

## INDEX

<b>Chapter 1. <i>Introduction</i> .....</b>	<b>1</b>
1.1 Catalysis.....	3
1.2. Heterogeneous catalysis .....	4
1.3. Oxidation reactions .....	7
1.4. Condensation reactions.....	8
1.5 Metal-organic frameworks .....	11
1.6 MOFs as heterogeneous catalysts.....	12
1.7 MOFs for oxidation reactions .....	15
1.8. MOFs as catalysts for condensation reactions.....	20
1.9. Metal-free heterogeneous catalysts .....	23
1.10. Graphene-based materials as metal-free catalysts.....	25
1.11. References.....	31
<b>Chapter 2. <i>Objectives</i> .....</b>	<b>35</b>
<b>Chapter 3. <i>Desing of stable mixed-metal MIL-101(Cr/Fe) materials with enhanced catalytic activity for the Prins reaction</i> .....</b>	<b>39</b>
3.1 Abstract .....	43
3.2 Introduction.....	43
3.3 Results and discussion.....	46
3.3.1 Catalyst preparation.....	46
3.3.2 Catalytic activity .....	53
3.3.3 Characterization of acid sites .....	62
3.4 Conclusions.....	66
3.5 Experimental section.....	66
3.5.1 Materials .....	66
3.5.2 Catalyst preparation.....	66
3.5.3 Characterization .....	67
3.5.4 Catalytic reaction.....	68

3.6 Acknowledgments .....	69
3.7 References.....	71
3.8 Supplementary Material.....	74
<b>Chapter 4. Nitro functionalized chromium terephthalate metal-organic framework as multifunctional solid acid for the synthesis of benzimidazoles.....</b>	<b>85</b>
4.1 Abstract .....	89
4.2 Introducion .....	89
4.3 Experimental .....	92
4.3.1 Materials .....	92
4.3.2 Catalyst preparation.....	92
4.3.3 Catalyst characterization.....	93
4.3.4 Reaction procedure.....	94
4.4 Results and discussion.....	95
4.4.1 Structural description of catalysts.....	95
4.4.2 Catalytic activity .....	97
4.4.3 Characterization of the Lewis acid centers .....	107
4.5. Conclusions.....	109
4.6 Acknowledgements .....	110
4.7 References.....	111
4.8 Supporting information .....	114
<b>Chapter 5. MIL-101(Cr)-NO<sub>2</sub> as efficient catalysts for the aerobic oxidation of thiophenols and the oxidative desulfurization of dibenzothiophenes .....</b>	<b>133</b>
5.1 Abstract .....	137
5.2 Introduction.....	137
5.3 Materials and methods .....	140
5.3.1 Materials .....	140
5.3.2 Catalyst characterization.....	140
5.3.3 Catalytic experiments.....	141

5.3.4 Product analysis .....	141
<b>5.4 Results and discussion.....</b>	<b>142</b>
5.4.1 Catalyst properties .....	142
5.4.2 Aerobic oxidation of thiophenols to disulfides .....	143
5.4.3 Aerobic oxidative desulfurization of diesel fractions. Use of DBTs .....	149
5.5 Conclusions.....	156
5.6 Acknowledgements.....	157
5.7 Reference .....	158
5.8 Supplementary Material.....	160
<b>Chapter 6. <i>Tuning the active sites in reduced grapheme oxide by hydroquinone functionalization for the aerobic oxidations of thiophenol and indane.....</i></b>	<b>167</b>
6.1 Abstract .....	171
6.2 Introduction.....	171
6.3 Experimental section.....	173
6.3.1 Materials .....	173
6.3.2 Catalyst preparation.....	173
6.3.3 Instrumentation .....	175
6.3.4 Aerobic oxidation of thiophenol .....	175
6.3.5 Aerobic oxidation of indane .....	175
6.4 Results and discussion.....	176
6.4.1 Functionalizations of rGO.....	176
6.4.2 Aerobic oxidation of thiophenol .....	179
6.4.3 Aerobic oxidation of indane .....	183
6.5 Conclusions.....	189
6.6 Acknowledgements .....	190
6.7 References.....	191
6.8 Supplementary information .....	193
<b>Chapter 7. <i>Conclusions .....</i></b>	<b>195</b>
<b>Chapter 8. <i>Abstract of the Thesis.....</i></b>	<b>201</b>

8.1 Abstract .....	203
8.2 Resumen.....	204
8.3 Resum .....	205
<b>Annex I. <i>Publications and conferences</i>.....</b>	<b>207</b>
Publications derived from the present Thesis.....	209
Other Publications.....	210
Conferences.....	211