg. 1723



1724

1725 Figure 29: Experimentally measured ITEg plot as a function of EGR and intake

pressure [235].

1726

The authors reported that ITEg increased with decreasing EGR at a fixed intake air pressure of 240 kPa. The relatively stronger effect of EGR than the intake air pressure on ITEg was another important observation of this study, reflecting a more promising contribution of air dilution on ITEg improvement (figure 29). To better compare air and EGR dilution effects on ITEg, Wang et al. [235] used 'fuel-to-charge base equivalence ratio 1732 (Φ'),' which decreased with increasing EGR. Therefore, for maintaining a constant Φ' , Φ 1733 should be increased with increasing EGR. Figure 29 showed that the ITEg contours were 1734 closely related to air and EGR dilution at similar engine loads and combustion phasing. 1735 Results showed that ITEg increased with decreasing EGR; however, at a constant mass 1736 of air, ITEg decreases with increasing EGR dilution. They concluded that the thermal 1737 efficiency increased with increasing air dilution; however, thermal efficiency was 1738 hampered by the EGR dilution.

Olmeda et al. [142] also investigated the effects of mineral diesel's premixed ratio, EGR, and SoI sweep. They reported that both EGR and premixed ratio of gasoline affected the combustion efficiency significantly. Increasing the EGR beyond 20% at higher premixed ratios of gasoline (more than 70%) resulted in inferior combustion, leading to higher HC and CO emissions. Although varying SOI timing of mineral diesel provided insights into its effects on the heat losses, however, for SoI timing variations from 30 to 60° CA bTDC, the heat losses did not show significant variations.

1746 Desantes et al. [303] focused on the thermal efficiency of the RCCI combustion mode at 1747 lower engine loads. They performed simulations and experimental investigations to 1748 explore the effect of oxygen concentration on the RCCI combustion mode. They reported an improvement of $\sim 1.5\%$ in combustion efficiency using the combined effect of oxygen 1749 1750 concentration and in-cylinder fuel blending (ICFB). Bora et al. [274] performed the RCCI 1751 combustion mode investigations to assess the effect of compression ratio on the RCCI combustion mode and reported relatively superior engine combustion and performance; 1752 1753 however, NOx and CO_2 emissions increased with increasing compression ratio. This 1754 increase was due to improved combustion mainly, leading to higher peak in-cylinder temperature. Singh et al. [148] also explored the EGR and intake charge temperature 1755 1756 (ICT) as control parameters to effectively control the RCCI combustion mode.



1758Figure 30: Qualitative correlation between the combustion, performance, emissions, and1759particulate characteristics of RCCI combustion mode (at $r_p = 0.50$) at varying (a) EGR1760rates and (b) ICT [148].

1757

The authors performed experiments at different premixed ratios and reported that 1761 1762 increasing EGR resulted in relatively more stable combustion (Figure 30 a); however, at 1763 higher premixed ratios of methanol, combustion degraded due to the excessive cooling 1764 effect of the EGR. In contrast to EGR, the effects of ICT were significant at higher premixed ratios to reduce the in-cylinder cooling and charge cooling effects of EGR and 1765 methanol, respectively. They concluded that the combined effect of EGR and ICT 1766 variations could be an effective solution to achieve stable RCCI combustion mode along 1767 with lower HC and CO emissions (Figure 30). 1768

In summary, this section presented an overview of recent literature on the effect of control parameters on RCCI combustion mode. By the definition of RCCI, the main control parameter explored is the reactivity that defines this combustion mode; hence the quantities of LRF and HRF were extensively investigated. Some studies indicated that by optimizing the proportions of LRF to HRF, RCCI combustion mode engine should easily comply with steady-state EU-VI regulations for NOx and soot. It was revealed that the optimum premixed ratio was dependent on the engine load, being the highest possible at 1776 medium loads since at low loads, higher proportions of LRF may lead to higher CO and 1777 HC emissions or unstable combustion. In contrast, at high loads, it could lead to excessive 1778 maximum pressure and PRR. Some other combustion control strategies were explored to enhance the combustion control and prevent relying exclusively on the reactivity 1779 1780 gradients of the fuels. One such strategy was to increase the intake air's temperature, which resulted in higher combustion stability depending on the proportion of the LRF. 1781 Another commonly explored strategy was to optimize the fuel injection timing, the 1782 1783 number of injections, and the FIP of the HRF. It was concluded that for the RCCI combustion mode, higher FIP could be beneficial for improving the combustion stability 1784 and reducing the PM emissions, contrary to CDC mode. However, FIP didn't have a great 1785 1786 influence on CO and HC emissions. Advancing the SoI of the HRF also improved the 1787 combustion stability of RCCI combustion mode. Pilot injection controlled the RCCI combustion mode, especially at lower loads, increasing the in-cylinder temperature and 1788 pressure, allowing higher premixed ratios. Air management strategies studied by 1789 1790 researchers to control the combustion and emissions in RCCI combustion mode were EGR 1791 and boosting. Albeit ITEg reportedly increased with reducing EGR rate. EGR helped 1792 maintain more stable RCCI combustion mode, though.

3. Challenges and Limits of RCCI Combustion mode

1794 HCCI and PCCI combustion have several drawbacks. The main difficulties lie in the 1795 combustion control, high load operating range extension, and high CO and HC emissions 1796 at low load. Diesel (or HRFs) facilitates the auto-ignition and combustion under the PCCI mode at low loads; however, combustion phasing and excessive PRR pose a challenge at 1797 1798 high loads. The RCCI combustion mode adopts in-cylinder fuel mixing using a PFI to induct the LRF and DI to induct the HRF. An ideal fuel reactivity stratification can be 1799 obtained using this approach by varying the LRF/HRF proportions over a wide range of 1800 engine loads and speeds [304]. This concept was preceded by Inagaki et al. [104], who 1801

worked on PCI combustion. Gradual combustion progress was achieved by precise in-1802 1803 cylinder stratification, using a PFI system to supply the iso-octane and a DI system for 1804 diesel supply, and adjusting the mixture reactivity to meet the load requirements. This study effectively demonstrated that the operational map for the LTC concepts derived 1805 1806 from the HCCI could be extended for maintaining the soot and NOx at low levels and keeping the PRR under control. The PCI concept evolved into the RCCI concept after the 1807 advancements made by Kokjohn et al. [106, 123, 144], who coined this term and verified 1808 1809 the results by Inagaki. Their work indicated the domination of the combustion sequence 1810 by successive ignition of zones having different mixture reactivities, from the most 1811 reactive to the least reactive ones. It was established that controlling the fuel blend for 1812 spatial stratification of the fuel reactivity could control the combustion duration. Several 1813 researchers stressed that RCCI combustion mode could overcome many limitations of the HCCI and PCCI combustion mode [130, 305]. RCCI combustion mode, however, was not 1814 1815 immune to drawbacks. On its own, it has been extensively studied, and several reviews 1816 have been done on this topic [25, 306], which cover some of the drawbacks and potential 1817 of the RCCI combustion mode concept. However, a comprehensive review on overcoming 1818 the challenges and real-world applicability is unavailable in the open literature. The next section summarizes studies related to the challenges which must be addressed for the 1819 1820 commercial use of the RCCI combustion mode concept, proposed solutions of various 1821 challenges, few examples of real-world applications, and the prospects of this new concept.

1822 3.1 Challenges in Implementing RCCI Concept in Real Engines

There is general agreement in the scientific community about the main issues faced in the commercialization of RCCI combustion mode. These include higher CO and unburned HC emissions at low loads and excessive PRR at high loads. Other identified problems include slower flame propagation, knocking, and lower thermal efficiency at low loads, lower exhaust gas temperatures which reduce the efficiency of after-treatment systems, and

increased complexity for the fuel injection system and controls. Other issues include 1828 1829 higher specific fuel consumption, which is closely related to the RCCI mode engine's 1830 efficiency. However, the main issue lies in the operating range since both high and low 1831 loads cause undesired effects. Often, the strategies to extend the range in one direction 1832 negatively impact the other load extremes. Solutions proposed for the range issue are from 1833 hybrid combustion concepts that operate under one combustion mode, or others depending 1834 on the load demands, such as the dual-mode dual-fuel (DMDF) concept given by Benajes 1835 et al. [307], to the utilization of alternative fuels with attractive properties that could potentially offset the negative effects at low and high loads [306], and the implementation 1836 1837 of the so-called single-fuel RCCI, where through the addition of cetane improvers or 1838 reforming of the fuel onboard, a fuel derivate with different properties can be obtained. The problems associated with RCCI combustion mode are identified, and solutions are 1839 1840 discussed to develop this concept closer to reality.

1841 3.2 Engine Speed-Load Limits in RCCI Combustion mode

RCCI is an LTC that uses at least two fuels with different reactivities to control 1842 1843 combustion timing and phasing. This control is achieved by varying the ratio of the two 1844 fuels and adjusting their injection timing. This same principle also has undesirable side effects; for example, a high fraction of premixed combustion at high loads promotes 1845 excessive PRR, knock, and noise. The low combustion temperature at low loads does not 1846 1847 allow complete fuel-air mixture oxidation and increases the CO and HC emissions. 1848 Another unforeseen consequence of the advanced LTC concept is the low exhaust temperatures, at times lower than the lighting-off temperature of DOC and air 1849 1850 management system requirements. These requirements are almost impossible to achieve 1851 outside the test rigs, therefore requiring some complex system additions.

Defined operational limits for RCCI have been investigated [308]. They reported that on
a 17.1:1 CR, serial production 1.9 L engine platform at 1000 rpm, the combustion concept

limits were within 2-5 bar BMEP, while at 3000 rpm, the limits were from 4-8 bar BMEP. These limits respect a set of limitations such as smoke under 0.1 filter smoke number (FSN) and NOx below 0.4 g/kWh. However, the problem remains in extending the operational limit to the ranges of conventional CI and SI engines. For this reason, the main limiting factors are addressed in this review paper so that the focus can be objectiveoriented. These limiting factors of discussed in the following sub-sections.

1860 3.2.1 Intake Air Loop Requirements (EGR and Turbocharging)

1861 For resolving the issue of excessive HRR, the researchers have been prompted to increase 1862 the charge dilution, and for managing this, excess air and EGR are proposed quite often. 1863 Wu and Reitz [309] reported that combustion and emissions improved with a higher boost while simultaneously reducing EGR requirement and sensitivity to emissions in RCCI 1864 1865 combustion mode at high loads. High inlet pressures bring some implementation difficulties to the light. While a boosted engine is necessary to induct excess air, the 1866 existing turbochargers cannot always supply the required inlet air pressure and flow 1867 1868 rates. This can be resolved by using a large air management system that surpasses any 1869 commercially installed turbocharger capacity. For a real-world application, the design of 1870 a higher capacity air management system would be necessary or a significant improvement in the control of RCCI combustion mode to not require high air-flow rates. 1871

1872 On the other hand, EGR induces similar flow problems with an additional factor of 1873 temperature limitations; hence any new system must consider these constraints. EGR reduces the PRR; however, RCCI combustion mode is greatly sensitive to variations in 1874 1875 EGR rate, especially at high loads. Nevertheless, an early diesel injection can slightly 1876 reduce EGR sensitivity compared to a late diesel injection [309]. In this case, the early injection changes the charge composition, reducing the chances for auto-ignition of the 1877 1878 premixed charge of the LRF. It is worth mentioning that contradictory results have been reported for boosting and EGR for HCCI mode (and by extension to other LTC methods, 1879

1880 like RCCI mode). Some studies reported that the mixture dilution by Boost and EGR could 1881 help the combustion remain within peak pressure and PRR limits. In contrast, some other 1882 studies reported an increase in both these parameters. These contradictions have been 1883 noted [63]. The latter effect was attributed to reducing the heat transfer to the cylinder 1884 walls, increasing the combustion speed. It is, however, generally agreed that EGR extends 1885 the load limits for the LTC modes without breaching the imposed limits for peak pressure 1886 or the PRR.

1887 High EGR rates are necessary to extend the load range. Nonetheless, large quantities of 1888 EGR at low loads must be accompanied by increasing the HRF fraction. Low load dieselfueled LTC (IMEP ~0.23-0.26 MPa) has been investigated and compared with the diesel-1889 1890 gasoline RCCI strategy [310] on a single-cylinder heavy-duty engine. The results 1891 indicated that the proposed method was suitable for low load operations, albeit produced 1892 slightly higher NOx and soot than RCCI combustion mode. In Figure 31 a, an increase in EGR was necessary for the increased load, and the injection timings were more advanced 1893 1894 to prevent the LRF from generating knock. Figure 31 b shows the emissions and 1895 maximum PRR, which is of particular interest in this study. A plateau existed at medium 1896 loads and speeds for HC and CO emissions.





1898 Figure 31: Operational parameters and associated exhaust emissions corresponding to 1899 the maximum load conditions in RCCI mode at each engine speed investigated [310]. With the application of EGR in RCCI combustion mode, CO and HC emissions reportedly 1900 increased in another study [230]. However, with an early diesel injection, a slight 1901 1902 reduction in HC and CO was observed with a penalty of higher NOx emissions [309]. EGR 1903 rate is also highly dependent on the initial temperature [311]. If the initial temperature 1904 is high, EGR is essential to extract the NOx benefits and prevent undesirable autoignition 1905 of the LRF. EGR, however, has some effect on the ringing intensity of the engine when 1906 maintaining a constant combustion phasing. At different (higher) loads, only a small fraction of EGR would be required, further indicating that a fraction of LRF can control 1907 combustion [311]. RCCI combustion mode regime needs increased global reactivity of the 1908 in-cylinder charge (in the same way as CDC mode) to extend the low-load operating range. 1909 1910 However, this neither prevents higher CO and HC emissions nor higher fuel consumption that LTC strategies often suffer from [310]. A similar principle would be reviewed later to 1911

1912 extend the RCCI concept by incorporating multi-mode operation, where more good1913 combustion is obtained at each load and speed condition.

1914 High boost pressure requirement emerged as a limitation to realize the full potential of the RCCI combustion mode concept at high load conditions [312] because the LRF fraction 1915 1916 has to be reduced, and the HRF fraction has to be increased to cater to the necessary load 1917 conditions. A further study, however, demonstrated that this might not be as limiting as 1918 previously thought. At full load, the boosting and EGR rate requirements were similar to 1919 those required in the production-grade engine (3 bar boost and 30% EGR) [313]. Hanson 1920 et al. [314] demonstrated the effect of these properties on the RCCI combustion mode, 1921 while investigating the emissions and performance of a 13L multi-cylinder heavy-duty 1922 diesel engine modified for dual-fuel operation (PFI of natural gas and direct injection of 1923 diesel) both with and without the EGR over the EPA Heavy-Duty 13 mode supplemental emissions test. They confirmed that EGR reduced the engine noise to below 97 dB while 1924 simultaneously reducing NOx emissions by 48%, even though a slight increase of soot and 1925 1926 thermal efficiency were also observed. The objective of using EGR in this study was to 1927 achieve the lowest possible NOx without significant thermal efficiency loss. Hence, to 1928 control pumping losses and soot emissions, only a minimum required EGR was applied. EGR also reduced HC and CO emissions due to the engine's lower exhaust mass flow rate. 1929 1930 With EGR, inlet temperature was also higher, which helped oxidize cycle-averaged CO and HC. With EGR, 1.53 g/kWh of HC emissions were observed, while without EGR, they 1931 were 2.27 g/kWh. 1932

1933 3.2.2 Mechanical Limitations (Pressure Rise Rates and Noise)

High speed and high load conditions are problematic due to the mechanical limitations
related to the structural integrity of the engine. Higher PRR and peak pressures are often
a consequence of more efficient combustion, which heavily affect the engine noise
characteristics and lead to higher stresses on the engine components, reducing their

durability [315]. PRR is an important factor affecting engine knock and is an issue in SI engines. However, with premixed combustion in LTC modes, PRR can also affect the CI engines. It is generally accepted that high PRR occurs because of the auto-ignition of gasoline (in this case, the LRF) before the flame front initiated by the spark plug (or autoignition of the HRF) reached the combustible mixture. This, in turn, causes a ringing sound, originated by detonation waves. Prolonged knocking may cause high wear of piston rings, cylinder-head erosion, the disintegration of the piston, and piston melting.

1945 Hence PRR also limits the thermal efficiency achieved, besides undesirable noise [316]. 1946 High peak in-cylinder pressure is undesirable since it can exceed the safe operating 1947 pressure range, causing high stresses and fatigue in the engine components and adversely 1948 affect the engine. High HRR in RCCI combustion mode at high-speed, high-load 1949 conditions can cause ringing. Studies on charge stratification by Li et al. [317] concluded that peak PRR could be reduced by retarding the injection timing. Researchers [310] 1950 examined the operational range for RCCI combustion mode from 900 to 2500 rpm engine 1951 1952 speed and concluded that the engine load limit increases with the engine speed. Limiting factors for extending the high-load limit of RCCI combustion mode were excessive NOx 1953 1954 formation at low engine speeds (because the LRF fraction needs to be reduced), excessive 1955 PRR at moderate speeds, and unacceptable in-cylinder peak pressure at high speeds. The 1956 exploration campaign found a low fuel consumption of 168.6 g/kWh at 1900 rpm speed and a 56% EGR rate. The importance of boosting for extending the operational range of 1957 RCCI combustion mode at low speeds was assessed. It reduced fuel consumption but 1958 1959 generated excessive PRR. A high EGR rate and gasoline fraction were required at high 1960 speeds, and advanced diesel injection was required at low speeds to extend the RCCI combustion mode operating range. Figure 32 shows the PRR achieved with and without 1961 1962 the EGR [314]. The images make it clear how EGR aided in reducing problematic levels of PRR across the entire operating map, reaching levels >12 bar/deg only in the higher 1963

1964 load range. At the same absence of EGR promoted these values at medium loads for low



1965 and high engine speeds.



1967 Figure 32: MPRR for EGR (top) and non-EGR (bottom) operations over the entire engine
1968 operating map [314].

1969 3.2.3 Engine-Out HC and CO Emissions and Aftertreatment System Efficiency

One of the main issues for RCCI is the higher emissions of incomplete oxidation products, 1970 i.e., HC and CO. The injected and premixed LRF at advanced crank angles in the RCCI 1971 1972 mode can get trapped in the crevices [19], increasing the unburned HC emissions due to 1973 incomplete burning. This can happen particularly at lower engine loads because the 1974 combustion propagation is weaker due to insufficient fuel-air mixing. RCCI and other LTC 1975 modes suffer from reduced CO conversion, posing challenges to the after-treatment 1976 systems. Although RCCI can help avoid the need for SCR after-treatment system due to its very low NOx emission levels; however, the DPF and the DOC become important to 1977

1978 convert the higher quantities of CO and HC that remain unoxidized in the LTC modes. 1979 After-treatment systems need suitable boundary conditions to operate satisfactorily, and 1980 inlet temperature is one of those important boundary conditions. The exhaust gas temperature of the RCCI mode engine is the inlet temperature for the after-treatment 1981 1982 system. Obtaining adequate inlet temperature has been a challenge for efficient operation 1983 of the DOC, more so at low engine loads. Hence unburned emissions of HC and CO remain disproportionally higher. DPF regeneration also suffers from negative consequences 1984 1985 because of lower inlet air temperature, leading to inefficient passive regeneration. The 1986 lower soot and the NO₂ concentrations do not properly activate efficient passive 1987 regeneration. Therefore, active regeneration using higher oxygenation at higher 1988 temperatures is more effective in cleaning the DPF.

1989 Several attempts have been made to reduce engine-out HC and CO emissions before they 1990 reach the after-treatment devices. Among the options, direct injection of both the HRF 1991 and the LRF is thought to simplify the reactivity and equivalence ratio independently 1992 [318], controlling the fuel quantity in the crevice regions, reducing unburned HC. This 1993 strategy reduced higher NOx emissions than the traditional incorporation of LRF via PFI. 1994 However, direct injection of LRF did not show the desired reduction of unburned HC and CO emissions. Appropriate spray targeting and control of the crevice flow [19, 144] 1995 reduced the CO and HC emissions by reducing the localized fuel-rich zones. 1996

Higher CO and HC emissions in RCCI mode might not impede commercial implementation of this concept [318-319]. The oxidation of CO and HC can be achieved by using conventional DOCs with RCCI mode operation, as long as the exhaust gas temperatures remain >200°C to ensure catalytic activity. Advancing the evaluation of the performance of DOCs with RCCI mode, Garcia et al. [320] developed and calibrated a 1-D model for the DOC with RCCI mode to define the device's size to comply with current emission standards. They analyzed the response of DOC in a vehicle system simulation 2004 under different driving cycles to find that the CO and HC emissions at the DOC outlet 2005 surpass the desired range. The researchers then sized the device to achieve an acceptable 2006 emission level. They concluded that a volume four to six times bigger would be necessary 2007 to comply with the prevailing emission standards. It can be summarized from these 2008 studies that although DOC reduces exhaust emissions, to comply with current and future 2009 emissions regulations, resizing the DOC is necessary along with fine-tuning the 2010 combustion to reduce engine-out CO and HC emissions.

2011 3.2.4 Transient Cycles and Control Systems

2012 Vehicle operation and, consequently, engine operation are not restricted to stationary 2013 conditions. The complexity of transient RCCI mode operation was explored by Gross and 2014 Reitz [321]. They indicated an expressed need for additional controls to avoid undesirable 2015 effects to achieve the RCCI mode. A comprehensive review of the transient operation under RCCI mode was presented by Paykani et al. [322]. The main theme was that the 2016 2017 RCCI concept was mostly tested under stationary conditions, and there could be 2018 differences under transient conditions. They showed how open-loop (OL) control systems 2019 based on maps could be more expensive to calibrate than the close-loop (CL) control based 2020 on the in-cylinder pressure signals. The authors referred to Saracino et al. [323] and 2021 Hanson [324] to suggest that CL systems can account for the variability in exhaust gas 2022 temperature and EGR among the cylinders. The literature indicated that transient operation in RCCI mode is a real possibility by adjusting the EGR, airflow rate, engine 2023 speeds, intake pressure, and pedal position for a wide range of engine operating 2024 2025 conditions. Control systems are vital for implementing the RCCI combustion concept. 2026 Controllers have demonstrated an accurate tracking performance for desired combustion phasing. The study focused on a single-fuel RCCI using gasoline and a cetane improver to 2027 2028 increase the reactivity of the directly injected gasoline [321]. They performed a step change of load from 1 to 4 bar BMEP at 1500 rpm. Before transient operation 2029

2030 investigations, experiments at four steady-state points helped assess engine performance 2031 and emissions. Intermediate points were interpolated to improve smooth transitions in 2032 the instantaneous step changes. These points were then calibrated by changing the 2033 injection strategy, EGR, and fuel rail pressure to reach a predefined combustion phasing 2034 (CA₅₀). A CL calibration was used for these tests by employing a next-cycle (NC) controller to adjust the PFI fraction of each cycle to obtain the preset CA₅₀ values, while engine 2035 2036 parameters were adjusted following the 2-D maps. Results indicated that the single-fuel 2037 transient operation is possible without significantly increasing the emissions.

2038 In summary, RCCI mode emerged as an important LTC mode to resolve the issues posed by methods such as HCCI and PCCI. RCCI mode indicated that an ideal charge reactivity 2039 2040 could be obtained by varying the HRF and LRF quantities. However, researchers also experienced some limitations that need to be resolved. This section reflects the challenges 2041 in adopting RCCI combustion mode for commercial applications. From the emissions 2042 2043 perspective, it is generally agreed that the main shortcoming of RCCI combustion mode 2044 is relatively higher CO and HC emissions. Charge dilution strategies such as deploying 2045 higher EGR to increase the operational limits and prevent other problems can be useful 2046 if exhaust gas temperatures are not sufficiently high. Other factors limit the RCCI 2047 combustion mode, such as knocking probability at high loads and high premixed ratios, 2048 low thermal efficiency at low loads, and unstable combustion. Low exhaust temperatures 2049 are the other effect of RCCI combustion mode, which remains to be addressed. Low 2050 exhaust gas temperatures can hamper the after-treatment system operations, such as for 2051 DOCs. With these main issues in mind, appropriate solutions must be devised. These 2052 solutions include superior spray targeting and control of crevice flows, which reportedly reduce HC and CO emissions. Several studies on the after-treatment system capacity 2053 2054 have been undertaken to assess whether higher CO and HC emissions are a limiting 2055 factor for the commercial application of RCCI mode. These studies considered reduced

exhaust gas temperatures of the RCCI mode engines to find whether the after-treatment
systems could efficiently operate within these limitations. Finally, the control systems
must be more robust for operations under transient conditions due to relatively higher
injection system complexity for the RCCI mode engines than the CDC mode engines.
Experimental studies have conclusively shown that though RCCI mode operations might
be more complex, they will soon be a real commercial possibility.

4. Implementation of RCCI Combustion mode in Real Engines

2063 Most studies on RCCI mode address the combustion control issues in reactivity because 2064 it is the core of the working principle of this advanced combustion mode. Li et al. [325] 2065 classified the reactivity in two types: (i) global reactivity and (ii) reactivity gradient or 2066 stratification. The first was determined by the quantity and characteristics of the fuel 2067 (cetane number, octane number, LHV, etc.), while the second is dependent on the injection 2068 strategy, spray penetration, and entrainment of the HRF in the premixed charge. Kokjohn et al. [135] found that reactivity stratification is a leading factor for controlling the 2069 combustion phasing and ignition location, followed by equivalence ratio having a 2070 2071 significant influence. At the same time, the temperature stratification effect was 2072 negligible. Because of the importance of the reactivities of the fuels in combustion 2073 characteristics of the RCCI mode, their management, injection strategies, control, and 2074 concept extensions are worthy of investigations for engine implementation. For the RCCI 2075 combustion concept to work, at least two fuels with different reactivity are required. The 2076 source for these can vary from the use of a single-fuel to multiple fuels. The need for different fuels in the same engine/ vehicle platform increases control system complexity 2077 2078 and cost. There are hurdles in adapting current generation engines to the RCCI mode or designing clean sheet RCCI engines, but this concept has become feasible for 2079 2080 commercialization. Hanson et al. [326-327] demonstrated the RCCI concept using gasoline 2081 and ultra-low sulfur diesel in a hybrid platform on a 2009 Saturn Vue vehicle. Besides

2082 this, one engine control system supplier has a commercially available vessel that affirms 2083 the RCCI mode retrofitting capabilities [328]. Recently, Argonon [329] showed promising 2084 commercial application possibilities of the RCCI combustion concept in large size engines by retrofitting a Caterpillar 3512 engine on an inland vessel MTS, which reduced up to 2085 2086 10% energy consumption when operated with biofuel instead of diesel while also 2087 complying with emission regulations without the need for DPF. The vessel's engine took 2088 advantage of the fuel flexibility of the RCCI concept to refuel without completely emptying 2089 the tank. This implied that it is possible to have a commercially available RCCI engine 2090 that relies on the fuel flexibility of the RCCI concept for its use. However, it is important 2091 to explore possible solutions for the downsides of the RCCI concept using the current 2092 state-of-the-art technologies available.

2093 4.1 Multi-Mode Concepts to Cover the Entire Engine Map

2094 RCCI concept has limitations over the load extremes, both high and low. Hence 2095 researchers have focused their efforts on finding a solution for these limitations without sacrificing benefits (such as reduced NOx and PM emissions) or further deteriorating the 2096 2097 weaker aspects (such as higher HC and CO emissions). Another boundary condition of 2098 RCCI combustion mode limits the peak pressure or PRR. The multi-mode concept offers 2099 an alternate solution to this issue. The multi-mode concept limits RCCI mode operation 2100 to an engine operating range where the combustion is optimized. Then, the engine 2101 transitions to another mode, which would offer superior performance at other engine operating conditions. Not only have that, mode-switching RCCI engines retained the 2102 2103 advantage of lower NO_x emissions compared to the CDC.

2104 4.1.1 Dual-Mode Dual-Fuel (DMDF) Concept

The dual-mode dual-fuel (DMDF) combustion mode extends the RCCI mode over the high and low loads. Different load zones are then catered by different combustion modes depending on constraints imposed by the PRR, NOx, soot, and maximum in-cylinder

2108 pressure. These modes could be fully premixed combustion and dual-fuel diffusion 2109 combustion modes. PRR and in-cylinder pressure limitations are imposed to prevent 2110 mechanical failures. The optimized multi-mode combustion strategy combines fully and 2111 highly premixed RCCI regime at low and medium loads and dual-fuel diffusion 2112 combustion at full load [155-156]. In the cited studies, the authors indicated that this strategy allows maintaining PRR below 15 bar/CAD and maximum in-cylinder pressure 2113 2114 below 190 bar while simultaneously covering the entire engine map with engine-out NOx 2115 emissions below EURO VI limits [155]. The authors could reach up to 14 bar IMEP and 2116 maintain soot emission under 0.8 FSN in most engine operational zones, reaching values 2117 as low as 0.02 FSN at 7 bar IMEP. DMDF combustion relies on reducing the CR to resolve 2118 the limitations of the RCCI mode at full load [330]. The reduced CR then helps mitigate 2119 the undesirable ignition of LRF and reduce the PRR. Benajes et al. [330] emphasized extending the RCCI mode because of the emission improvements in the entire global 2120 engine map. To do that, they evaluated the RCCI/CDC mode-switching vis-à-vis CDC, 2121 2122 depending on the coverage of the RCCI combustion mode regime, while trying to maximize its share to complete the Real Driving Emission cycle. Garcia et al. [294] compared the 2123 2124 DMDF combustion to the RCCI/CDC mode-switching combustion to cover the 2125 unattainable load range of RCCI with CDC. They reported that DMDF reduced specific 2126 fuel consumption by 7%, and engine-out NOx emissions complied with EURO VI limits and were 87% lower than the RCCI/CDC mode. DMDF still showed higher CO and HC 2127 2128 emissions (up to 10 times higher than other modes) which could be addressed by using 2129 exhaust gas after-treatment systems.

2130

4.1.2 RCCI/ CDC Mode Switching

Benajes et al. [304] explained that the dual-fuel RCCI/CDC concept hinges upon switching 2131 2132 between the RCCI and CDC modes to cover the entire engine map. They evaluated the 2133 performance and emissions of this concept by simulating vehicle systems operating under

2134 different driving cycles (Real Driving Emissions (RDE) Cycle, Worldwide Harmonized 2135 Light Vehicle Test Cycle (WHTC), Federal Test Procedure (FTP-75), and Japanese cycle 2136 (JC08)), with the experimentally obtained diesel-E85 and diesel-gasoline engine maps on a light-duty diesel engine having a CR of 17.1. Their results indicated that this concept 2137 2138 could be used in flexible fuel vehicles (FFV). They concluded that E85 as LRF could extend the operating limits of the RCCI mode with lower NOx and soot emissions but with higher 2139 2140 HC and CO emissions. Another interesting conclusion of the real-world application of this 2141 concept was that the amount of fuel necessary for dual-mode RCCI mode using gasoline was almost the same as required for the CDC mode. Hence no additional fuel storage 2142 2143 space would be required in the vehicle. A 32.5 L tank for diesel and 27.5 L tank for gasoline 2144 would be sufficient while maintaining the same vehicle range as the CDC. Prikhodko et 2145 al. [319] and Benajes et al. [331] indicated dual-mode RCCI mode as an alternative combustion mode (CDC in this case) without the need to reduce the CR when the RCCI 2146 2147 mode operating window was rather limited.

2148 Benajes et al. [313] indicated that the operating range of RCCI mode was between 25% 2149 and 35% load because of the limits imposed by PRR and peak in-cylinder pressure. The 2150 rest of the engine operating conditions in the engine map were catered by CDC mode, improving the overall oxidation of HC and CO, however maintaining their peak values at 2151 2152 \sim 37 g/kWh and 23 g/kWh, respectively. It is worth noting how the CO and HC emissions are superior to the results produced by a higher compression ratio engine [312]. The 2153 investigations were conducted in a single-cylinder engine using a gasoline-ethanol blend 2154 2155 (80%-20%) as LRF and diesel with 7% biodiesel as HRF. The RCCI operational range 2156 provided a 2% improvement in gross indicated efficiency. Very low NOx and soot emissions from RCCI complied with EURO VI emission norms, as shown in Figure 33. 2157



Figure 33: NOx and soot emissions mapping of RCCI mode operation on a high CR
EURO VI engine [313].

2158

Recent works on LTC mode switching explore how to cover the entire engine map by 2161 2162 varying between CDC, conventional dual-fuel combustion, HCCI, RCCI, PCCI, and 2163 piston-split dual-fuel combustion (PDFC) [332]. The authors divide the operation modes 2164 according to the proportion of LRF and HRF, as well as the timing of the injections of the HRF to early or advance CAD, as can be seen in Figure 34. Test and driving cycle 2165 simulation results indicate that selecting the correct mode for each zone of the operational 2166 2167 engine map can decrease NOx and soot emissions by around 20% when compared to CDC, 2168 and, as the thermal efficiency is increase, also provide a reduction of near 1% of CO₂ emissions. It should be understood that since the multi-mode concept uses the CDC 2169 approach to expand the operating map of the RCCI mode engine, the SCR and DPF cannot 2170 2171 be eliminated from the after-treatment system of the engine. Finally, the RCCI mode

- 2172 engine emerged as a very attractive concept exhibiting excellent efficiency and reduced
- 2173 fuel consumption.



2174

Figure 34: The Manifold/Early DI/Late DI Triangle diagram or 'MELT" diagram plots
the distribution of the total fuel energy into the three primary injection types considered
(a) shows isolines of fuel energy percentage, while (b) applies acronyms from the

2178 literature of combustion modes that are expected to occur with certain fuel distributions.

2179 4.1.3 Hybrid RCCI Mode Coupling

Electric vehicles are supposedly important in the decarbonization of the transport sector
and reducing urban air pollution. Light-duty vehicles have been substituted by plug-in
electric vehicles (PHEV) without significantly impacting the end-user experience. New

2183 developments in batteries and energy administration controls have increased the range 2184 to a market average of 315 km [333]. The availability of publicly accessible charging 2185 stations increased by 60% in 2019 compared to the previous year [334], and energy costs have become comparable to conventional vehicles [335]. Even though electric vehicles are 2186 2187 gaining relevance in the transport sector [335], IC engines would remain the most important workhorses for the transport sector globally in the near and medium-term 2188 2189 future. IC engines have not yet encountered a feasible electric powertrain challenge, 2190 which can fulfill the distance-range and cargo-weight demands of the medium-and-heavy-2191 duty vehicle segments while maintaining operational and logistics costs. As more electric 2192 vehicles penetrate the automotive fleet, an intermediate step between full electrification 2193 and straight IC engines is required. Hybrid Electric Vehicles can serve as a bridge between both realms, the EVs and the ICEs, and offer a superior solution, taking 2194 advantage of the benefits of both. A hybrid RCCI-electric powertrain is highly attractive 2195 2196 because of its potential to get emissions to regulation levels and even lower while also 2197 reaping benefits of lower fuel consumption. Additionally, the electric operation can serve as a primary power source in the load range where the RCCI has trouble. 2198

2199 Hybrid powertrains would allow RCCI mode operation with its load constraints, as demonstrated by Solouk and Shahbakhti [336]. They designed and implemented three 2200 2201 energy management control (EMC) strategies, namely rule-based control (RBC), dynamic programming (DP), and model-predictive control (MPC), to improve the fuel economy of 2202 the hybrid system combined over a combined driving cycle entailing the UDDS (urban 2203 2204 dynamometer driving schedule), HWFET (highway fuel-economy test), and US06 driving 2205 cycles (Figure 35) [336]. The investigators considered switching modes between the electric operation and combustion leads to a fuel penalty, adding this insight into their 2206 2207 results. The work was focused on fuel optimization, and it was observed how the RCCIseries hybrid electric vehicle (SHEV) exhibited superior fuel economy than a modern SI 2208

engine (12.6%) and a CI engine-based HEV (2.2%). Results from this work also discussed the state of charge (SOC) influence on the fuel economy. A battery at low SOC in charge sustaining mode has a fuel economy advantage over a higher initial SOC. The authors indicated that the battery at low SOC demands the engine to run longer to compensate for the higher battery energy loss, providing the RCCI mode with instances to save fuel that the tested SI and CI modes do not offer.



2215

Figure 35. The combined driving cycle for evaluating the designed EMC [336]. 2216 2217 Advancements in hybrid powertrains combined with dual-fuel RCCI mode were explored by Benajes et al. [337]. They evaluated Mild hybrid (MHEV), Full hybrid (FHEV), and 2218 2219 Plug-in hybrid (PHEV) vehicles with diesel-gasoline RCCI combustion mode using 0-D 2220 numerical modeling. The simulation inputs were based around the calibration maps of a 2221 CDC mode and diesel-gasoline dual-fuel RCCI mode of a GM 1.9L light-duty engine. The simulated vehicle consisted of a Class D passenger car, which had an additional electric 2222 2223 motor and battery in the model since the vehicle was not originally an electric vehicle. 2224 Hybrid powertrain results were compared to their non-hybrid counterparts under the World Harmonized Light Vehicles Test Procedure (WLTP). They demonstrated that it is 2225 2226 possible to attain NOx and soot levels below the EURO VI regulation. They achieved a 5% 2227 reduction in fuel consumption for only dual-fuel combustion mode while maintaining the 2228 NOx levels same as CDC mode. With the same fuel consumption that the OEM has, a 30% 2229 reduction in NOx emissions was obtained (Figure 36). The dual-fuel combustion increased CO emission to 1.6 g/km compared to 0.8 g/km from the CDC mode. It was possible to obtain similar emissions from a hybrid powertrain as that of the OEM because of the use of high BMEP for recharging the batteries, thus promoting CO conversion. HC emissions did not improve by using a hybrid powertrain and exhibited values higher than those reported by the OEM. Benajes et al. [337] performed a lifecycle analysis (LCA) for these systems. They indicated that the RCCI mode-based PHEV reduced CO₂emissions by 30% compared to the baseline CDC mode vehicle in the cradle-to-grave approach.





Figure 36. (a) NOx emissions for the CDC mode calibration map and (b) reactivity-

2239

controlled compression ignition (RCCI) gasoline calibration map [337].





2240

Figure 37. UW hybrid vehicle drivetrain [337].
Hanson et al. [326-327] tested a series-hybrid vehicle with an RCCI engine coupled to a 2242 2243 90 kW AC motor that worked as a generator for a 14.1 kW-hr Li-ion traction battery pack powering a 75 kW drive motor (Figure 37). They reported that HC, CO, and NOx 2244 emissions were similar to those reported in previous laboratory tests. With appropriate 2245 2246 modifications, the vehicle could comply with US EPA Tier-2 bin-5 NOx and CO levels for several test cycles with a fuel economy ranging from 4.4 L/100 km to 7.47 L/100 km. 2247 Additionally, they reported that exhaust gas after-treatment systems could reduce HC 2248 2249 and CO emissions by 98.5% in the HWFET with a hot start. During the ORNL HWFET, 2250 the NOx emissions were higher than the steady-state laboratory tests because of a 2251 probable unoptimized fuel injection calibration. Fuel consumption reduced to 0.3 L/km 2252 when operating at higher engine power demands because of the increased thermal 2253 efficiency it represented.

Solouk et al. [338] experimentally coupled different LTC modes with an Extended Range 2254 2255 Electric Vehicle (EREV). The EREV allowed the decoupling of the engine from the 2256 drivetrain, thus making the LTC operation achievable only in its optimum limits. Their experimental setup allowed the use of HCCI, RCCI, and conventional SI modes. They used 2257 2258 the data acquired to generate a brake-specific fuel consumption (BSFC) map and 2259 determine the load limits for each combustion mode (Figure 38). Tests were performed for 2260 single-mode EREV coupling (HCCI-EREV, RCCI-EREV, and SI-EREV) and a multi-mode LTC-EREV. They reported 9% and 10.3% improvements in fuel economy in a single-mode 2261 RCCI-EREV compared to SI-EREV coupling for the city driving cycle (UDDS) and the 2262 highway driving cycle (HWFET), respectively. However, with HCCI-EREV, the 2263 2264 improvement was higher (12% and 13.1%). The improvement over one driving cycle to the other was explained by the longer engine operation duration in the high-power demand 2265 2266 cycle (HWFET). There were no reported noise vibration harshness (NVH) or ringing issues, primarily because the LTC modes (RCCI and HCCI) were kept within the low 2267

2268 engine speed range where these issues are not prominent. The LTC modes exhibited 2269 improvement over the SI mode since the LTC modes at low vehicle speeds could operate at low engine speeds while the SI mode has to do so at high engine speeds. Another 2270 conclusion from this study was that the LTC-EREV multi-mode operation could improve 2271 2272 the fuel economy in the vehicle more than the single-mode operation could. Thus, the RCCI+SI-EREV emerged as the best combination in the high-power demand driving 2273 cycles, and HCCI+RCCI emerged as the best in the low to mid-power range driving cycles 2274 2275 (Figure 39).



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2277

Figure 38: BSFC map of multi-mode LTC-SI engine [338].



2278

2279 Figure 39: LTC-Hybrid electric powertrain experimental test-bed utilizing a double-

ended 465 hp AC dynamometer at Michigan Technological University [338].

2281 4.2 Single-Fuel RCCI Combustion mode

Functionally, the concept of RCCI with two different fuels can have substantial 2282 2283 advantages in obtaining NOx and soot emissions below the regulatory limits; however, 2284 operationally, the use of two fuels can bring in some complications. These include: (i) need for two fuel tanks in the vehicle, (ii) different depletion and recharging rates for the fuels, 2285 2286 which could potentially deter users from adopting the mode due to slightly increased fuel 2287 recharge scheduling complications, (iii) increased complexity of fuel injection control 2288 systems, as well as the use of two separate fuel injection systems, which would increase 2289 the cost of the vehicle. For these reasons, single-fuel RCCI mode was explored as a viable 2290 alternative. There are several ways to realize single-fuel RCCI mode. The main 2291 approaches for this include use of fuel additives and reforming. Both methods seek 2292 changes in the fuel composition and reactivity of the primary fuel so that the derivate fuel 2293 can act as a second fuel to manage the reactivity stratification. These approaches are 2294 discussed in the following sub-sections.

2295 4.2.1 Single-fuel + Additives

2296 To apply the RCCI combustion mode concept with a single-fuel tank, a method to change 2297 the reactivity of the fuel to allow reactivity stratification can be used. Cetane improvers 2298 are the most common path to achieve single-fuel RCCI mode. These additives could be DTBP and 2-EHN [285]. Cetane improvers are applied to a portion of the LRF, which is 2299 2300 then directly injected into the cylinder as the HRF. The rest of the LRF is injected through 2301 the PFI system [131, 133]. Many studies have been done on single-fuel RCCI mode engines 2302 using cetane improvers [76, 122, 178]. Single-fuel RCCI mode could be accomplished by 2303 using only the LRF (e.g., gasoline, methanol, ethanol, iso-butanol) with a cetane improver, 2304 which enhanced autoignition characteristics of the LRF. The required fractions of cetane improver were reportedly quite low, ranging between 0.75% and 5% v/v. However, the 2305 2306 amount of secondary fuel (the HRF obtained by the mixture of LRF and additives) will 2307 vary significantly from application to application. In the case of iso-butanol with DTBP,

2308 high concentrations of DTBP were required to raise the reactivity sufficiently [178]. 2309 Higher quantities of DI fuel were required (60-70%), compared to relatively lower 2310 quantities of diesel required for similar conditions (25-40%). Splitter et al. [133] tested DTBP with gasoline at mid-load conditions and used relatively smaller quantities of 2311 2312 improver (0.75%, 1.75%, and 3.5% v/v). The same study reported that a lower fuel injection pressure (400 bar) for the HRF was necessary with gasoline-DTBP than a diesel 2313 counterpart (800 bar). Peak gross indicated efficiency of 57% was obtained with acceptable 2314 emissions in this study. 2315



Figure 40: Examples of dual-fuel and single-fuel injection strategies for RCCI mode[131].

2316

2319 It was reported by Liotta [340] that a2-EHN fraction increase caused a linear increase in 2320 NOx emissions from the engine. The work states the response was due to the presence of 2321 the nitrate group chemically bonded to the additive. To realize ultra-low NOx emissions 2322 from conventional RCCI engines, Kaddatz et al. [180] used a light-duty single-cylinder 2323 engine with 10% ethanol-blended gasoline (E10) and 2-EHN as an additive (up to 3%v/v) 2324 in RCCI mode. They then compared the results with dual-fuel RCCI combustion using E10 and diesel. Since the percentage of 2-EHN in this study was low, the NOx penalty 2325 was not very high due to single-fuel and additive use. NOx penalty was below 1 g/kWh 2326 2327 over a range of loads. In addition, the best peak indicated efficiency was quite similar to that of dual-fuel RCCI mode at 50% load (9 bar gross IMEP). Single-fuel RCCI was also 2328

2329 investigated under transient conditions in a General Motors (GM) Z19DTH 1.9-L diesel 2330 engine having a CR of 13.75 [321]. Gasoline mixed with 2-EHN (3% v/v) was compared to 2331 conventional dual-fuel RCCI combustion mode under transient conditions. HC emissions were maintained under 1500 ppm, NO below 10 ppm, and smoke lower than 0.1 FSN 2332 2333 while simultaneously keeping a 10 bar/CAD limit on the PRR and a moderate maximum noise up to 95 dB during calibration. The researchers proved that single-fuel RCCI mode 2334 operation could be achieved with similar HC levels to the dual-fuel RCCI mode 2335 2336 counterpart, but with NOx penalty, while using gasoline with 2-EHN as the HRF.

2337 Regarding cetane improvers, one aspect that should be highlighted is the possible effects 2338 changing the fuel reactivity could cause into the storage stability and safety of the 2339 mixtures. Cetane improvers have been included into fuels from as far as 1940 [341]. More 2340 than two decades ago, the use of these peroxides with dual-fuel applications that included liquefied petroleum gas (LPG) and diesel [342] was tested finding that the thermal 2341 2342 stability of the LPG-diesel CI engine configuration operating with 5% was comparable 2343 with an engine running exclusively diesel fuel. In a posterior work, Sugiyama et al. 2344 produced a prototype truck operating with an LPG and 1% in weight cetane enhancer and 2345 lubricity improver fuel blend and found that the system could reach about 70000 km without major failures, establishing a long precedent of the use of such additives. It is 2346 2347 important to comment that cetane improvers usually are reactive and exothermically 2348 unstable peroxides, which in large concentrations it is conceivable that some of these properties can be transferred to the fuel with which is blended, nonetheless for 2349 2350 applications of RCCI concentrations of these composites remain relatively low and no 2351 significant effect reports were found regarding the stability of the fuel. The work of Eng et al. [343] tested concentrations as high as 15% v/v finding that the addition of larger 2352 2353 concentrations (above 2%) does not produce large changes in ignition timing in the fuel.

2354 4.2.2 Single-fuel + Reformate

2355 Although cetane improvers theoretically require only one fuel tank for the RCCI mode to 2356 work, the single-fuel alternative does require an additional tank of a similar size as that 2357 of a diesel exhaust fluid (DEF) [322], which would be required to be recharged periodically. The fuel reformation strategy to obtain a secondary fuel having different reactivity from 2358 2359 the same fuel to achieve RCCI combustion mode has been proposed to bypass this hurdle completely. The concept takes a fraction of the primary fuel and directs it to an onboard 2360 2361 fuel reformer to produce a reformed gaseous mixture of partially reacted hydrocarbon 2362 species, hydrogen, and CO [344]. Catalytic partial oxidation (CPOX) is a reforming, where 2363 a rich fuel-air mixture is reacted over a catalyst to produce CO, H_2 , and other partially 2364 combusted hydrocarbon species [345]. Another reforming alternative is steam reforming 2365 (SR or SMR for the specific case of methane). This option requires a high-temperature 2366 heat source, steam, and fuel (methane), which are reacted over a catalyst to produce CO and H₂ [344]. SMR and CPOX are different. The first is endothermic, while the second is 2367 exothermic. Variable valve actuation can also lead to in-cylinder reforming by using 2368 2369 negative valve overlap (NVO). The NVO principle increases the auto-ignition capabilities 2370 of high-octane fuels at a CR typical of SI engines without the need to preheat the air [346] 2371 because the exhaust mass is compressed. This concept is useful for on-board fuel reforming because the hot recompressed exhaust provides the necessary conditions to 2372 2373 initiate fuel reformation reactions gas [347]. The last reforming method mentioned in this review is the thermo-chemical fuel reformer (TFR) from dedicated EGR (D-EGR). D-EGR 2374 2375 has a dedicated cylinder running stoichiometrically; whose exhaust gases are supplied 2376 directly into the intake manifold [291]. It can be concluded from various studies that 2377 single-fuel RCCI mode is feasible by using onboard fuel reforming. However, the addition 2378 of hardware elements increases the complexity of the fuel and control systems.

2379 4.3 Dual Direct Injection (DDI)

Research shows that the most common RCCI configuration is using an engine with a DI 2380 2381 injector in addition to a PFI. However, other configurations are also proposed, e.g., the 2382 dual direct injection (DDI) configuration. Unlike the DI/PFI configuration, the DDI involves direct injections for the LRF and the HRF at different injection timings. The 2383 2384 intention behind direct injection of the LRF is to avoid fuel entrapment in the crevice region, which would reduce CO and HC emissions [339]. Lim and Reitz [349] performed 2385 2386 RCCI mode engine simulations using direct injection of iso-octane using a gasoline direct 2387 injection (GDI) injector. The LRF injected directly into the cylinder reduced CO and HC emissions by 27.1% and 7.1%, respectively, increasing combustion efficiency. DDI 2388 2389 enhances the control over the mixing process and combustion phasing. It provides greater 2390 flexibility in reactivity stratification due to the possibility of distributing the LRF across multiple injections throughout the compression stroke. Some of the previously mentioned 2391 noise issues can be mitigated by this strategy while reducing the total unburned 2392 hydrocarbon emissions by up to 91% [350]. The direct dual-fuel stratification (DDFS) 2393 2394 strategy reduced the combustion noise and improved combustion stability with lesser 2395 EGR required than conventional RCCI mode, with a combustion phasing near TDC [351]. 2396 Direct injection of the LRF and HRF stratifies the fuel directly in the engine cylinder and reduces the charge premixing to maintain it under desired limits [352]. This approach is 2397 2398 quite different from GCI combustion since RCCI mode has superior control over the 2399 combustion phasing. Besides this advantage, higher loads (up to 21 bar IMEP) were attained in the simulation of DDI of the RCCI mode using iso-octane and n-heptane [353]. 2400 2401 An ITEg of 48.7% was obtained. On the other hand, n-heptane (as the HRF) mass and 2402 injection timing were reported to have a larger effect on the injection control by allocating a smaller quantity of HRF in the squish region before introducing the rest of the HRF to 2403 2404 promote the ignition delay. Some drawbacks of the DDI are the need for higher injection 2405 pressure for both injectors, which adds complexity to the fuel injection and control

2406 systems. Additional space is also required in the cylinder-head for mounting both the 2407 injectors. Yang et al. [354] proposed a low-pressure dual-fuel direct injection (LPDDI) 2408 concept based on air-assisted direct injection (AADI) technology (Figure 39). The AADI 2409 consists of a built-up nozzle that incorporates a liquid injector and a gaseous injector, 2410 injecting diesel and gas mixture. The work substituted the compressed air traditionally used with the AADI system by methane, thus adapting the system for RCCI mode 2411 2412 application. The proposed system can produce direct injections for both the LRF and the 2413 HRF. The authors demonstrated that LPDDI could achieve RCCI combustion mode with 2414 diesel/CH₄. They underscored the importance of the LRF, and the HRF injection timings 2415 over the combustion phasing, specifying that optimized combustion was achieved by advancing the HRF timings (-250 °CA aTDC) and retarding the LRF injection 2416 timings(-112 °CA aTDC). 2417



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2419

Figure 41: Cross-section of AADI diesel/CH₄ dual-fuel injector model [354]

To summarize, commercially available RCCI engines are not yet a reality. However, the real-world implementation of RCCI mode has made significant advances in recent years. One of the main tasks to implement the RCCI mode in commercial vehicles is adapting the dual-fuel injection system, but this increases the complexity of the vehicle systems and the control paradigms. Even with these design hurdles, the implementation of the concept has already been tested in cars and maritime vessels. In addition to the real-world

2426 proof of concept, other alternatives have been investigated to bring RCCI mode closer to 2427 commercial feasibility. For example, implementing multi-mode concepts, such as DMDF 2428 and RCCI/CDC, intends to resolve the limitations of the operational range by switching to another combustion mode when RCCI mode proves to be inefficient or unattainable. An 2429 2430 additional strategy proposed was combining RCCI combustion mode in hybrid form for road vehicles. In particular, this strategy has proven to comply with US EPA Tier-2 bin-2431 5 NOx and CO emissions in several driving cycles and indicated that the after-treatment 2432 2433 systems could address the CO and HC emissions.

2434 On the other hand, some studies focused on simplifying the systems necessary for 2435 obtaining the RCCI combustion mode. Strategies for single-fuel RCCI combustion mode 2436 required improving the cetane number of the LRF, which two approaches could obtain: 2437 adding additives to the LRF and then using it as the HRF, and fuel reforming. Fuel reforming consisted of applying on-board processes to modify the reactivity of the LRF to 2438 2439 facilitate controlled auto-ignition. Finally, the dual-direct injection was applied to achieve 2440 RCCI combustion mode. It was possible to reduce CO and HC emissions while 2441 maintaining high thermal efficiencies using DDI technology.

2442 5. Conclusions, Recommendations, and Way Forward

2443 The RCCI combustion mode has gained significant attention from the engine research 2444 community in the last few years due to its excellent combustion stability, efficiency, and 2445 emission characteristics. RCCI combustion mode could be adopted in production-grade engines offering a unique possibility of adopting alternative fuels. This review article has 2446 covered all features of RCCI combustion mode from end to end. The basic mechanism of 2447 2448 all LTC variants is the same; however, RCCI combustion mode deals with reactivities of 2449 two different fuels, which are mainly governed by chemical kinetics. In extensive research related to LTC, it has emerged that all LTC modes have significant potential for emission 2450 2451 reduction; however, a comprehensive analysis of HCCI and PCCI modes of combustion

2452 revealed several limitations, especially related to their application at higher engine loads. 2453 RCCI combustion mode exhibited significantly greater potential in these aspects, explored 2454 in many research studies off-late, including in-cylinder combustion and optical diagnostics. This review article discusses detailed experimental methodologies, 2455 2456 conclusively showing that the RCCI combustion mode has great potential to be adopted in production-grade engines. For this, the dual-mode operation has been explored 2457 2458 significantly among various other techniques due to its lesser complexity and greater 2459 applicability to cater to full engine load operations.

2460 Research and development studies have demonstrated good potential for RCCI 2461 combustion mode to be applied to various engine platforms. Both light-duty and heavy-2462 duty applications showed that the engine-out NOx emission levels were well below the 2463 limits imposed by stringent emissions regulations. In addition, lower soot emissions were observed, without the need for after-treatment devices such as DPF and SCR.RCCI also 2464 delivers higher thermal efficiencies than other LTC modes. A flexible array of fuels can 2465 2466 be used in this combustion mode, which opens the possibility of using fuels synthesized from renewable resources, thus reducing the overall carbon footprint of the transport 2467 2468 sector. Besides these advantages, the RCCI mode has some challenges, such as higher HC and CO emissions at lower loads and excessive PRR at higher loads. These issues restrict 2469 2470 the full map application to only medium engine loads. RCCI mode offers several advantages; however, it still has some problems to overcome. These challenges limit the 2471 engine operation due to high PRR/ knocking. Before the commercialization of the RCCI 2472 2473 combustion mode engines, these issues must be resolved. Literature review shows that 2474 researchers are working to resolve these problems to make the RCCI engine work in a low-environmental impact combustion mode, having good reliability and higher energy 2475 2476 conversion efficiencies. This could make these RCCI mode engines commercially 2477 attractive to the consumers and reduce the CO_2 footprint at the same time.

RCCI offers very low NOx emissions, which can eliminate dependence on SCR systems,
along with reduced PM emissions. Higher HC and CO emissions are a penalty because of
low combustion temperatures. However, the after-treatment systems have shown
excellent conversion efficiencies to comply with the emission legislations.

2482 Various combustion control strategies have shown promising control over the PRR, such 2483 as retarding the HRF injection timings and introducing the EGR. These strategies help 2484 increase the mixture's resistance (previously introduced as LRF) to autoignition and help 2485 charge dilution. Higher boost pressures have shown a reduction of PRR, which prompts 2486 finding a way to obtain required airflow under real-world operating conditions. Pathways 2487 to resolving these issues are partially completed by developing mixed combustion modes 2488 such as DMDF, which go from the pure RCCI mode concept to combustion suitable for 2489 varying loads. Electrical hybridization of the RCCI combustion mode engine is also hugely beneficial since the electric motor can work over a load-speed range where the RCCI 2490 2491 combustion mode engine struggles. The transient operations were verified under the 2492 discussed concept. The next logical step towards commercial implementation would be 2493 proving this concept in real driving conditions. Retrofitting of current engines has also 2494 been of some interest to researchers and manufacturers. To further improve the prospect for this concept, the use of unconventional fuels with suitable properties facilitating the 2495 2496 RCCI mode could be explored. These would promote ultra-low soot emissions due to higher 2497 oxygenation (like OMEx) or a closer to net neutral CO_2 footprint due to its production 2498 cycle and even tank-to-wheel operation, but are aspects that need to be explored further. 2499 Once technical problems are resolved, commercial adoption of the RCCI mode concept 2500 would require addressing functional issues such as reliability and fuel flexibility. A technical solution should be developed for an efficient control method to operate the 2501 2502 engine in different combustion modes if one of the two fuels is unavailable. Further 2503 research and development of a single-fuel RCCI combustion mode engine development

2504 would be a topic of interest for researchers, which would resolve several hurdles of having 2505 to different fuel systems for the LRF and the HRF. Since costs are an important decision factor for the consumer and corporations, reducing the complexity of the fuel injection and 2506 2507 control systems would also be essential for the commercialization of the RCCI mode 2508 engine technology. With increasingly stringent emissions limitations, a methodology should be developed to assess whether the improvements in NOx and soot emissions are 2509 2510 worth the CO and HC penalty, as well as the increased system complexity and possible 2511 costs of having two distinct fuel injection systems. Besides some of the representative examples of real-world operation the concept of RCCI has not been commercially 2512 2513 widespread, and other, simpler, alternatives have been prioritized by manufactures that 2514 can have similar increases in thermal efficiency or emissions buffering. The conversion 2515 efficiency of the after-treatment systems for the dedicated RCCI mode engine would be an important development in the near future, as it could help determine whether the concept 2516 2517 can make possible the emissions reductions necessary to reach global targets.

2518 Nomenclature

2-EHN	2-ethylhexyl nitrate
AADI	air-assisted direct injection
AMP	accumulation mode particles
ATAC	active thermo-atmosphere combustion
BC	black carbon
BMEP	brake mean effective pressure
BSFC	brake-specific fuel consumption
CAD	crank-angle degree
CAxx	crank angle position for xx% cumulative heat release
CBG	compressed biogas
CDC	conventional diesel combustion

CHR	cumulative heat release
CI	compression ignition
CL	close-loop
CN	cetane number
CO	carbon monoxide
CO2e	CO_2 equivalent
CPOX	catalytic partial oxidation
DDFS	direct dual-fuel stratification
DDI	dual direct injection
DEF	diesel exhaust fluid
D-EGR	dedicated EGR
DI	direct injection
DICI	direct injection compression ignition
DMCC	diesel methanol compound combustion
DMDF	diesel-methanol dual-fuel
DMDF	dual-mode dual-fuel
DME	dimethyl ether
DOC	diesel oxidation catalyst
DP	dynamic programming
DPF	diesel particulate filter
DPI	diesel pilot injection
DTBP	diterbutyl peroxide
EGR	exhaust gas recirculation
EMC	energy management control
EREV	extended range electric vehicle
EV	electric vehicle

FFV	fuel flexible vehicles
FHEV	full hybrid electric vehicle
FIP	fuel injection pressure
FoV	field of view
FSN	filter smoke number
FTP-75	Federal Test Procedure
GCI	gasoline compression ignition
GDI	gasoline direct injection
GHG	greenhouse gas
GM	General Motors
HC	hydrocarbon
HCCI	homogeneous charge compression ignition
HCHR	homogeneous combustion heat release
HRF	high reactivity fuel
HRR	heat release rate
HTHR	high-temperature heat release
HVO	hydrotreated vegetable oil
HWFET	Highway Fuel Economy Test
ICE	internal combustion engine
ICFB	in-cylinder fuel blending
ICT	intake charge temperature
ITE	indicated thermal efficiency
ITEg	gross indicated thermal efficiency
JC08	Japanese cycle
LCA	lifecycle analysis
LIVC	late intake valve closing

LPDDI	low-pressure dual-fuel direct injection
LPG	liquefied petroleum gas
LRF	low reactivity fuel
LTC	low temperature combustion
LTHR	low-temperature heat release
MHEV	mild hybrid electric vehicle
MPC	model-predictive control
MPRR	maximum pressure rise rates
MSR	methanol substitution ratio
NC	next cycle
NFL	natural flame luminosity
NL	natural luminosity
NMP	nucleation mode particles
NOx	nitrogen oxides
NP	nanoparticles
NVH	noise vibration harshness
NVO	negative valve overlap
OC	organic carbon
OL	open-loop
PAHs	polycyclic aromatic hydrocarbons
PCCI	premixed charge compression ignition
PCHR	premixed combustion heat release
PCI	premixed compression ignition
PDFC	piston-split dual-fuel combustion
PHEV	plug-in hybrid electric vehicle
PLIF	planar laser-induced fluorescence

PM	particulate matter
PN	particle number
PODEn	polyoxymethylene dimethyl ether
PPC	partially premixed combustion
PPCCI	partially premixed charge compression ignition
PRF	primary reference fuel
PRR	pressure rise rate
PSD	particle size distribution
RBC	rule-based control
RCCI	reactivity controlled compression ignition
RDE	real driving emissions
RON	research cetane number
R-RCCI	reverse RCCI
SCR	selective catalytic reduction
SHEV	series hybrid electric vehicle
SI	spark ignition
SMR	steam methane reforming
SoC	star of combustion
SOC	state of charge
SPCCI	spark controlled compression ignition
SR	steam reforming
TC	total carbon
TDC	top-dead-center
TFR	thermochemical fuel reformer
TPM	total particle mass
TPME	Thevetiaperuviana methyl ester

UDDS	urban dynamometer driving schedule
v/v	volume over volume
VCR	variable compression ratio
VVT	variable valve timing
WHTC	Worldwide Harmonized Light Vehicle Test Cycle
WLTP	World Harmonized Light Vehicles Test Procedure

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