



UNIVERSITAT
POLITÈCNICA
DE VALÈNCIA

**ESTABILIZACIÓN DE REVESTIMIENTOS DE TIERRA
USANDO CERATONIA SILIQUA L.**

Programa de Doctorado en Arquitectura, Edificación, Urbanismo y Paisaje

TESIS DOCTORAL

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Marzo 2021

AGRADECIMIENTOS

Esta tesis fue depositada en la Universitat Politècnica de València en 2021, y me gustaría finalizar agradeciendo a toda la gente que ha contribuido en la redacción de esta.

En primer lugar, agradecer a mis tutores, Carlos Hidalgo Signes, Begoña Serrano Lanzarote y Gabriel Barbeta Solà, por su apoyo profesional y moral, siendo siempre un seguro y respaldo para poder continuar con la investigación. Gracias al Departamento de Ingeniería del Terreno por cederme sus instalaciones para realizar los ensayos de laboratorio, en especial a Carlos Hidalgo Signes y a Ángel Morilla Rubio. También al Instituto Valenciano de la Edificación por cederme el espacio para realizar los prototipos, en especial a Begoña Serrano Lanzarote por abrirme las puertas de éste y a Francisco Pla por su total disponibilidad y servicio.

Agradecer también al Departamento de Ingeniería Química, en especial a Jaime Primo por su predisposición a ayudar en la extracción del extracto de la *Ceratonia Siliqua L.*, al igual que a Victoria Lizama por su apoyo y paciencia, al mismo tiempo que dedicación desinteresada en la investigación en productos no afines a su labor profesional común.

También me gustaría dar las gracias a mi equipo de trabajo a título profesional, a la cooperativa Okambuva.coop, la cual inició mi interés por esta disciplina, y cuya experiencia y motivación por la construcción con tierra fueron esenciales para poder disfrutar plenamente de la investigación.

A mi familia, en especial a mi madre por su constante apoyo, interés y alegrías compartidas, siempre un pilar básico en mi evolución profesional y personal. A Salut, mi compañera de vida, pieza esencial en mi camino profesional y quien me escucha y anima sin descanso a las buenas y a las malas, y que siempre ha creído en mí.

RESUMEN

En todos los continentes, un rico patrimonio arquitectónico atestigua el uso de la tierra como apto para la construcción, acuciando unas bondades difícilmente alcanzables por sus homólogos contemporáneos. Hoy en día, la tierra como material de construcción es utilizada por aproximadamente la mitad de la población mundial, mediante diversas técnicas.

Durante las últimas décadas ha surgido un creciente interés por parte de arquitectos e ingenieros investigadores, aplicando la tecnología moderna en construcción e investigación para facilitar el uso de la tierra cruda, así como en la mejora de las técnicas y sistemas tradicionales readaptándolos a las exigencias normativas contemporáneas.

La dificultad más importante detectada en la incorporación de la construcción con tierra al flujo de trabajo del sector de la construcción actual es su velocidad de secado y, por tanto, la velocidad de puesta en obra del sistema, ya que esto repercute en gran medida en los rendimientos unitarios de las partidas de ejecución, dejando la construcción con tierra relegada a casos puntuales y extraordinarios de ejecución de obra o a la rehabilitación.

El objetivo principal de la investigación ha sido determinar y aplicar medidas de mejora de la puesta en obra de elementos constructivos ejecutados con técnicas de construcción con tierra para su conciliación a la logística y rendimientos de trabajo del sector convencional actual.

Para ello se han detectado medidas de mejora mediante aditivos naturales para aumentar y mejorar las posibilidades mecánicas del material tierra cruda demostrando su idoneidad técnica mediante ensayos mecánicos, subrayando finalmente su importancia en clave sostenible en comparación con sus homólogos convencionales.

Palabras clave: construcción con tierra; tierra vertida; circularidad; superplastificantes; análisis de ciclo de vida.

ABSTRACT

On all continents, a rich architectural heritage testifies to the use of the earth as suitable for construction, stressing benefits that are difficult to achieve by its contemporary counterparts. Today, land as a building material is used by approximately half of the world's population, through various techniques.

During the last decades there has been a growing interest on the part of architects and research engineers, applying modern technology in construction and research to facilitate the use of raw land, as well as in the improvement of traditional techniques and systems, readapting them to regulatory requirements contemporary.

The most important difficulty detected in the incorporation of construction with earth into the workflow of the current construction sector is its drying speed and, therefore, the speed of putting the system into work, since this has a great impact on the unit returns of the execution items, leaving the construction with land relegated to specific and extraordinary cases of work execution or rehabilitation.

The main objective of the research has been to determine and apply measures to improve the implementation of construction elements executed with earth construction techniques for their reconciliation to the logistics and work performance of the current conventional sector.

For this, improvement measures have been detected using natural additives to increase and improve the mechanical possibilities of the raw earth material, demonstrating its technical suitability through mechanical tests, finally underlining its importance in a sustainable key compared to its conventional counterparts.

Keywords: earthen construction; poured earth; circularity; superplasticizers; life cycle analysis.

RESUM

En tots els continents, un ric patrimoni arquitectònic testifica l'ús de la terra com a apte per a la construcció, acuitant unes bondats difícilment assolibles pels seus homòlegs contemporanis. Avui dia, la terra com a material de construcció és utilitzada per aproximadament la meitat de la població mundial, mitjançant diverses tècniques.

Durant les últimes dècades ha sorgit un creixent interès per part d'arquitectes i enginyers investigadors, aplicant la tecnologia moderna en construcció i investigació per facilitar l'ús de la terra crua, així com en la millora de les tècniques i sistemes tradicionals readaptándolos a les exigències normatives contemporànies.

La dificultat més important detectada a la incorporació de la construcció amb terra a el flux de treball de sector de la construcció actual és la seva velocitat d'assecat i, per tant, la velocitat de posada en obra de sistema, ja que això repercuteix en gran mesura en els rendiments unitaris de les partides d'execució, deixant la construcció amb terra relegada a casos puntuals i extraordinaris d'execució d'obra o la rehabilitació.

L'objectiu principal de la recerca ha estat determinar i aplicar mesures de millora de la posada en obra d'elements constructius executats amb tècniques de construcció amb terra per a la seva conciliació a la logística i rendiments de treball de sector convencional actual.

Per això s'han detectat mesures de millora mitjançant additius naturals per augmentar i millorar les possibilitats mecàniques de l'material terra crua demostrant la seva idoneïtat tècnica mitjançant assaigs mecànics, subratllant finalment la seva importància en clau sostenible en comparació dels seus homòlegs convencionals.

Paraules clau: construcció amb terra; terra abocada; circularitat; superplastificants; anàlisi de cicle de vida.

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Unidades y variables:

| | |
|----------------------------|--|
| <i>kgCO₂-eq</i> | kilogramos de CO ₂ equivalente |
| <i>MJ</i> | Uso de recursos, energía |
| <i>kg Sb eq</i> | Uso de recursos, mineral y metal |
| <i>pK_a</i> | Constante de disociación ácida (K _a) |
| <i>ρ</i> | Density |
| <i>Δρ</i> | Diferencial de densidad |
| <i>F_{cm}</i> | Resistencia media a compresión |
| <i>F_{ctm}</i> | Resistencia media a flexión |
| <i>PI</i> | Índice de plasticidad |
| <i>WS</i> | Límite de Retracción |
| <i>pH</i> | Concentración de iones de hidrógeno en la disolución |
| <i>RH</i> | Relative humidity |
| <i>G_s</i> | Specific gravity |
| <i>w%</i> | Porcentaje en peso |
| <i>w/w%</i> | Relación peso/peso en porcentaje |

Acrónimos:

| | |
|-------------|---|
| <i>GA</i> | Ácido Gálico |
| <i>CT</i> | Tanino de algarrobo |
| <i>CEC</i> | Capacidad de intercambio catiónico |
| <i>FAAS</i> | Flame atomic absorption spectroscopy |
| <i>USDA</i> | United States Department of Agriculture |
| <i>HPLC</i> | High Performance Liquid Chromatography |
| <i>XRD</i> | Test de difracción X-ray |

| | |
|-------------|---|
| <i>IS</i> | Suelo Illítico |
| <i>KS</i> | Suelo kaolinitico |
| <i>SS</i> | Suelo Esmectítico |
| <i>USCS</i> | Unified Soil Classification System |
| <i>AF</i> | Fine aggregates |
| <i>FG</i> | Fine granulometry |
| <i>AG</i> | Gravel aggregates |
| <i>GG</i> | Gravel granulometry |
| <i>PYBC</i> | Pastas y barbotinas cerámicas |
| <i>LCA</i> | Analisis de ciclo de vida |
| <i>SCCC</i> | Self compacting clay-based concrete |
| <i>UNE</i> | Normas de Ensayo Españolas (Una Norma Española) |
| <i>ASTM</i> | ASTM International - Standards Worldwide |
| <i>PNE</i> | Rules for presentation of European Standards |
| <i>GWP</i> | Global warning potential |
| <i>IPCC</i> | Intergovernmental Panel on Climate Change |
| <i>CTE</i> | Código técnico de la edificación |

CAPITULO 1: INTRODUCCIÓN

1.1. Antecedentes

La construcción con tierra forma parte inherente de nuestra arquitectura vernácula (Red and Cytel 2003). En las últimas décadas, un renacimiento de dicho material está provocando un creciente interés en el sector de la investigación y de la industria de la construcción, con nuevas tendencias del uso de la tierra en distintas aplicaciones, desde su uso en revestimientos hasta el estructural (Easton 2012).

Mayormente, el trabajo de investigación en el área de la construcción con tierra normalmente queda acotado dentro del marco de la rehabilitación o el trabajo de recopilación de ejemplo de las distintas técnicas de la arquitectura vernácula, relegando a este material a un campo de desarrollo meramente de arquitectura de otro tiempo, sin proyección en su incorporación en el campo de la construcción contemporánea.

Uno de los objetivos principales de la unión europea (EU) según la agenda 2030 es el desarrollo sostenible y la transición ecológica traducida en distintos objetivos principales (Kroll, Warchold, and Pradhan 2019), uno de ellos repercute directamente sobre la descarbonización del sector de la construcción, uno de los sectores con mayor impacto medioambiental según (European Commission 2015).

Es por ello que, en los últimos años vemos como renace un nuevo tipo de arquitectura, usando técnicas y materiales cuya materia prima conlleva una energía embebida mucho menor (Ben-Alon et al. 2019). La tierra es protagonista en este cambio de paradigma en la construcción sostenible, renovando técnicas de construcción ancestrales, y adaptándolas a la logística, condiciones de trabajo y normativa actuales como es el caso de la normativa específica, siendo un claro ejemplo de transformación y resiliencia dentro del sector que más energía demanda, la construcción.

Uno de los campos de desarrollo que la técnica de la construcción con tierra ha tomado, es la investigación en el trabajo de la prefabricación, pues la velocidad y la puesta en obra de esta técnica ha sido uno de los lastres que ha ralentizado su incorporación en obra, dada su directa repercusión en su viabilidad económica. Esta velocidad de puesta en obra varía en dependencia de la técnica constructiva, pasando por técnicas de trabajo de la tierra en consistencia seca, como por ejemplo la tapia, hasta técnicas de consistencia

húmeda, como el muro de mano (*cob* en inglés), hasta consistencias líquidas como la tierra vertida (Ma, Chen, and Chen 2016).



Fig. 1 Revestimiento de arcilla illítica en consistencia líquida mediante dispersión.

Europa demanda de forma creciente una transformación clara en materia de sostenibilidad. Herramientas como el LCA pretenden traducir este requisito una parametrización clara del proceso de producción, uso y destrucción de los materiales de construcción, y es por ello por lo que una producción de materiales de construcción, no solo eficiente, sino inocua para el medio ambiente, es primordial para el cumplimiento de los objetivos cercanos de la UE.

En Europa, varios países ya han adaptado en su normativa materiales sostenibles como la tierra, estando incorporados en su código de edificación. En España algunas técnicas de construcción con tierra ya cuentan con normativa específica, como el bloque de tierra compactado del bloque de tierra compactado BTC UNE 41410: Bloques de tierra comprimida para muros y tabiques. Definiciones, especificaciones y métodos de ensayo (UNE 41410 2008), sin embargo, por norma general procedimientos de ensayos en técnicas paralelas de construcción con tierra aún han de acogerse a estándares de análisis prediseñados para otros conglomerantes como el cemento (González Serrano 2016).

Como resumen, las dificultades a resolver detectadas y a acometer por la tesis se pueden resumir en:

- La falta de inventariado de protocolo de análisis para el desarrollo de identificación de aditivos.

- La falta de estandarización de ensayos del sector de la construcción convencional a la construcción con tierra.
- La falta de comparativas medioambientales objetivas entre técnicas de construcción convencionales y sostenibles.

1.2. Objetivos

1.2.1. Objetivos generales

Los tres objetivos generales de esta tesis se podrían resumir en:

- 1- Identificar un aditivo natural asequible y cercano, susceptible de mejorar las condiciones mecánicas de la tierra (artículo 1).
- 2- Establecer un protocolo de extracción del compuesto activo reactivo con el suelo, y evaluar las mejoras mecánicas de un conglomerado de arcilla (artículo 2) (artículo 3).
- 3- Aplicar el aditivo estudiado sobre un material comercializable y compararlo mediante un LCA (artículo 3).

1.2.2. Objetivos específicos

Para lograr los objetivos anteriores, se establecieron estos objetivos específicos:

- 1A: Estudiar bibliografía específica para identificar tipos de aditivos usados a lo largo de la historia, reconociendo recetas y metodología de extracción claras y comparativas.
- 1B: Identificar productos del inventariado con potencial desarrollable en la zona.
- 1C: Estudiar los mecanismos de interacción entre los componentes activos de los aditivos y los suelos arcillosos y sus posibles medidas de mejora.
- 2A: Establecer un procedimiento de extracción del componente activo desde la materia prima y estudio de su rendimiento de extracción.
- 2B: Establecer un procedimiento estandarizado de identificación de la reactividad del compuesto con los suelos arcillosos.

-2C: Identificar las mejoras en las propiedades mecánicas de un conglomerado de arcilla.

-3A: Extrapolar el procedimiento de identificación de reacción con un aditivo comercial sobre un conglomerado tipo hormigón.

-3B: Crear un escenario comparativo entre los dos elementos constructivos, uno de base arcillosa y otro de base cementosa, para realizar un LCA.

No todos los objetivos específicos están plasmados en los artículos publicados, pero sí han sido desarrollados en el cuerpo de la investigación que ha dado lugar a los mismos.

1.3. Alcance e hipótesis de partida

Como se mencionó anteriormente, el objetivo es tratar de enlazar las técnicas de construcción con tierra y enfrentarla a las condiciones logísticas y de trabajo de la construcción convencional. Entre los distintos aditivos identificados, el que resultó más atractivo a desarrollar eran los superplastificantes basados en el ácido gálico de algunos productos vegetales. El uso de estos superplastificantes pretendía incidir directamente en las siguientes propiedades:

- *Hipótesis 1: Se puede mejorar la rapidez de puesta en obra del mortero de arcilla, reduciendo la cantidad de agua de amasado y reducir el tiempo de secado.*
- *Hipótesis 2: Se puede aumentar la resistencia a compresión de los elementos conformados con mortero u hormigón de arcilla.*
- *Hipótesis 3: El hormigón de arcilla puede cumplir con la normativa aplicable para ciertos escenarios y solicitudes.*
- *Hipótesis 4: El hormigón de arcilla puede sustituir al hormigón de cemento en ciertos escenarios siendo mucho más sostenible.*

1.4. Metodología

La metodología utilizada en esta tesis se compone de los siguientes pasos:

Fase 1: Estado del arte

La tesis trata sobre el estudio y mejora de las propiedades mecánicas de un material del sector de la construcción sostenible, la tierra. Primeramente, se necesita recopilar información sobre cuáles son los aditivos aplicables a la tierra para mejorar de forma importante su comportamiento reológico, y a ulterior sus propiedades mecánicas:

- Estado del arte sobre aditivos naturales o artificiales usados en la construcción con tierra.
- Estado del arte sobre ensayos y estudios extrapolados de la normativa existente del sector convencional a la construcción con tierra.
 - o Ensayos de caracterización de suelos.
 - o Ensayos mecánicos en morteros de arcilla.
 - o Ensayos mecánicos en hormigones de arcilla.
- Protocolos de aditivado de argamasas en base arcillosa similares
 - o Porcentajes admisibles de aditivado.
 - o Esqueleto granulares para argamasas de arcilla.
- Estado del arte de análisis de ciclo de vida de técnicas de construcción con tierra existentes.

Fase 2: Estudio del aditivo y su metodología de extracción

Tras una búsqueda pormenorizada de la bibliografía tanto contemporánea como histórica en el campo de los aditivos de la construcción con tierra:

- Se decide trabajar sobre un comportamiento y mejora mecánica específica: la modificación la reología de la tierra.
- Se escoge el espécimen disponible en cantidad y cercanía para la extracción del compuesto reactivo, la *ceratonia siliqua l.* (albarrobo).
- El último paso es escoger el método de extracción de entre los distintos existentes en la bibliografía, comparando rendimientos, fiabilidad y reproducibilidad.

Fase 3: Protocolo de análisis de materia prima

Para poder hacer reproducible el estudio, un trabajo importante fue la caracterización de la materia prima, tanto del aditivo como de los suelos, y establecer un protocolo claro de análisis:

- Se caracterizan física y químicamente los 3 tipos de suelos distintos: suelos illíticos, suelos esmectíticos, suelos caoliníticos.
- Se estudia su capacidad de intercambio catiónico para evaluar el potencial de reacción.
- Se realiza un estudio de protonación del aditivo para establecer condiciones de entorno ideales.

Fase 4: Creación de patrón de evaluación de reactividad reproducible

- Para medir la reactividad de este se crea una parrilla de pruebas para comparar la eficacia del aditivo según las variables condicionantes: el % de agua de amasado y su pH ideal.
- Una vez determinado el potencial de reacción, se crea un protocolo de actuación para poder reproducirlo en otros suelos de condiciones naturales distintas. El procedimiento de ensayo se puede ver en la Figura 2.

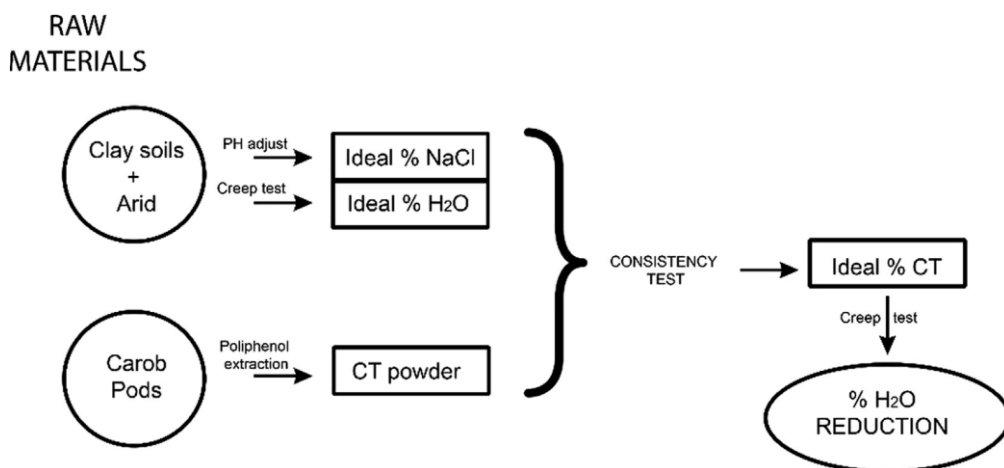


Fig. 2 Protocolo de actuación para determinar la reducción de % agua de amasado.

- Finalmente se realiza una serie de ensayos mecánicos para evaluar el potencial de la medida de mejora propuesta. Algunos de estos ensayos se importaron de

la normativa existente de los materiales convencionales como el cemento, otros se implementaron de ensayos aplicables adaptándolos al caso de estudio.

Fase 5: Puesta en práctica del protocolo de análisis sobre materiales de obra disponibles en el mercado. Caso de estudio.

La metodología se extrapola a materiales disponibles cercanos, tanto de los suelos arcillosos, como los áridos de cantera y el aditivo comercial para recrear un elemento constructivo afín a las posibilidades de mejora del aditivo, en este caso una losa estructural de vivienda residencial.

- Se caracteriza el árido disponible de la cantera suministradora. Éste sirvió para recrear el esqueleto granular ideal para un hormigón de arcilla según su capacidad de adhesión y para facilitar un comportamiento autocompactante.
- Se estudia el suelo arcilloso suministrado por una cantera cercana. Se procedió a caracterizarlo según el protocolo de laboratorio.
- Se preparan las probetas necesarias para poder justificar el elemento constructivo a la normativa aplicable.

Fase 6: Análisis de ciclo de vida comparativo

Finalmente, con el elemento constructivo ejecutado y ensayado, se procede a realizar un ACV (análisis de ciclo de vida) mediante la metodología IPCC.GWP 100a para determinar la reducción y por tanto mejora en términos de sostenibilidad de un material de construcción convencional y el propuesto.

1.5. Difusión y divulgación de los resultados

El objetivo principal de una Tesis doctoral es compartir y difundir los conocimientos adquiridos, repercutiendo en la mayor medida posible sobre la comunidad científica, principalmente mediante artículos científicos, cuya finalidad es crear redes y sinergias entre los investigadores afines, y avanzar en la materia objeto de la investigación en pro de la comunidad científica.

Por otro lado, paralelamente es importante que esta información, en otro registro, cale en la sociedad y en última instancia en el interés comercial e industrial. Para ello es importante la difusión del material desarrollado a través de distintos medios, como

simposios, intervenciones en instituciones públicas afines, congresos o medios de comunicación de mayor o menor entidad.

A parte del trabajo de desarrollo de artículos científicos, y dada la naturaleza de los centros en los que se desarrollaron las estancias, se pueden acometer un trabajo de difusión de la materia a través de formación y cursos que a día de hoy se sigue conduciendo en formación reglada en materia de construcción sostenible.

En todos los artículos expuestos a continuación el autor principal es el autor de la presente Tesis Doctoral, y se recogen en el anexo situado al final del documento, y son los siguientes:

- 1- **ARTÍCULO 1:** J. Romero Clausell, G. Barbeta, Stabilisation of earthen surfaces using carob (*Ceratonia siliqua* L.), CRC Press. Vernacular (2017) 789–796. [doi:10.1201/9781315267739-129](https://doi.org/10.1201/9781315267739-129).
- 2- **ARTÍCULO 2:** J. R. Clausell, C. H. Signes, G. B. Solà, B. S. Lanzarote and M. Inclán. “Improvement in the rheological and mechanical properties of clay mortar after adding *Ceratonia Siliqua* L. extracts,” *Constr. Build. Mater.*, vol. 237, p. 117747, 2020, [doi:10.1016/j.conbuildmat.2019.117747](https://doi.org/10.1016/j.conbuildmat.2019.117747).
- 3- **ARTÍCULO 3:** Romero Clausell, J., Quintana-Gallardo, A., Hidalgo Signes, C. Lanzarote Serrano, B. Environmental evaluation of a self-compacted clay based concrete with natural superplasticizers. *Mater Struct* 54, 20 (2021). [doi:10.1617/s11527-020-01586-6](https://doi.org/10.1617/s11527-020-01586-6).

Los directores de tesis son el Dr. Carlos Hidalgo Signes, Dra. Begoña Serrano Lanzarote y Dr. Gabriel Barbeta Sola. La tesis doctoral se enmarca en el Programa de Doctorado en Arquitectura, Edificación, Urbanismo y Paisaje dirigido por Francisco Juan Vidal.

La tesis doctoral está adscrita al proyecto competitivo europeo DRIVE0 (*Driving decarbonization of the EU building stock by enhancing a consumer centred and locally based circular renovation process*) del programa HORIZON 2020.

Los autores de los artículos son:

- El candidato a doctorado.
- Los directores y supervisores de la tesis.

- Alberto Quintana y Mario Inclán, colaboradores en la investigación.

Todos (excepto el candidato a doctorado) son doctores.

Por otro lado, el desarrollo de la tesis doctoral contó con la publicación y participación en distintos congresos y actividades específicas y transversales como se lista a continuación:

Publicaciones - Participación en congresos:

Prefabricated modular straw walls and panels for houses building and building renovation. II Congreso Internacional y IV Nacional de Construcción Sostenible y Soluciones Eco-Eficientes (CICSE), mayo de 2017, Sevilla (España).

Paramentos y paneles modulares prefabricados con paja para la construcción de viviendas y rehabilitación energética de edificación. I Jornada de Investigación Universitaria sobre Cambio Climático, octubre de 2018, Valencia (España).

Glocal architecture against climate change: rice straw in Valencia. EAAE-ARCC 2020 International Conference. 2nd Valencia International Biennial of Research in Architecture (VIBRArch 2020), noviembre 2020, Valencia (España).

Calibration with real measurements of a straw-bale passive building model. CYTEF, X Congreso Ibérico - VIII Congreso Iberoamericano de las Ciencias y Técnicas del Frío, Pamplona, (España).

Contratos de investigación derivados de la tesis:

Contrato de investigación para el desarrollo de productos basados en arcilla con la empresa Epifanio campo, S.L, Lugo (España).

Contrato de investigación en el IVE. desarrollo de análisis del ciclo de vida de la base de datos, 2020, Valencia (España).

Formación - Seminarios y cursos recibidos e impartidos:

Curso impartido sobre construcción de una casa de paja, madera, barro, yeso y cal. UNIZAR (universidad de Zaragoza), 2017, Morillo de Tous (España).

Curso de impartido en bioconstrucción práctica con paja, barro, madera y cal., UNIZAR (universidad de Zaragoza), 2018, Aínsa (España).

Participación en el II Seminario Internacional SOS Tierra La protección y la restauración del patrimonio construido con tierra. Experiencias y oportunidades, 2016, Madrid (España).

Docencia impartida en la asignatura Construcción sostenible del Máster en Eficiencia Energética y Sostenibilidad de la UJI (Universidad Jaume I), 2017, 2018 y 2019 y 2020 Castellón (España).

Ponencia en plataforma específica de bioconstrucción sobre la técnica de la tierra vertida: <https://bioconstruccionfutura.com/sesion/hormigon-de-tierra-joan-romero/>

Estancias en otros centros

Estancia de colaboración en empresa de bioconstrucción (Okambuva.coop.V), de septiembre de 2018 a septiembre de 2019, Sagunto (España).

Estancia de colaboración en el IVE (Instituto Valenciano de la Edificación), de enero de 2020 julio de 2020, Valencia (España).

Estancia internacional individual de investigación en la Sapienza University of Rome (Sapienza University of Rome), de julio de 2020 a octubre de 2020, Valencia (España).

CAPITULO 2: COMPENDIO DE ARTÍCULOS

2.1. ARTÍCULO 1

J. R. Clausell, G. B. Solà. “Stabilisation of Earthen Surfaces Using Carob (*Ceratonia Siliqua* L.)” *Vernacular and Earthen Architecture: 2017*, pp. 789–96. Proceedings of SosTierra 2017 (Valencia, Spain, 14-16 September 2017) (1st ed.). CRC Press.

[Doi: 10.1201/9781315267739-137](https://doi.org/10.1201/9781315267739-137)

STABILISATION OF EARTHEN SURFACES USING CAROB (CERATONIA SILIQUA L.)

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CRC Press/Balkema (2018 Taylor & Francis Group, London.):

CRC Press. Vernacular (2017) 789–796.

doi: 10.1201/9781315267739-129 CRC Press/Balkema. All rights reserved.

Received 16 March 2017

Publication year: 2017

Abstract:

Additives have been used in traditional construction throughout history. A significant body of literature endorses the potential and use of natural additives, establishing the suitability of these materials for construction purposes, contingent on the proper extraction, manufacture and application under certain conditions. In this paper we conclude that *Ceratonia siliqua* L., a common tree in the Mediterranean area contains the proper components to be used as consolidant for earthen surfaces. Biopolymers as tannins or polysaccharides contained in their pods are capable, in determinate medium conditions, of reacting in different ways with clay constituents improving mechanical properties of the colloid and endowing new possibilities to the earthen mortar.

1. Introduction

1.1. Background

Traditionally, additives have been used in construction in order to compensate for the mechanical weaknesses of low quality resources, usually in the form of residues or subproducts from related production areas such as blood, cheese, lung, animal hair and urine.

According to Lauren-Brook (2013), natural additives may be divided based on their animal, vegetable or mineral origin. He produced a detailed and complete taxonomy of the historic references of the use of these organic materials, various kinds of admixtures and their desired effects. For example:

| Type of admixture | Desired effect | Organic material |
|-------------------|---|---|
| Waterproof | Decrease permeability | Animal glue plus tannin Bitumen Wax emulsion |
| Water reducer | Reduce water required for given consistency | Sugar |
| Adhesive | Increases bond | Rosin Gelatin Animal glue Gluten Casein Blood Albumen |

Table 1. Effect obtained by organic additives.

With the arrival of cement mortar, the view of additives changed, monopolizing the construction sector attention because of cements new possibilities. However, there were some authors still writing about them, such as (Vicat & Smith 2014) who explained how sugar could be used in small doses to make a mortar called Jaghery.

There are several, sometimes contradictory opinions about the effectiveness of natural additives. In fact, some tests, which compared the properties of renders containing the additive with others which did not, produced negative results with the additives making

no improvement Vargas Neuman, J., Heredia Zavoni, E., Bariola Bernales, J., & Mehta (1986).

On the other hand, several experiments with some of these products were performed with satisfactory results in patrimonial construction restoration and also in new applications. In Cedillo A. (1991) the author, based on his observation and experience of the archaeological site of Balamkú, emphasises the importance of the urgent eradication of the use of synthetic polymers in real archaeological property and points out that a preliminary investigation of the case and a systematic study should be made.

In the same vein, Hoyle (1990) revealed that mucilages are sulfuric acid esters (complex polysaccharide), contained in the vesicular cells of the parenchyma tissue from *Opuntia*. They are insoluble in water, but they can absorb and retain it; when in contact with it they form viscous solutions. This mucilage has conglomerating properties and also inhibits and selects germs, so its use brings material consolidation and avoids bacterium and lichen proliferation.

1.2. Additive families

Besides Sickel's classification in Vissac et al. (2013) a different grouping was proposed, based on molecular composition, as follows:

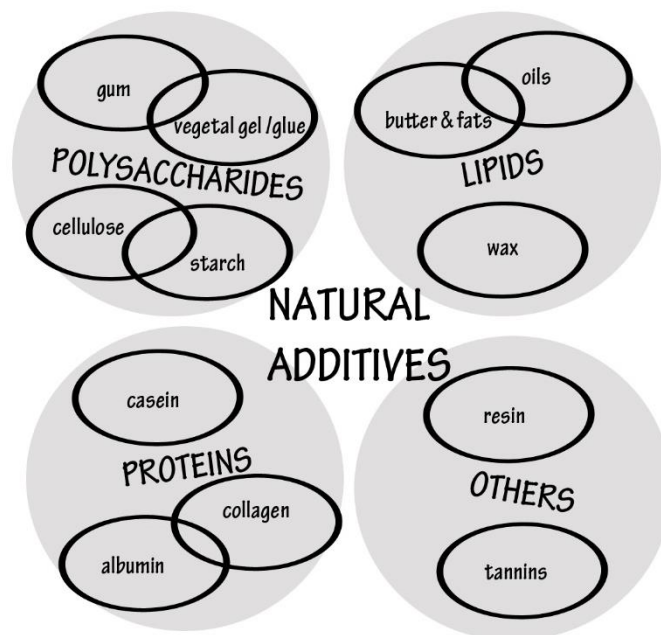


Figure 1. Grouping of additives according to their molecular components. Adapted from Vissac et al. (2013).

Proteins, important molecule for the animal and plant worlds, are biomolecules formed by lineal chains of amino acids, and are considered normally amphiphilic particles.

They are essential for organism growth, playing an important role in different areas, such as structure (collagen), protection (fibrinogen), sending signals (rhodopsin), enzymatic (sucrase) and transport molecules (casein).

There are two types of proteins: fibrous (collagen - normally insoluble in water) and globular (casein or albumin - normally soluble). The structure of each protein will determine its behaviour and interaction with clays.

According to Vissac et al. (2017), proteins create strong bonds with clays, linking their hydrophobic part (head) to the surface of the render, with the remainder of the hydrophilic part outside, creating an external water-repelling coat.

In the polysaccharides group we find sugars in different structures, as shown in Figure 2. This is the largest group, which contains various structures: branched, linear, twisted,

short or long, depending on the nature of the body from which the sugar was extracted. A key characteristic of these types of additives is that they carry a hydroxyl (OH) group.

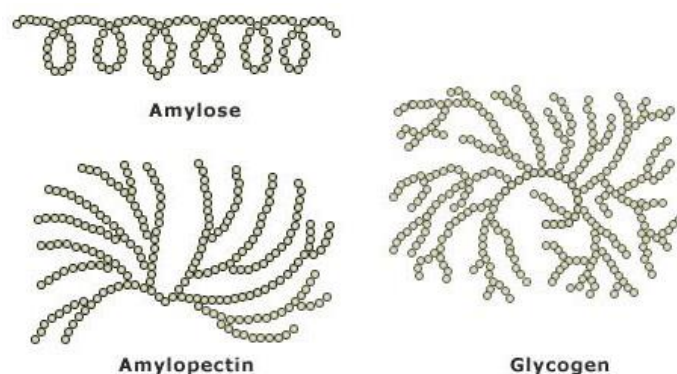


Figure 2. Polysaccharides' different molecular structures (Josh Scheerer).

The molecular structure is not necessarily available as a directly applicable additive and must be subjected to a predetermined previous transformation or extraction process such as grinding, decoction, maceration, etc. The extraction process can release those components susceptible to adsorption in the surface of the clay, which would allow the polysaccharides' chains to form micro networks with the clay particles in different mechanisms as we will see later.

A lipid is a molecule, either completely apolar (neutral lipid) or amphiphilic with a polar head linked strongly to an apolar chain (tail). The main biological functions of lipids include energy storage, signaling, and acting as structural components of cell membranes.

Temperature plays an important role in this group, as it modifies flexibility; depending on the origin of the fat, the substance will present a solid or liquid state: oils, waxes, fats (triglycerides), sterols, butters, among others.

According to Fontaine, Anger, Vissac, et al. (2013) the fatty acids, being small hydrophobic molecules, only allow weak interactions with the clays. Nevertheless, during the siccation step of the unsaturated fatty substances, it is possible that new bonds are created, consolidating the coating.

In the others' group, we find the rest of the biomolecules. Most of them are phenolic compounds such as tannins. Most plants include high percentages of this component as a protection against herbivores or parasites.

Historically, tannin has been historically used to make black metallo-gallic ink, ground and mixed with iron sulphate forming iron tannate, an interesting product for earthen stabilization. We will explore his possibilities later.

1.3. *Extraction methods*

For the proper identification the most suitable extraction methods and in order to obtain the best possible yield with traditional methods, a thorough literature study was conducted in which many sources were consulted. Many of these sources lacked scientific rigour and produced contradictory results or conclusions. It is therefore important to highlight the relevance of the selection and contrast the methods of extraction depending on the additive application.

For instance, Drovou et al. (2015) studied two different ways of extraction for *Parkia biglobosa*, a specimen similar to *Ceratonia siliqua l.*. The first is an artisanal and traditional maceration method, such as those presented in the following Table 2. below. The other method was akin to more commercial methods in presence of bisulfit, meaning that the results of the two methods were rather different. Mostly polyphenolic oligomers are present in the case of the more commercial extraction, while for artisanal maceration technique resulted in the unusual presence of long carbohydrate residues attached to the flavonoids.

These differences in the extracts obtained by different methods indicate that an extraction procedure needs to be chosen according to the purpose for which the tannin extract is to be used.

We framed our study in terms of only two of the additive groups in order to narrow the scope and focus of the research. We chose polysaccharides and tannin extracts, of which the applicable research literature was more focused to the construction sector. These specimens included the *Ficus carica*, *Heuphorbia Characias*, *Ceratonia siliqua*, *Agabe*, *Ficus Indica* and, *Elm tree*, among others.

Now we present some extraction methods from one of the selected specimens, the *Ceratonia siliqua l.*:

Algarrobo (*Ceratonia siliqua l.*)

| | |
|---------------------|--|
| Other names | Carob tree, kharuv, caroubier |
| Similar specimens | Locust tree, Clappertoniana, le néré (<i>Parkia biglobosa</i>), nitta (<i>Parkia filicoidea</i>) |
| Part used | Pods |
| Extract | Carob water |
| Extracting method A | Maceration |

Once the pods were dried, they were collected and cut into pieces of 50mm length. They are allowed to ferment in water and after 15 days the resulting liquid is stored. Manca et al. (2014)

| | |
|---------------------|-------------|
| Part used | Pods |
| Extract | Carob water |
| Extracting method B | Decoction |

Boil the *néré* pods in large pots, until obtaining a viscous liquid of red color. This water will be used for both kneading water and final protection. Pibot (2001)

| | |
|---------------------|-------------|
| Part used | Pods |
| Extract | Carob water |
| Extracting method A | Maceration |

The carob pods were cut into pieces approximately 20 mm in length and then boiled in water. The amount of material used was 10% by weight with respect to the water weight. This preparation boiled until 2/3 of its total volume was consumed. The remnants of the pods were then removed, and the preparation allowed to settle. A liquid of slightly higher viscosity was obtained from the water, orange in color and containing sediments of brown color. Vargas Neuman, J., Heredia Zavoni, E., Bariola Bernales, J., & Mehta (1986)

| | |
|---------------------|-------------|
| Part used | Pods |
| Extract | Carob water |
| Extracting method B | Decoction |

40 g of pods were introduced into 1 liter of water taken at room temperature for 72 h. After filtration, the concentration of tannins in the decoction was estimated (6.33 ± 0.10) g / l. Sorgho et al. (2016)

Part used Pods
Extract Carob water
Extracting method B Decoction

Soak dried carob pods in water for at least one night. The next day they were boiled and result in a hot water of very dark color between red and black to apply on the support. Taxil (2006)

Part used Seeds
Extract Carob gel
Extracting method B Boiling

It has been done by grinding only the seed of the carob tree was ground after being soaked in water for 15 days. Once crushed it was mixed with water. Subsequently, it was heated to 80 °C resulting in a gel. Manca et al. (2014)

Part used Seeds
Extract Carob rubber
Extracting method B Boiling

The seeds were boned by treating the beans with heat-mechanical treatments. Then the peeled seeds were milled and sieved to obtain the endosperm.

The gum of this pre-treated dry powder was extracted with distilled water (197: 1 water-endosperm), a water bath, temperature of 9 °C for a time of 36 min.

The solution and the solid phase were separated by centrifugation at (218 °C, 1 h). The locust bean gum was precipitated with an excess volume of isopropanol. The formed white fibrous precipitate was collected by filtration with 45 µm sieve and washed twice with isopropanol and acetone. After vacuum drying overnight at 30 °C, the precipitate was triturated to a fine powder. El Batal et al. (2013)

Part used Pods
Extract Carob powder
Extracting method B Decoction

The shell was heated to 150 °C for 3 h. It was then cut into pieces, crushed in a RETSCH blade grinder and passed through 2 mm and 0.125 mm sieves. The recovered powder was used as a binder to be mixed with the laterite soil. Banakinao et al. (2016)

Part used Pods
Extract Carob powder
Extracting method A Maceration

The pods were crushed and reduced to grains. They were macerated with water in a container for several weeks. The juice was collected and sprayed onto the coating two to three times at one or two day intervals. Keré (1991)

Part used Seeds
Extract Carob rubber
Extracting method B Decoction

According to a recent patent Audibert, English Patent 241,186 (1925) the seed endosperm is first separated from its shell and sheath by rolling and crushing, roasting up to 150 °C to a golden brown color, and then is extracted with boiling water in quantity 20 times its weight. The filtered clear liquid is evaporated and the dried slurry is sprayed. This powder mixed in water in different proportions gives us rubber with different viscosities. Hart (1930)

Part used Pods

Extract Carob water

Extracting method A Maceration

The pod of *Parkia biglobosa*, dried, was finely ground with the type of mill RETSCH SM 100 knives equipped with a 5 mm grid. The total phenolic compounds were extracted by decoction or maceration of these powders in distilled water for 60 min at temperatures ranging from 70 to 110 °C and following a liquid / solid (L / S: mass / mass) ratio from 6 To 16. The obtained filtration is lyophilized. Nenonene (2009)

Part used Pods

Extract Carob tannins

Extracting method A-B Maceration & decoction

The shells were mechanically axed until a fine powder was obtained from which the tannin extraction was done.

1) Method: Before grinding the shells were dried for 3 h at 110 °C. They were ground to a powder and passed through a sieve to obtain particles of 0.125 mm or less. The powder was left in water (powder / water ratio of 1/6 by weight) at room temperature for 5 days without any added additive. The suspension was filtered and the liquid obtained was concentrated by cooking in the open air without any temperature control. The extract obtained was a dark liquid solution of 50% solids content and high viscosity. Drovou et al. (2015)

2) Industrial method: The powdered shells were placed in a beaker and water was added in a 1:6 powder: water weight ratio. The water had been pre-dissolved 2% sodium bisulfite and 0.5% sodium bicarbonate based on the weight of the powder to be extracted. The extraction was then carried out for 2 hours under continuous mechanical stirring at a temperature of 70-74 °C. The mixture was then filtered and the tannin was recovered by spray-drying with a laboratory spray dryer (Buchi mini-spray dryer B-290), with an inlet temperature of 169°C and an outlet temperature of 70°C. Drovou et al. (2015)

Part used Pods

Extract Carob tannins

Extracting method A Maceration

The pods were pre-dried at 50°C, sliced and mixed with water (40g pods per liter of water) at room temperature for 3 days. After filtration, the dry matter concentration in the solution was 9.6 g / l. The concentration of tannins was 6.3 g / l. Keita et al. (2014)

Table 2. Traditional extraction methods.

2. Local application

2.1. Mediterranean specimens

After the extraction method research, we focused our attention on *Ceratonia siliqua*, given that the literature review reveals the empirical reports and extrapolations from similar specimens from all over the world. Moreover, *Ceratonia siliqua* has components which may potentially work as soil stabilizers; polysaccharides and tannins.

According to Avallone et al. (1997) carob pod meal contained high levels of carbohydrates (45%) mainly sucrose, glucose, fructose and maltose, appreciable amounts of protein (3%), and low levels of fat (0.6%).

The chemical composition of the pulp depends on the cultivar, origin and time of harvest. The authors also proved that 70% of acetone was the most effective solvent for the extraction and recovery of tannins.

Carob pod contains a mean value of 19 mg of total polyphenols / g, 2.75 mg of condensed tannins (proanthocyanidins)/g, and 0.95 mg of hydrolysable tannins (gallo- and ellagitannins)/g. Germ contained higher concentration of total polyphenols (40.8 mg/g) and tannins (16.2 mg of condensed tannins/g and 2.98 mg of hydrolysable tannins/g) while only traces of these compounds were detected in carob seed. Puhan & Wielinga (1996) show a similar composition in his study of Carob Tree:

| Constituent | % |
|----------------------------|---------|
| Total sugars | 48-56 |
| Sucrose | 32-38 |
| Glucose | 5-6 |
| Fructose | 5-7 |
| Pinitol | 5-7 |
| Condensed tannins | 18-20 |
| Non-starch polysaccharides | 18 |
| Ash | 2-3 |
| Fat | 0.2-0.6 |

Table 3. Average composition of *Ceratonia siliqua* pulp.

2.2. Possible interactions

As we showed previously, clays can interact in s with tannins and polysaccharides.

In order to obtain the correct interacting components, it is important to choose the proper extraction methods.

In the case of tannic use, when mixed with mineral phases tannins can act as dispersing agents for clays when pH and ionic forces are controlled, and change the rheology and

plasticity of mixtures. Tannins are also able to form chemical bonds with active sites on mineral surfaces, leading to the change of macroscopic behavior Kaal et al. (2005).

According to Vissac (2013) tannins are formed in several structures of hydroxyl groups, that in certain conditions of pH are able to trap a cation, effect known as chelation.

According to Kaal et al. (2007), tannic acid (TA) and condensed tannin (CT) are strongly retained by mineral phases consisting of quartz with or without coatings of either goethite or ferrihydrite ,confirming that TA and the CTs used are efficient Fe, Al and Si mobilising agents. In natural soil, the fate of tannins after entering the mineral soil is probably affected by the soil's moisture regime and especially by the abundance and type of Fe (oxy)(hydr)oxides. Also, in quartzitic soil poor in secondary oxides, tannins may be tightly bound to Qtz surfaces. The mineral soil is probably affected by the soil's moisture regime and especially by the abundance and type of Fe (oxy)(hydr)oxides.

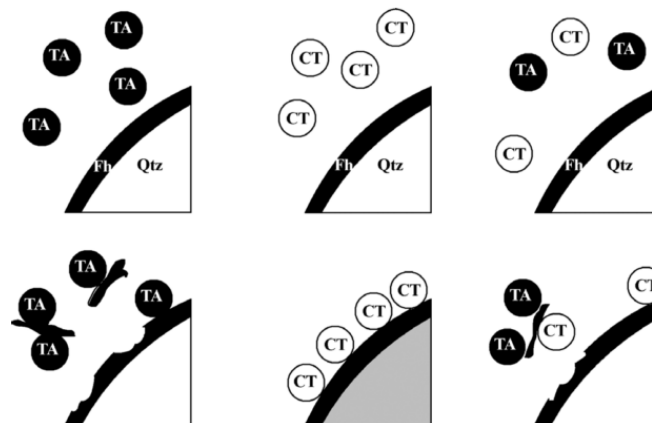


Figure 3. Proposed interaction between tannins and Qtz-Fh complexes. Kaal et al. (2005).

It seems obvious that not only the medium conditions will be determinant, but that the type of soil will also have an important role in this interaction. In Banakinao et al. (2016), the researchers mixed *Parkia Biglobosa* husk powder with a lateritic soil, achieving positive results. On the other hand, *Ceratonia siliqua* has a very good percentage of polysaccharides which have an important potential as consolidants. This interaction is

different from the one with tannins, since now we speak of adsorption (Figure 4.) of polysaccharide macromolecules into the clay surface.

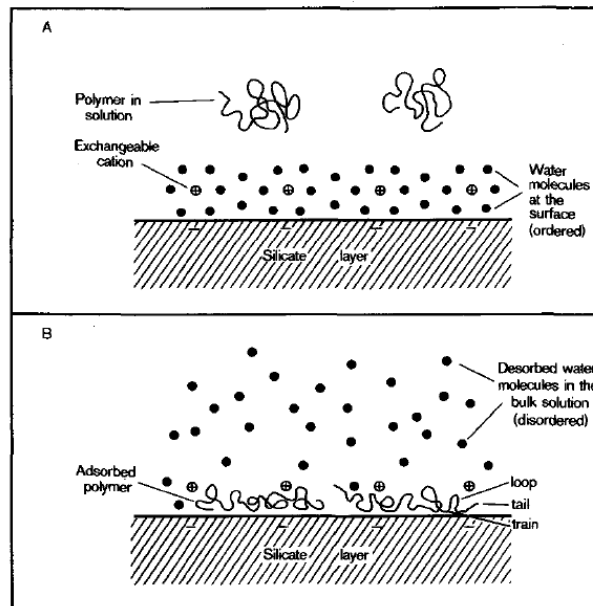


Figure 4. Diagram illustrating the desorption of numerous water molecules from a clay surface during the adsorption of an uncharged linear polymer, leading to a net gain in entropy by the system. Theng (1982).

Adsorption of natural biopolymers have been used and thoroughly researched in the soil and mineral industry for several uses, such as the biostability of soil organic matter Schloesing (1874).

There are several kind of forces related to adsorption effect. Oliveira (1997) explains the most important of these: attractive Van der Waals forces, hydrogen bonds, electrostatic double-layer forces, hydrophobic interactions, short-range repulsive forces, steric interactions, ion bridging and hydration pressure.

Natural polysaccharides, such as starches, dextrans and guar gums that are widely used in mineral flotation adsorb through the interaction with mineral surface metal-hydroxylated species. Based on the fact that the surface metal hydroxyl groups with a lower acidity exhibit stronger interaction with the natural polysaccharides, the nature of the interaction

is likely an acid/base interaction. The extent of the acid/base interaction probably determines whether the adsorption is hydrogen bonding or chemical complexation Liu et al. (2000).

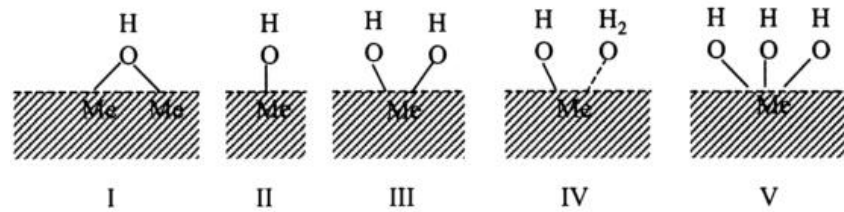


Figure 5. Mechanism of adsorption of a dextrin on the surface of a mineral by acid-base interaction. Case of a basic surface: complexation. Liu et al. (2000)

In Vissac (2013) is undertaken a deep analysis about these interactions, concluding that each kind of polysaccharide has a different manner of interaction to its shape, size and charge.

For instance, the relatively weakly charged, long (cellulose, amylopectin) and fairly rigid molecules would rather act by bridge flocculation, linking several mineral particles, a similar work that straw does in adobe bricks. On the other hand the more highly charged or short-lived molecules (amylose) would act by charge neutralization and total covering of the surface. This phenomenon would lead to repulsion or attraction between the particles depending on the ionic strength and pH conditions.

Conclusion

In default of laboratory test assays, we can confirm that *Ceratonia siliqua* is an important specimen available for use in the adaptation of existing recipes for earth stabilization extract.

A future line of research in this field could include the study of the ageing properties of these combinations, and in the case of positive results, undertake an economic and ecological feasibility study of this material.

The extraction and pretreatment of *Ceratonia siliqua* can be differentiated based on soil type and especially the different clay crystallographic facies presented. The study of pH and cation exchange capacity can provide references for the degree of reactivity and the type of links that may be obtained. So surely tannins can react very well on montmorillonite as happens in lateritic soils, on the other hand current polysaccharides can generate bonding bridges and longer chains in kaolinite. It would therefore be interesting to know how natural *Ceratonia siliqua* extract would react with each clay. This leaves settled the laboratory basis of the research which has just begun.

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2.2. ARTÍCULO 2

J. R. Clausell, C. H. Signes, G. B. Solà, and B. S. Lanzarote, M. Inclán
“Improvement in the rheological and mechanical properties of clay mortar after
adding Ceratonia Siliqua L. extracts,”*Construction and Building Materials* , vol.
237, p. 117747, 2020

<https://doi.org/10.1016/j.conbuildmat.2019.117747>

Improvement in the rheological and mechanical properties of clay mortar after adding *Ceratonía Siliqua L.* extracts.

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Construction and Building Materials, 237 (2020) 117747

<https://doi.org/10.1016/j.conbuildmat.2019.117747> 0950-0618/2019 Elsevier Ltd. All rights reserved.

Article history:

Received 18 July 2019

Received in revised form 19 November 2019 Accepted 28 November 2019

Abstract

In this study we have investigated the use of one of the components of the carob tree, *Ceratonía Siliqua L.* polyphenols, as an additive to improve the rheological properties of clay mortars used for sustainable construction. To obtain comparable results, soils with different clay bases have been used as raw materials. The most relevant results were the increased fluidity of the material and, consequently, a reduction in mixing water, especially in kaolinite soils with a decrease of 57.16 w% and in smectitic soils with 66.25 w%. Significant improvements in the mechanical behaviour of the mortars supplemented with the polyphenols derived from carob were observed.

keywords: *Ceratonía Siliqua L.*, Carob tannin, clay mortar, Vegetal additive.

1. Introduction

The carob tree (*Ceratonia Siliqua* L.) is common in the Mediterranean Basin. Its by-products are essentially centred on the use of its sugars for the production of gums in thickeners, stabilisers, emulsifiers, or gelling agents. Depending on their applications and methods of extraction, some of the by-products generated may have application in construction [1].

There is well-founded documentation on the effect of polyphenols on clay compounds, also in the field of construction, and their applicability varies depending on the origin of the polyphenol and its typology [2].

The objective of this study is to determine the effect of these polyphenols derived from the carob tree in different states, applicable as additives to clay mortars, while trying to emulate various scenarios sufficiently well to obtain a conclusive result regarding their potential and reactivity.

The additive and extraction method that offers the greatest possibilities according to the inventory presented in [1] is carob tannin added in dry mass with the goal of reducing the water in the mixture.

2. Materials and methods

2.1. Materials

2.1.1. *Ceratonia Siliqua L.*

The product under investigation is the fruit of the *Ceratonia Siliqua L.*, known as the carob tree. The carob by-products used in this study were provided by the company, FruitSecs SL, whose intensive carob plantations are located in Spain, close to Alcudia (Valencia, Spain). For the extraction of the carob tannin additive (hereinafter CT), raw material was acquired from the dry and crushed carob pods in different sizes (1 cm+/-3 mm and 0.2 cm+/-1 mm) (Fig. 1) to test the extraction yield of the tannin. Only the pod was used because of its high content of hydrolysable tannins, such as gallic acid, which can be used as an additive [3].



Fig. 1. Sample of carob pod. Particles 1 cm in size.

2.1.2 Soils

Three types of clay soils as different as possible (**Table 1**) were selected for the analysis of the change in their mechanical behaviour after adding additives obtained from the fruit of the carob tree.

An illitic soil (hereinafter IS), a kaolinitic soil (hereinafter KS), and finally, a smectitic soil (hereinafter SS) were selected.

The IS was supplied by the company Arcillas Atomizadas de Onda (Atomised Clays of Onda, Spain), with the product name, GRES ONDAGEN. The extraction quarry is located near Villar del Arzobispo (Valencia, Spain).

The KS was supplied by the company, VICAR de Manises (Valencia, Spain), with the product name CFK-PO. The extraction quarry is located in the area of Losa del Obispo (Valencia, Spain).

| Components | IS | SS | KS |
|-------------------|-----------|-----------|-----------|
| % Smectites | 0.00 | 34.61 | 0.0 |
| % Illitic | 60.71 | 24.09 | 5.4 |
| % Chlorite | 6.23 | 33.93 | 0.0 |
| % Kaolinitic | 33.06 | 7.38 | 94.6 |

Table 1. Mineralogical analysis provided by suppliers.

The SS was supplied by the company MYTA Grupo Samca (MYTA Samca Group) with the product name Esmectita Activada (activated smectite). The extraction quarry is located near Teruel (Spain).

2.1.2. Aggregates

A quartz aggregate with a maximum size of 2 mm, free of silt, and with a standardised granulometric curve, supplied by NORMENSAND, GmbH (Beckum, Germany), was used to obtain a well graded mortar.

The use of this sand is important for various reasons. Firstly, it is inert and therefore has no ion exchange capacity that could disturb the soil-additive reaction. It also provides a

precise standardised granulometric curve for the mortar compression strength test according to UNE-EN 1015-11:2000/A1:2007 [4].

2.2. *Methods.*

2.2.1. *Soil characterisation: CEC and FAAS analysis.*

The CEC (cation exchange capacity) test was conducted to quantify the total amount of interchangeable cations in the raw materials. This test shows the number of cations that can be released or retained from the surface of the soil colloid towards the medium. The ammonium acetate pH=7 method was used [5].

Quantification was carried out specifically on those free cations that were of interest to the study in Ca^{2+} , K, and Na, using the FAAS method (flame atomic absorption spectroscopy), and according to USDA (United States Department of Agriculture) standards, using a Sherwood photometer.

2.2.2. *Protonation test*

Various potentiometric valuations were completed to determine the protonation constants of the carob tannin and its greatest load by pH. For this purpose, the first test was carried out on standard gallic acid SIGMA-ALDRICH $\geq 99\%$ (HPLC). Protonation constants of gallic acid and commercial tannin are determined by potentiometric titrations of solutions in 0.15 M NaCl at 25 ° C. These measures were done by Inclán M. and García-España E., in the Supramolecular Chemistry Group of the University of Valencia, following the protocol developed and previously described in [6]. These constants were used to calculate the species distribution diagrams and the load values versus pH.

2.3. Additive preparation. CT extraction.

The fruit of *Ceratonia Siliqua* L. has a wide variety of polyphenols such as proanthocyanidins, ellagitannins, and gallotannins [3]. Within the family of polyphenols, some are soluble and others insoluble. The highly polymerised proanthocyanidins, for example, are insoluble in water while gallic acid, the polyphenol extracted as an additive, is soluble in water. The extraction methodology used absorbent and cationic resins [7]. The resins were a SEPABEADS SP207 Mitsubishi absorbent resin and a RELITE EXC14 cationic resin.

2.4. Mortar and test preparation.

Mortars were prepared with 30% clay and 70% aggregate by weight. The clay content in the mixture was higher than in commercial mortars (15-20%) such as to enhance the reaction of the clays and not the mechanical suitability of the mortars. The mortars were prepared according to DIN specifications 18947:2012-08 [8] y UNE-EN 1015-2:1999 [9]. In addition to the applicable regulations, the methodological prescriptions proposed in [10] were followed, the modifications are justified by the nature of the clay mortars, whose preparation and conditioning for testing is different from those described in conventional regulations. The mixture was left prepared and moistened up to 24 hours prior to its use to achieve a complete behaviour of its properties, especially its plasticity. The specimens were prepared for the mechanical tests in accordance with the regulations shown in **Table 2**. Trials were proposed to identify their improvement in strength, both superficial and structural. Since the clay mortar does not need high ambient humidity for hardening, the samples were kept in a controlled environment chamber under constant conditions of 20°C and 50% RH and broke at 90 days.

| Tests | Regulation | Dimensions (mm) | Samples | | | | | |
|-------------------|---|--------------------|----------|----------|----------|----------|----------|----------|
| | | | KS /B | KS/ T | IS /B | IS /T | SS /B | SS /T |
| Compression | UNE-EN 1015-11:2000 [4] and DIN 18555 [18] | 40x40x160 | 6 | 6 | 6 | 6 | 6 | 6 |
| Flexotraction | UNE-EN 1015-11:2000 [4] and DIN 18555 [18] | 40x40x160 | 3 | 3 | 3 | 3 | 3 | 3 |
| Wide disc wear | UNE-EN 13748-1:2005 Terrazzo Paving Stones [19] | 80x80x25 | 3 | 3 | 3 | 3 | 3 | 3 |
| SAET Erosion | Swinburne Accelerated Erosion Test (SAET) [20] | 80x80x25 | 3 | 3 | 3 | 3 | 3 | 3 |

Where:

For soils: Kaolinitic soil (KS); Illicit soil (IS); Smectitic Soil (SS)

For additives: Without additive (B); with additive (T)

Table 2. Mechanical tests chart.

2.4.1. Determination of the optimal water content %H₂O

With the materials completely dry, 800 g were prepared in the proportions indicated, and placed in the mixing bowl of a Proeti 085 mortar mixer. Given that the object mortars vary in components and additive percentages according to the nature of each clay, it was decided to set a practical constant that would serve as a reference and thus be able to produce comparable results. A fixed fluidity value of 160 mm ± 5 mm was set in the test according to the norm [11]. The mortar was tested in the shaking table according to the norm [12] in order to adjust the water content to this fixed fluidity value.

2.4.2. Determination of the optimal CT %

With the percentages of mixing water controlled, the percentage of CT to be added to each mortar was determined. Considering the pH of the medium according to the protonation test for the CT, the pH of the mortar was increased with the addition of NaOH to reach the set alkalinity of the solution. The pH of the mortar was measured according

to UNE 77305 [13] with a Multi 340i WTW meter before, during, and after the addition of NaOH, maintaining the mixing hydration percentages until reaching the appropriate NaOH % for each mortar. A test grid was prepared (**Fig. 2**) to experiment with the CT %. The test range of CT % was set a priori between 0.5-1% by mortar weight, compared to the superplasticising cement additives of existing commercial products.

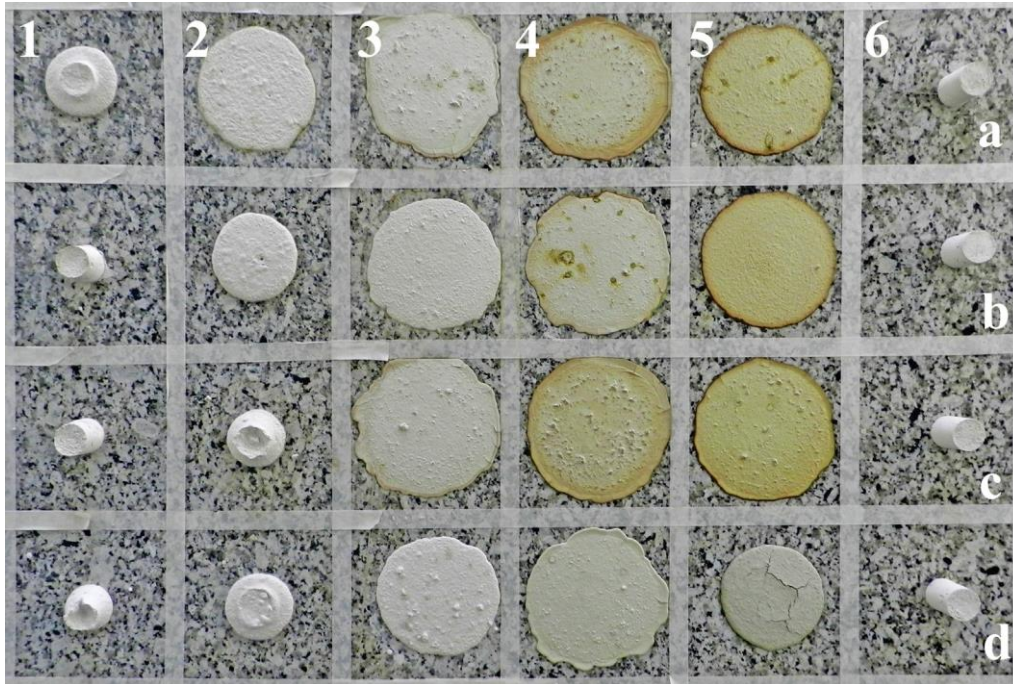


Fig. 2. Consistency test grid to determine the optimum CT %.

In each of the grid spacings, a consistency test was performed to visually determine the best reactivity of the additive in the mixtures, and thus verify at a mechanical level the macroscopic response of the cation exchange between the soil and the CT. For each of the tests, 160g of mixture was prepared, adding the demi H₂O (w/w%) according to the fluidity test with the corresponding NaOH (w/w%) of each soil. The mixture was introduced in the mixer and the mortar was poured into a cylindrical stainless-steel mould sized h=6 cm and ø=4 cm. The sample was compacted with a chopping bar and its upper face was levelled to obtain the same material content in all tests. Finally, the mould was

removed, obtaining a different fluidity in each test. To categorise acceptance or rejection values between the different reactions of the additive (**Fig. 3**) on the clay mortar, several criteria were established under constructive premises of workability and appearance, and thus to discern between APT, NOT APT, or IDEAL reactivity. The optimum CT dosage was established as the result of the maximum dispersion of the mortar and without showing any of the following failures: burns or significant pictorial changes (**Fig 3a**); mortar segregation (**Fig 3b**); significant mortar dispersion (**Fig 3c**).

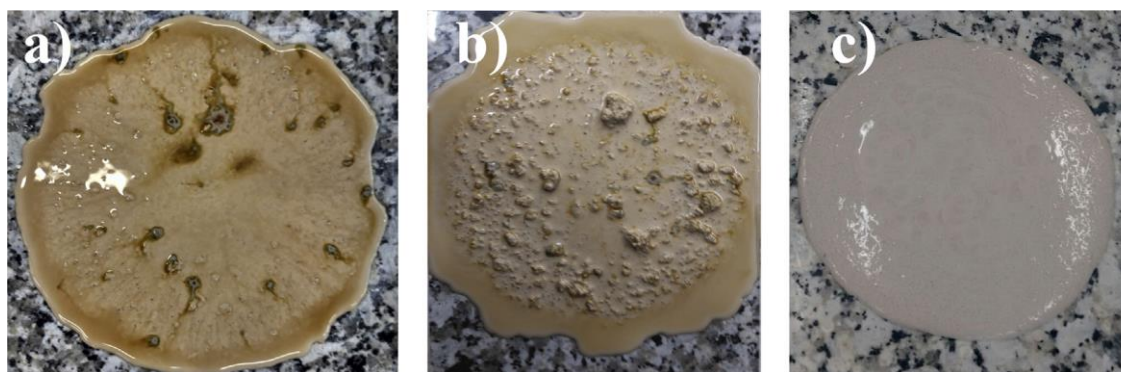


Fig. 3. Types of dispersion.

2.4.3. Determination of optimal H₂O % reduction

For the determination of the % reduction of mixing water, the fluidity test was repeated on a shaking table, this time adding demi H₂O with compensated pH and %CT according to the results of the reactivity grid.

The working methodology for obtaining the % reduction in mixing water is set out in **Fig.4**.

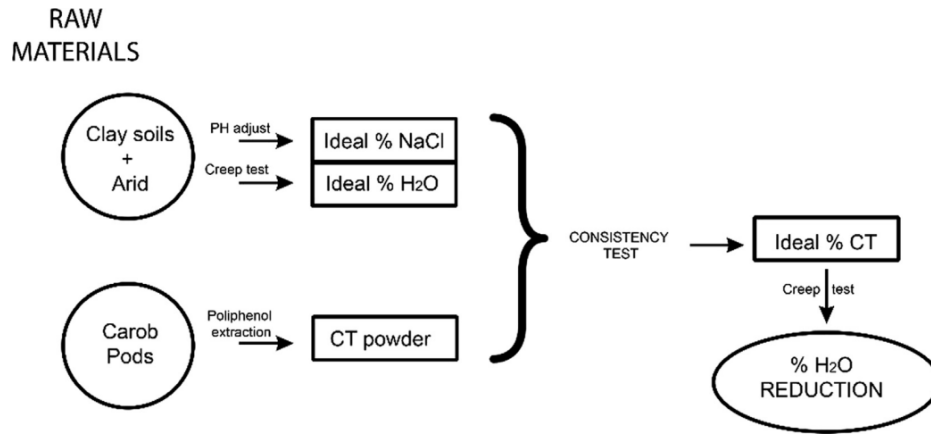


Fig. 4. Workflow scheme.

3. Results

The results of the tests carried out on the raw materials and on the specimens prepared for the mechanical tests are presented below.

3.1. Soil characterisation tests: CEC and FAAS analysis

With the results of the CEC and FAAS tests shown in **Tables 3** and **4**, the high reactivity capacity of KS and SS was confirmed, congruent with the reactions in the consistency test. In the FAAS test we were able to obtain information on the available and interchangeable cation provided by the CT and K^+ .

| Sample | CEC (meq/100g) |
|--------|----------------|
| SS | 26.17 |
| KS | 28.18 |
| IS | 10.07 |
| CT | 4.76 |

Table 3. CEC results.

| Sample | Na ⁺ (mg/l) | K ⁺ (mg/l) | Ca ²⁺ (mg/l) |
|--------|------------------------|-----------------------|-------------------------|
| SS | 394.86 | 0.07 | 6.54 |
| KS | 10.29 | 7.95 | 27.91 |
| IS | 405.22 | 10.79 | 4.68 |
| CT | 8.14 | 749.57 | 24.19 |

Table 4. FAAS results.

3.2. CT protonation test.

Following the protonation test, three acidity constants were determined for standard gallic acid. The first, with a value of pK_a=4.2, corresponds to the deprotonation of carboxylic acid. The second and third, with values of pK_a=8.6 and 10.9, respectively, can be assigned to the deprotonation of two of the phenol groups (**Fig. 5**). The charge group -3 only begins to predominate at pH values greater than 10.9. These constants were used to calculate the species distribution diagrams and the load values versus pH. Protonation constants of gallic acid and commercial tannin were determined by potentiometric titrations of solutions in 0.15M NaCl at 25°C. These measures are based on the Supramolecular Chemistry Group of the UV (University of Valencia), following the protocol developed and previously described in [6].

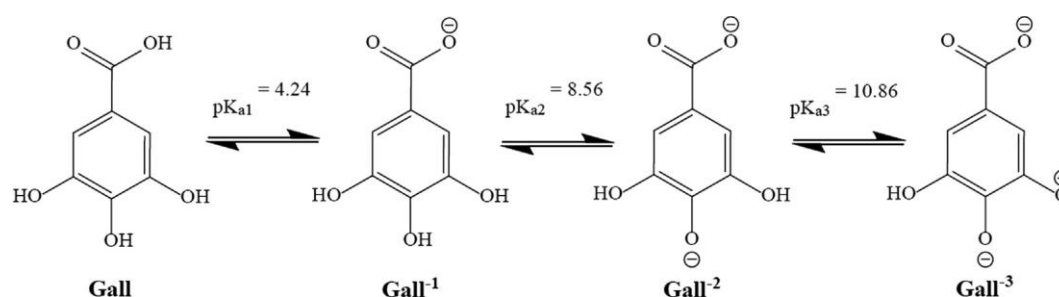


Fig. 5. Interpretation of GA protonation.

Given that CT is a complex mixture of oligomers of variable loads and sizes, it is not possible to deduce specific values for the acidity constants from the potentiometric valuations. However, it was possible to adjust the data obtained to a model in which the presence of up to eight protonable centres and an average molecular mass of 962 g/mol^{-1} was assumed, which was calculated from the analysis and distribution of oligomers according to Pica R. [7]. Starting from this setting, the number of charges will vary against pH as shown in **Fig. 6**.

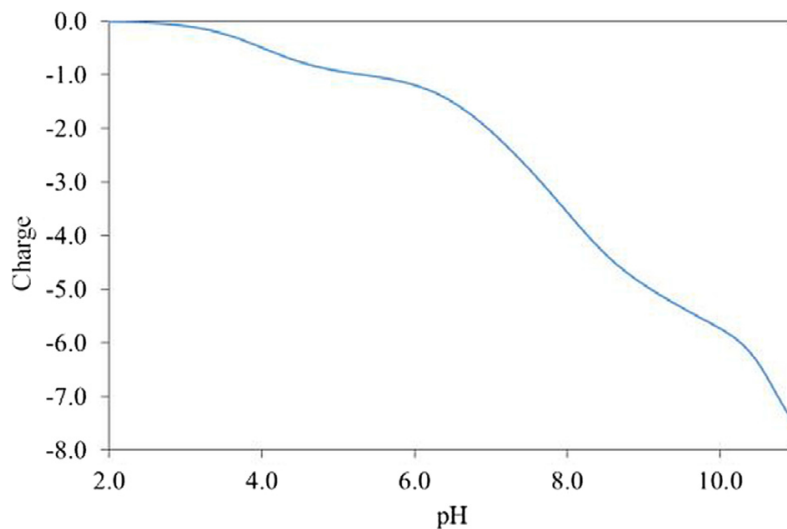


Fig. 6. Value of the load versus the pH for GA of the carob tree.

Analogous to the GA, it was only at pH values greater than 4.0 that a charge of -1 was reached. This is unsurprising given that the oligomers that make up the carob tannin all consist of a carboxylic acid unit that, in all cases, will have a similar pKa value. From pH 6.0 upwards, the value of the charge decreases fairly pronouncedly until it reaches a value of -6 at pH 10.0. From this point on, the load decreases rapidly from -7.5 at pH 11.0.

3.3. *Optimal hydration percentage*

The values of the hydration for each mortar for a fluidity value of $160\text{mm} \pm 5\text{mm}$ are shown in **Table 5**.

| Soil | IS | KS | SS |
|-----------------------|-----|-----|-----|
| H ₂ O w/w% | 15% | 24% | 80% |

Table 5. % Mixing water for 160mm fluidity.

3.4. Optimal %CT

The results of the pH measurement of the various clay mortars are shown in **Table 6**.

| Sample | pH | |
|--------|-----------------------|------------------------------|
| | H ₂ O demi | H ₂ O demi + NaOH |
| IS | 10.05 | 11.39 |
| KS | 8.67 | 11.62 |
| SS | 10.40 | 11.36 |

Table 6. pH of the sample and mixtures.

The clay mortars reacted differently to the addition of CT, producing interesting effects in various additive percentages. It was found that within the additive range fixed on the basis of recommendations for the use of commercial products, the percentage of additive was too high, in many cases creating overcoagulations in the mortar. A much smaller amount was sufficient to satisfactorily disperse the clays (**Table 7**).

| Type of mortar (Samples of 160 g) | H ₂ O w/w% (H ₂ O/Mortar) | NaCl w/w% (NaCl/H ₂ O) | Additive w/w% (CT/Mortar) |
|--------------------------------------|--|--------------------------------------|------------------------------|
| IS | 15 | 0.55 | 0.09 |
| KS | 24 | 0.55 | 0.045 |
| SS | 80 | 0.55 | 0.015 |

Table 7. Percentages for optimal reaction.

Interesting heterogeneous dispersion effects were observed in some samples, with aggregate granules clearly clustering as coagulation or, in some cases, dispersing the mortar with excessive and counterproductive mechanical segregation (**Fig. 7**).

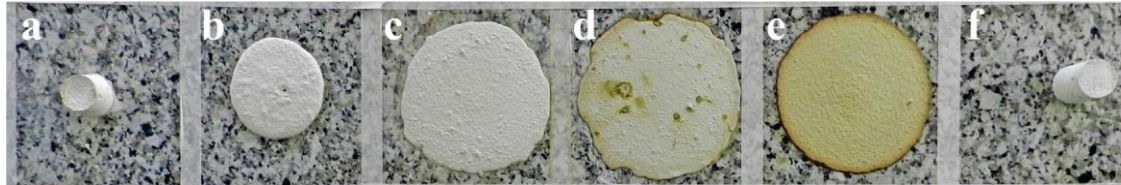


Fig. 7. Reaction table.

3.5. Reduction of mixing H₂O%

After the fluidity test, we obtained a different mixing water reduction in each of the soils tested as indicated in **Table 8**.

Note that the clays showing greatest reaction with the additive, and therefore having a greater reduction in mixing water percentage, were those that needed most water to acquire an adequate workability in their natural state. This was the case of the SS, which required a significant amount of mixing water. When using clay soils as raw material for renders, it is extremely important to reduce the mixing water as much as possible to avoid excessive shrinkage, especially in the case of clay coatings with a smectitic composition that requires a greater percentage of mixing water.

| Soil | H ₂ O w/w% without CT | H ₂ O w/w% with CT | Reduction of mixing water (%) |
|------|-------------------------------------|----------------------------------|----------------------------------|
| IS | 15 | 11 | 26.66% |
| KS | 24 | 11 | 54.16% |
| SS | 80 | 27 | 66.25% |

Table 8. Mixing water reduction.

3.6. Mechanical tests

3.6.1. Compression and flexural test

Tests show that the compressive strength increases significantly with a reduction of the mixing water in the mortars.

One of the significant reasons is the increase in the final density of the specimen, as in the case of KS with added tannin, whose density increased by 10% (**Table 9**).

| Soil | ρ (mg/m ³) with CT | ρ (mg/m ³) without CT | $\Delta\rho$ (%) |
|------|--|---|------------------|
| KS | 2.10 | 1.90 | 10 |
| IS | 2.13 | 2.01 | 6 |
| SS | 1.62 | 1.49 | 8 |

Table 9. Increased density ρ .

A further reason for the improvement of the compressive and flexural strength is the arrangement of the microstructure of the clay – the sorting of the clay, as we saw in **Fig. 8**, with the change in its electrical charge, as opposed to the case of having an aleatoric arrangement with positions that do not favour the transmission of compressive loads (See **Fig.9**).

The best results were obtained for the IS due to the basic properties of the clay. Nevertheless, the soil that showed the greatest improvement due to the presence of the additive was the KS.

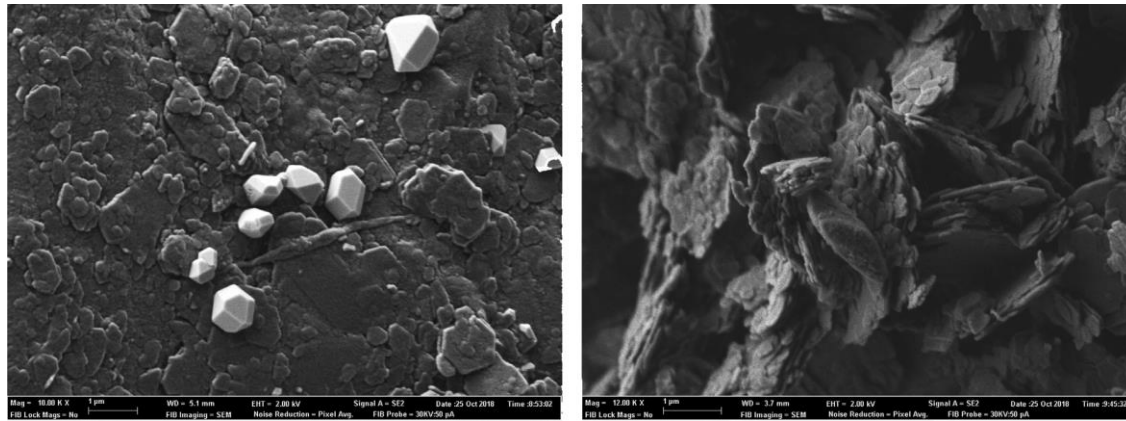


Fig. 8. Left: Dispersed structure. Dry KS mortar with CT. Right: Flocculated structure.

3.6.2. Wide disc and SAET wear test

For the abrasion tests, it was observed that the results follow the same pattern as in the compression and flexural tests, with the KS and IS displaying the greatest increase in strength after the addition of the CT (**Fig. 10 right**).

In the case of the SAET test, the results of the IS were not as good as those of the CT. This is because of the low quantity of silts in proportion to the granulometric skeleton

reduced its stability in humid conditions or water wear (**Fig. 10 left**).

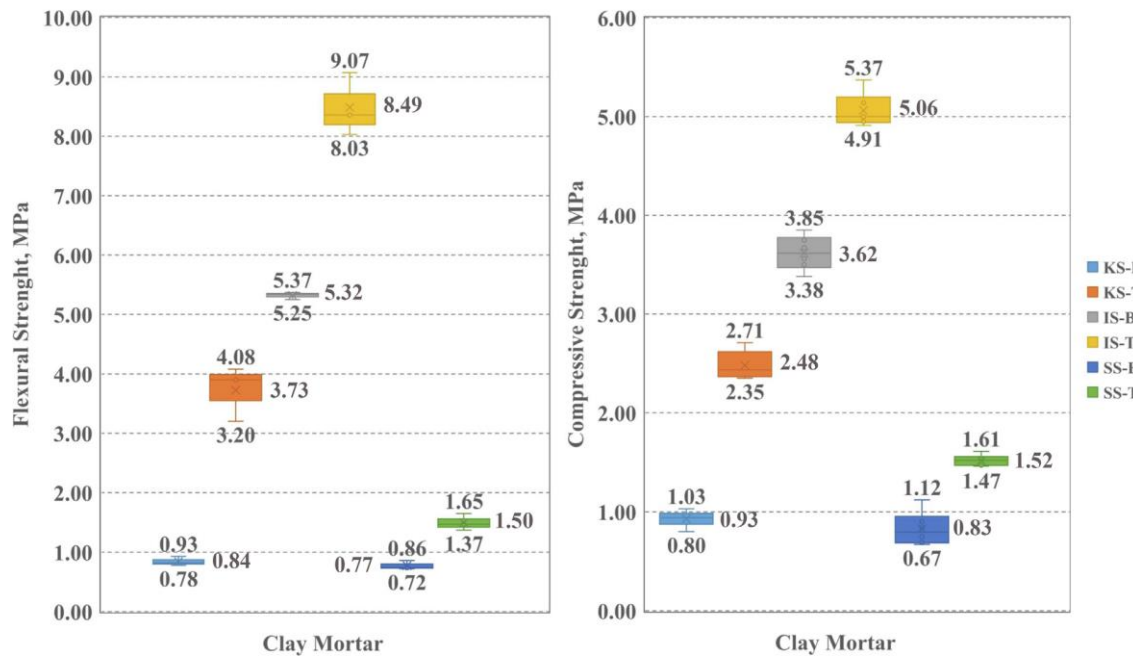


Fig 9. Left: Flexural strength test results. Right: compression strength test results.

Another interesting result of the SAET test is that, in the case of the SS, the SB specimen obtained a better result than the ST. This is because the clay particles in the SB specimen still maintain their expansive effect with the Na^+ cations that bind the clay sheets (which were detached when the tannin was added). This expansive effect creates an impermeable film that easily repels the shock of the water jet when the clay sheets clump together. The results in the wide disc test can be said to be parallel, although the improvement in the results is more modest, given that it is a test focused on products with greater surface strength.

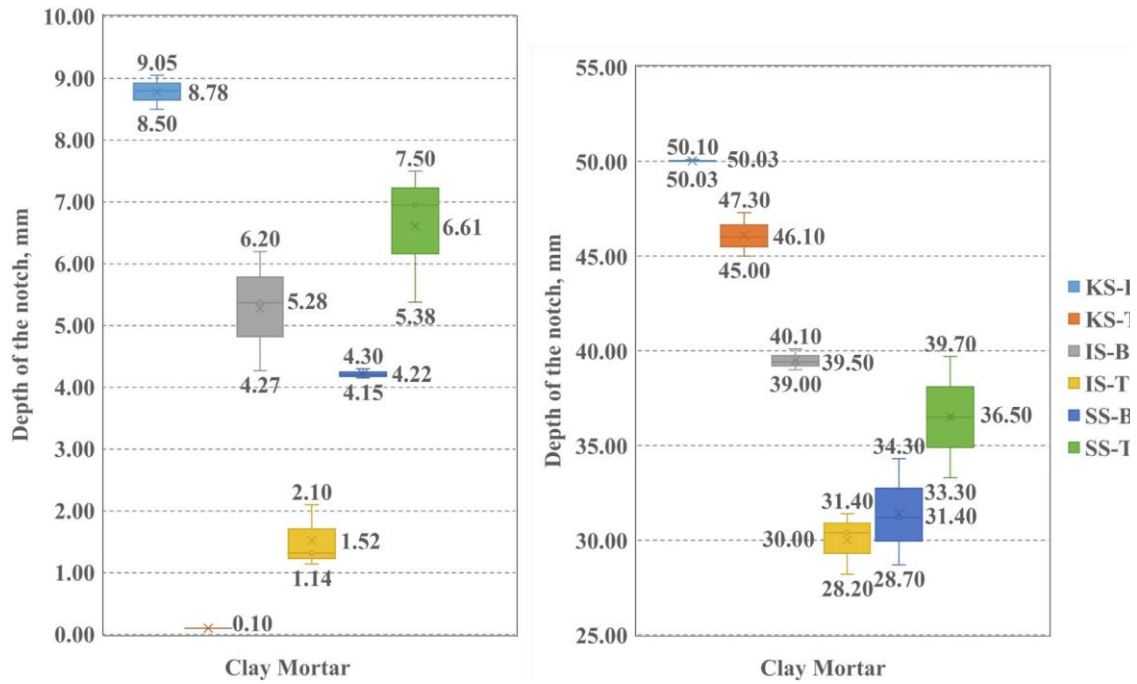


Fig. 10. Left: SAET test results. Right: Wide disc erosion test results.

4. Discussion

The behaviour of the CT on various clay particles has been tested and good diverse results were obtained in each case.

To obtain a conclusive value of the reactivity between the CT and the soils, the parameters that must be known and controlled are the pH of the mixture and its CEC (beyond its mineralogical or chemical composition). In the case of pH, the reason for its determinant influence on the CT-clay relationship has been defined.

The protonation test confirms that the CT charge reaches -8 at alkaline pH 10-11, which increases the possibility of a significant exchange of its readily available cations such as K^+ .

At high pH values the hydrogen from the edges of the sheet continues to have weak bonds and is easily lost, which can modify the charge at the edges and ultimately favour the

adsorption of CT molecules. Some of the molecule's negative charge neutralises the positive charge of the aluminium, while the free negative groups of the molecule increase the negative charge at the edge, according to Van Olphen [14]. This example can be given in the case of gallic acid bound by K^+ with the edges of the wafers. This adsorbed CT molecule has the advantage of special geometry that enables it to embrace the cation in order to neutralise it (binders). Therefore, in addition to being able to form compounds at the edges of the clay sheets, they can be joined simultaneously with free flocculant cations such as Ca^{2+} (**Fig. 11**), and so produce a double dispersant effect [15]. It is for this reason that not all tannins have the same dispersing effect; the effect depends on whether they are trivalent or monovalent phenolate groups.

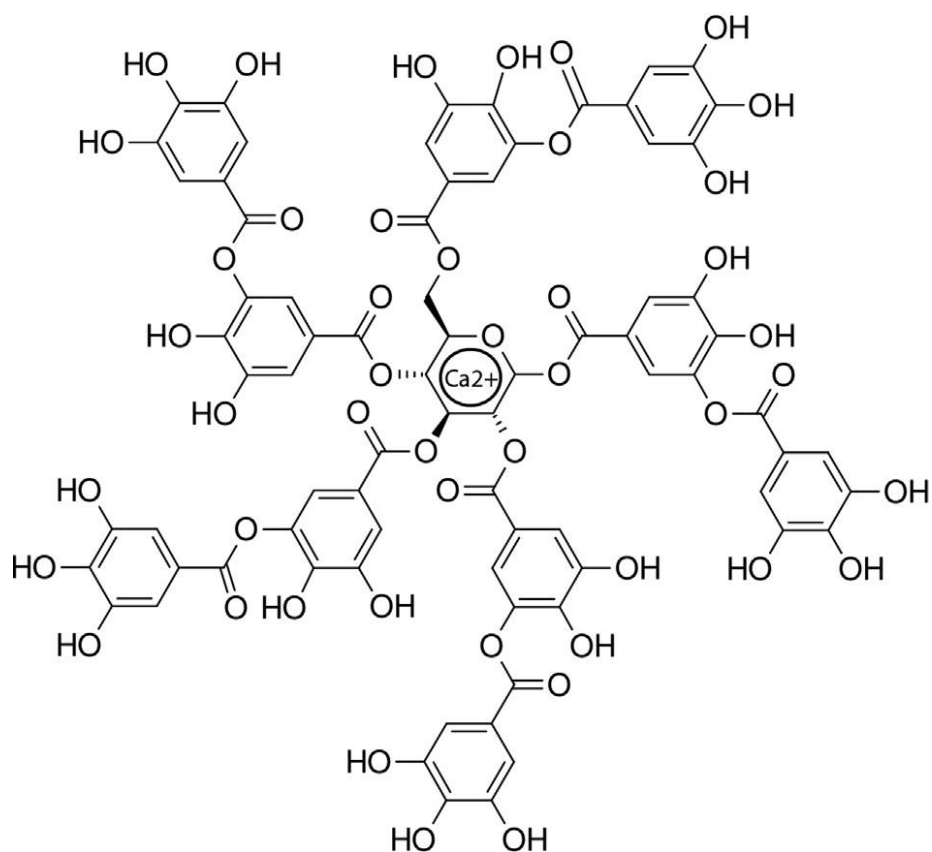


Fig. 11. Possible flocculant cation neutralisation scenario.

The CEC reflects the potential of the CT to cede these monovalent K^+ cations. Clays have interlaminar bipolar cations such as Ca^{2+} which are usually coagulants [16]. These are easily substitutable by available monovalent cations provided by the CT (such as unipolar cations of K^+). If this substitution occurs, a mismatch causes modification in the interlaminar distance of the clays which weakens the static attraction of variable force (depending on the nature of the clay and its components). If these Van der Waals forces are insufficient to keep the sheets together, the clay disperses, and in this case, the dispersion effect of the CT on the clays would be a dispersion by electrostatic repulsion.

5. Conclusions

The aim of this research has been to replace chemical additives used in conventional construction with more sustainable additives of plant origin. For this reason, it is important to emphasise that both the final product and its development must have the lowest possible cost in terms of energy, while avoiding the inclusion of chemical additives in the soil that would nullify its main advantage, which is its possible reusability. Carob tannin has been shown to be a product with significant potential as an additive for clay-based construction elements, especially as a superplasticizer as shown in [Video 1](#) [17], and so opening the door to market possibilities that were until now reserved for chemical additives.

This dispersing effect of the clays may be useful in the proposed field of study of wall renders, but it seems to have greater potential in the field of loadbearing construction elements, given the significant reduction in the mixing water requirements of mortars or concretes, and having a direct positive impact on the results of all mechanical tests carried out on strength to compression, flexion, and erosion.

Acknowledgements

The authors would like to give special thanks to the Geotechnical Department of the UPV (Universitat Politècnica de València) for its vital support in this research. Thanks are due to PhD. Victoria Lizama Abad of the Department of the Institute of Food Engineering for Development and PhD. Jaime Primo Millo for support in the polyphenol extractions, as well as to the bioconstruction cooperative Okambuva Coop, for the practical experimentation spaces, and finally to the IVE (Valencian Building Institute) for its assistance in carrying out tests.

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Romero Clausell, J., Quintana-Gallardo, A., Hidalgo Signes, C. Lanzarote Serrano, B.
Environmental evaluation of a self-compacted clay based concrete with natural
superplasticizers. Mater Struct 54, 20 (2021).

<https://doi.org/10.1617/s11527-020-01586-6>

ENVIRONMENTAL EVALUATION OF A SELF-COMPACTED CLAY BASED CONCRETE WITH NATURAL SUPERPLASTICIZERS

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Materials and Structures (2021) 54:20

<https://doi.org/10.1617/s11527-020-01586-6> SpringerLink. All rights reserved.

Article history:

Received: 29 July 2020 / Accepted: 17 November 2020 Ó RILEM 2021

Abstract

Cement concrete is the most widely used construction material worldwide due to its favourable mechanical characteristics. However, it is responsible for 8% of the total carbon emissions in the world, which are generated mainly during the production of clinker. Due to that fact, finding alternatives to cement for some applications in which it is not strictly needed should be a priority. In this study, a self-compacted clay-based concrete with natural superplasticizers based on natural tara tannins is presented. The main objective of the study is to determine if this clay-based concrete can be a sustainable alternative to conventional cement concrete as the main component in structural slabs. The methodology of the study is divided into two parts. First, the self-compacting clay concrete is characterized to determine its mechanical properties. Secondly, a comparative Life Cycle Assessment is conducted to determine the difference between the impacts generated by one square meter of self-compacting cement concrete and one of self-compacting clay concrete. The characterization of the material showed that this self-compacting clay concrete is suitable for some building elements such as structural slabs while avoiding the energy consumption needed to produce

conventional concrete. The environmental impact results showed that using self-compacting clay concrete instead of the cement-based material decreases 90% of the carbon emissions and 80% of the overall environmental impact. After the completion of the study, it can be stated that the presented material is a sustainable alternative to conventional concrete for building structural slabs.

Keywords: earth construction, clay concrete, self-compacted clay-based concrete, Life Cycle Assessment, sustainability.

1. Introduction:

Cement concrete is the most used building material in the world [1]. Its favourable mechanical properties paired with its affordable price makes cement concrete the easy choice for building any kind of structure. However, an immense quantity of CO₂ is emitted into the atmosphere during its production process. The production of cement concrete by itself generates 8% of the total carbon emissions in the world [2]. The reaction between limestone and clay to produce clinker is responsible for most of those emissions. Reducing the worldwide carbon emissions is one of the most pressing challenges society faces. Since the first climate emergency declaration, the effects of climate change have become apparent, and mitigating them is indispensable to protect the ecosystems [3]. Therefore, finding alternatives to conventional cement concrete is key to a sustainable future.

One of those alternatives is soil. Soil is used as a construction material by approximately half of the global population, with highly diverse techniques. It is the most abundant natural construction material in most regions in the world. The use of soil as a construction material is a constant in low-resource areas, and it is becoming increasingly popular in contemporary sustainable construction [4]. However, due to the lack of standardization in the different parts of the world, it is difficult to scale up its production.

Now more than ever, research on techniques to take advantage of the benefits of soil worldwide is a subject of great interest.

In this study, a soil-based concrete is presented. This concrete replaces cement by an illitic soil in its formulation. This concrete, named Self Compacting Clay Concrete (SCCC), has been thought as an alternative to cement concrete in some favourable scenarios. This technique has been studied previously, using chemical superplasticizers [5] or drying accelerators such as lime or cement [6] to improve the formwork removal. The idea behind this study has been to achieve a similar effect to that of the chemical superplasticizers in reducing the amount of water, but using a natural compound, gallic acid (hereinafter GA) from tara tannin (*Caesalpinia spinosa*).

The objective of this study is to determine if SCCC can, in some cases, be used as a sustainable replacement of conventional concrete. The methodology followed to achieve that goal can be divided into two parts. The first one is the mechanical characterization of SCCC. Evaluating the mechanical behaviour of the material gives the possibility of knowing which building typologies would be adequate for its use. After characterising the material, the second part consists of a comparative Life Cycle Assessment (LCA) between SCCC and conventional self-compacting concrete. By this comparison, it is possible to assess the actual existing difference between the impacts that both materials have over the environment. The LCA methodology has been applied to other earth-based construction systems on previous occasions [7]. The working methodology for the final LCA results is set out in (Fig. 1).

Figure 1. Methodology.



2. Materials

2.1. Soil

The illitic soil (IL) was provided by the company PYBC from Ribarroja del Turia (Valencia, Spain). The following characterisation tests were performed to establish an adequate dosage and reactivity evaluation strategy. The two goals of the characterisation tests (Table 1) were: to study the granular skeleton of the material either by density or particle size and to understand the chemical composition of the material. The characterisation was complemented using a sedimentation test to obtain complete information about the real percentage of clay and silt between the fine particles. A granulometric laser analysis was performed with a Mastersizer2000 Hydro 2000SM (A) and one minute of ultrasound as a dispersant agent.

A major component analysis was made through the quantitative interpretation of an X-ray diffraction test (XRD) to identify the crystalline phases of the object soils, using the Diffrac Plus Xrd Commander Software system with the JCP3 database of the ICDD (International Centre for Diffraction Data). To the basic dust pattern test were added the oriented clay aggregates, the glycol aggregates diagram/pattern, and the calcined aggregates diagram at 550°C.

2.2 Aggregates

The aggregates were supplied by La Torreta, a quarry in Castellón de la Plana (Spain). Dolomitic limestone and washed crushed stone aggregates were used so that the fines did not interfere in the proposed granulometric curve. The quarry provided data related to the properties of each aggregate to adequately adjust the dose needed (Table 2).

Table 2. Soil and aggregates properties.

| Soil Parameters | Unit | Soil | |
|--|------|---------------|-------------|
| Grain size | | | |
| Gravel (> 5000 μm) | % | 0.0 | |
| Sand (63–5000 μm) | % | 0.3 | |
| Fines (< 63 μm) | % | 99.7 | |
| Silt | % | 45.6 | |
| Clay | % | 54.1 | |
| Atterberg limits | | | |
| Liquid limit (W_L) | - | 17.7 | |
| Plastic limit (W_P) | - | 12.5 | |
| Plasticity index (PI) | - | 5.2 | |
| Soil classification - USCS | - | CL-ML | |
| Specific gravity (G_s) | - | 2.712 | |
| Calcium carbonate content | % | 11.0 | |
| Organic matter content | % | 0.02 | |
| <hr/> | | | |
| Aggregate Parameters | | AF-0/2 AF-0/4 | AG-4/11 |
| <hr/> | | | |
| Granulometry: Category (UNE-EN 12620:2003+A1:2009) [35] | | G_F 85 | G_C 90/15 |
| Fines, % (< 0.063 mm) (UNE-EN 933-1:2012) [36] | | ≤ 10 | ≤ 1.5 |
| Particle shape: elongation index, % (UNE-EN 933-3:2012) [37] | | - | ≤ 15 |
| Sand equivalent (UNE-EN 933-8:2000) [38] | | ≤ 75 | - |
| Resistance to fragmentation (LA), (UNE-EN 1097-2:2010) [39] | | - | ≤ 35 |
| Specific gravity, Mg/m^3 (UNE-EN 1097-6:2001) [40] | | 2.700 | |
| Water absorption, % (UNE-EN 1097-6:2001) | | ≤ 1.5 | |
| Chloride content, % (UNE-EN 1744-1:2010) [41] | | ≤ 0.03 | |
| Total sulphur content, % (UNE-EN 1744-1:2010) | | ≤ 1.0 | |
| Total soluble sulphates, % (UNE-EN 1744-1:2010) | | ≤ 0.2 | |
| Light organic pollutants, % (UNE-EN 1744-1:2010) | | ≤ 0.5 | |
| <hr/> | | | |

2.3 Gallic acid

The tannin powder (product name Tan'Activ T80) was supplied and produced by Silvachimica SRL (Italy) from a raw material imported from Perú, Tara (Caesalpinia

spinosa). The product was supplied as dry powder packed in 5 kg packages. We chose this product for its high gallic acid concentration in the extract (reaching up to 53%) [8].

3 Methods

3.1 SCCC Dosage

The protocol for the SCCC dose is based first on the readjustment and adaptation of the granulometric curve of the raw material to ensure an improved cohesion of the components of the concrete; and, secondly, on the reduction of mixing water, and therefore, the minimisation of the probability of microcracking (due to the shrinking effect) that decreases resistance.

Taking into account the singular curves of each component of the concrete, the percentages of each were adjusted to recreate an ideal curve that was close to the proposal made by Bollomey [9], which proposes an ideal granulometric curve based on the consistency of the concrete. The procedure of recognising the reactivity of the GA with the clayey soil IL according to [10] was followed to obtain an ideal pH of the mixing water and an ideal percentage of the GA as a superplasticizer.

The mixing procedure for the SCCC concrete was as follows: the first step was to add NaOH to the water to compensate the pH according to the results of the reactivity test. The aggregates, the IL, and the GA additive were homogenised in a dry state with an electric concrete mixer model HGN 150 for 30 s. The pH compensated water was added to the concrete mixer with the rest of the components. The mixture was then mixed as a concrete for 2 min until the materials reach a homogenised consistency.

3.2 Mechanical tests

The tests carried out are displayed in Table 1. It was proposed that the mechanical tests for the SCCC mixture under research be compared with the results of a sample without

additive (henceforth NA), set as a constant to obtain the same flow index for both mixtures.

Table 1. Soil characterisation and mechanical tests.

| Soil characterisation tests | Regulations | | |
|-----------------------------|--|--|--|
| Grain size | ASTM D7928-17 [28] | | |
| X-Ray powder diffraction | ICDD (International Centre for Diffraction Data) | | |
| Sedimentometry | UNE-EN ISO 17892-4:2019 [29] | | |
| Attemberg limits | ASTM D4318 [30] | | |
| Soil classification – USCS | ASTM D2487 [31] | | |
| Specific gravity (G_s) | ASTM D854 [32] | | |
| Calcium carbonate content | ASTM D4373 [33] | | |
| Organic matter content | ASTM D2974 [34] | | |

| Mechanical tests | Regulations | Dimensions (cm) | Samples |
|---------------------------|---------------------------|-----------------|---------|
| Slump test | PNE-EN 12350-2 [11] | 20x30 (top) | 3 |
| | | 10x 30 (base) | |
| Shrinking test | DIN 18952 [42] | 7x10x70 | 3 |
| Compressive strength test | DIN 18555-5 [43] | ø15x30 | 9 |
| Flexural strength test | DIN 18555-5 | 10x10x030 | 9 |
| Young's modulus test | UNE-EN 12390-13:2014 [44] | ø15x30 | 9 |

3.2.1 Fluidity and shrinkage test

The flow test was performed just after the mixing process in the concrete mixer. The fluidity to be achieved was of a 15-16 cone or fluid-liquid consistency according to the PNE-EN 12350-2 [11] for self-compacting concrete.

The next step was to perform the shrinkage test. A sample from the flow test was collected, and a parallelepiped specimen mould of 7x10x70 cm was filled. A distance of 20 cm and the line defining that distance were marked on the upper plane of the specimen using a knife and a pattern.

The specimen was placed then on a glass surface, previously greased with oil, for three days at a temperature not higher than 20° C. The drying was then completed in an oven at 60° for 24 h. The dry shrinkage value was determined by the average of the two tests,

which should not differ by more than 2 mm, and given that the SCCC is applied in continuous structural elements, the average of the total lineal shrinkage should not exceed 1%.

3.2.2 Compressive and flexural strength

The specimens were prepared for the mechanical tests following the regulations shown in Table 1.

Given that the clay-based concrete does not need high ambient humidity for hardening, the samples were kept in a controlled environment chamber under constant conditions of 20 °C and 50% RH. The SCCC does not have any time-dependent stabiliser or hardener, so the break date was set to its full hardening state (Table 3) and hygroscopic equilibration of 28 days. The compression test was performed using a testing machine with a range of 2000 kN and a speed of 0.60 kN/s, with a servo-hydraulic control console for axial load application and Mecatest-16 data acquisition. Also, strain gauges type PL-60-11 were used with a gauge factor of $2.09 \pm 1\%$ and an HBM Spider 8 daq data acquisition module to measure microstrains (to obtain Young's and Poisson coefficient under the standard regulations). The flexural test was performed using a testing machine model CODEIN MC0-30 with a range of 2000 kN and a speed of 2,5 kN/s.

Table 3. Mineralogical composition interpretation.

| Components | Illitic soil |
|--------------|--------------|
| % Smectites | 0.00 |
| % Illitic | 55.71 |
| % Chlorite | 6.23 |
| % Kaolinitic | 38.06 |

4 Methodology for the Comparative Life Cycle Assessment

A comparative Life Cycle Assessment (LCA) between Self-compacting clay concrete (SCCC) and regular self-compacting cement concrete has been performed. This study analyzes the life cycle from cradle to gate, accounting for all the impacts generated during the production of both materials. This LCA has been performed following the framework provided by the ISO 14040 [12].

4.1 Functional unit

The functional unit used for this study is one square meter of slab with a 10 cm thickness. Therefore, the comparative unit for both slabs is 0.1 cubic meters. This building element was chosen as the comparative unit since both materials meet the mechanical requirements to be used as a slab according to ACI 360R-92 [13] and Code on Structural Concrete EHE-08 [14]. A structural verification calculation was performed with Cype3D v. 2021.a. where the SCCC met the structural requirements for a standard single family-house established in the Technical Building Code (CTE SE-AE) [15] for residential use within the following conditions:

Slab dimensions: 10x6x0,1 m

Gravitational actions: Own weight 24,00 kN/m³; Dead loads 2,00 kN/m²

Variable actions: Surface load 2,00 kN/m²; Punching shear load 2,00 kN applied in 5x5 cm²

To obtain the solicitations, the principles of Rational Mechanics and the classical theories of the Resistance of Materials and Elasticity have been considered. The calculation method applied was the Limit States.

4.2 Inventory analysis

A Life Cycle Inventory (LCI) was performed to account for every activity, raw material, and process that can have an impact on the environment. The tool used to model the LCI was Simapro 8.3.1.0, one of the most well-known software used for LCA calculations. The dosages of both SCCC and self-compacting cement concrete are specified in Table 6.

Table 6. Concrete dosage for 1m² of a 10cm thick slab.

| | Materials | Mass (%) |
|-----------------|------------------------|----------|
| Cement concrete | Water | 7.50 |
| | Portland cement | 15.00 |
| | Sand 0/2, grinded | 20.62 |
| | Sand 0/4, grinded | 15.46 |
| | Sand 0/4, washed | 15.46 |
| | Gravel, round, washed | 25.78 |
| | Superplasticiser | 0.15 |
| | Anti-washout admixture | 0.03 |
| | Total mass | 100.00 |
| SCCC | Sand 0/4, washed | 32.92 |
| | Sand 0/2, grinded | 17.68 |
| | Gravel, round, washed | 28.05 |
| | Clay | 13.90 |
| | Tap water | 7.32 |
| | Galic acid | 0.09 |
| | Sodium hydroxide | 0.04 |
| | Total mass | 100.00 |

4.2.1 Data quality

The data used for the Life Cycle Inventory was collected during the testing and the production process of the SCCC. The cement concrete dosage was provided by industry partners. Additional data used to conduct this study was extracted from the Ecoinvent V3.5 database [16]. Ecoinvent is a not-for-profit association founded in the early 90s by the domain of the Swiss Federal Institutes of Technology. It compiles real data about the impacts generated by every industry field with over 14700 LCI datasets. The peer-

review process that every piece of data undergoes before being approved as a part of the database makes it a highly reliable source [17].

4.2.2 Production phase model

An overview of the production processes of the materials involved both in the SCCC and in the self-compacting cement concrete is included in this section.

Self-compacting cement concrete inventory

Cement: The most common technique for manufacturing cement is the dry method. First, limestone and clay are quarried. After the extraction, the rocks are crushed in two steps. The first step consists of crushing the rock to a maximum diameter of 152 mm. The rock then goes to hammer mills where it is reduced to around 76 mm or less. After that process, the rocks are combined with iron ore and fly ash and then ground. Once these materials are mixed, they are fed into a cement kiln and heated to around 1500 °C. After the heating process, the elements in the mix unite to form a new substance called clinker. Clinker comes out of the kiln as small grey balls the size of marbles. The clinker goes through various coolers to lower its temperature. In the final step, the clinker is then ground and mixed with small amounts of gypsum and limestone [18][19][20].

Sand and gravel: Sand is usually mined in open excavations. This process is carried out with power shovels, draglines, front end loaders, and bucket wheel excavators. In some rare cases, light charge blasting is required to loosen the deposits. After mining, the sand is then suctioned and transported to processing plants. Although sand is sometimes used straight from the quarry, it usually requires further processing. After being transported to the processing plant, the sand is directly loaded into a hopper,

typically covered with parallel bars to screen out big cobbles or boulders. Then the sand is transported on a conveyor belt to scalping screens. Scalping screens separate the oversize material from the smaller marketable sizes. The oversize material is usually crushed and returned to the process. The material is then fed into a battery of vibrating multi-deck sizing screens. Rotating trommel screens with water wash the sand and gravel. After the screening, the sized material is transported to stockpiles or storage bins. Then, water classification is then used to separate the different granulometry, and then the material is dewatered using hydro-separators. The resulting material is transported to stockpiles or storage bins on conveyors belts [21].

Superplasticiser: a polycarboxylate-based superplasticizer is considered for the production of the cement concrete in this study. Polycarboxylate ethers (PCE), which are linear polymers, contain groups with polyoxyalkylene (especially polyethylene or polypropylene glycol groups) as well as carboxylic acid or carboxylic acid anhydride monomers. Several components are submitted to a polymerization process to obtain polycarboxylate. Polyethylene glycol, acrylic acid, and maleic acid are obtained from crude oil. Hydrogen peroxide is extracted from natural gas and sodium hydroxide from rock salt. The batch polymerisation process requires a polymerisation plant and suitable industrial buildings [22].

Anti-washout admixture: admixtures are based on cellulose ether. Cellulose ethers are made by reacting cellulose first with aqueous sodium hydroxide and then with an alkyl halide [23].

Self-compacting clay concrete (SCCC)

Illitic soil: illitic soil is mined using excavators and load-hauling trucks. It is usually extracted with a bench-mining technique that enables the clay to be quarried separately. Depending on its category, clay is then aged for 3 to 12 months. The aged clays are then blended according to the desired formulation and roll crushed to a diameter of 5 cm. The clay is then put in a fluid bed dryer to reduce the moisture content to 10%. Once the drying process finishes, the last step is to use a roller mill to obtain clay powder [24].

Sodium hydroxide: commonly called caustic soda, is mainly produced by the electrolysis of soda. Currently, the most common procedure in the industry is called the ion-exchange membrane method. The process starts by dissolving salt in water and removing its impurities. Then the solution is fed to an electrolyser. The electrolysis process produces caustic soda and chlorine. The concentration of caustic soda obtained in the cathode is around 32%. The solution is taken to an evaporator that produces concentrated caustic soda. Thanks to a density meter installed in the evaporator, it is possible to control the steam temperature and the pressure inside the evaporator to maintain a suitable concentration of caustic soda [25].

Gallic acid: This can be obtained from Tara Spinosa. Tara is a small leguminous tree, native to Peru. The production process begins with the harvest. The pods are cleaned to remove impurities, and then deseeded and separated from the beans. After the deseeding, the pods become a mixture of powder and fibre, which then undergoes an extraction process using water at 70°C. The liquid extract is purified by decantation and filtration. The final product is obtained by submitting the purified liquid extract to an atomisation process [26].

4.3 *Life impact assessment*

4.3.1 *Allocation principle*

The allocation principle followed in this study is cut-off by classification, based on the cut-off system model. This model relies on the idea that the original producer of the material is responsible for the waste generated, not receiving any credit for using materials that can be recycled. The consequence of this is that recycled materials are burden free environmentally-wise [27]. Even though this study does not consider any recycled materials, this choice directly affects those pieces of data extracted from Ecoinvent. This model is considered to be the most adequate for this study.

4.3.2 *Evaluation methods*

Among all the available methods for performing the life cycle assessment, two are selected: The IPCC.GWP 100a [28], and the Environmental footprint (EF) method. Developed by the intergovernmental panel on climate change, the IPCC GWP 100a method is used to calculate the greenhouse gas emissions (equivalent CO₂ Kg) emitted by each material. The EF method was developed by the European Platform on Life Cycle Assessment. This method calculates the impacts of 19 different impact categories. The method describes the methodology for normalizing and weighting the results. By that process, it is possible to compare the results adequately and also to join them together in a single score result [29].

5 Results

The results of the tests carried out on the raw materials and on the specimens prepared for the mechanical tests are presented below.

5.1 Soil characterisation tests

The results of the soil characterisation tests are shown in Table 2. From the results, the soil has been classified as a silty-clay (CL-ML) of low plasticity according to USCS soil classification.

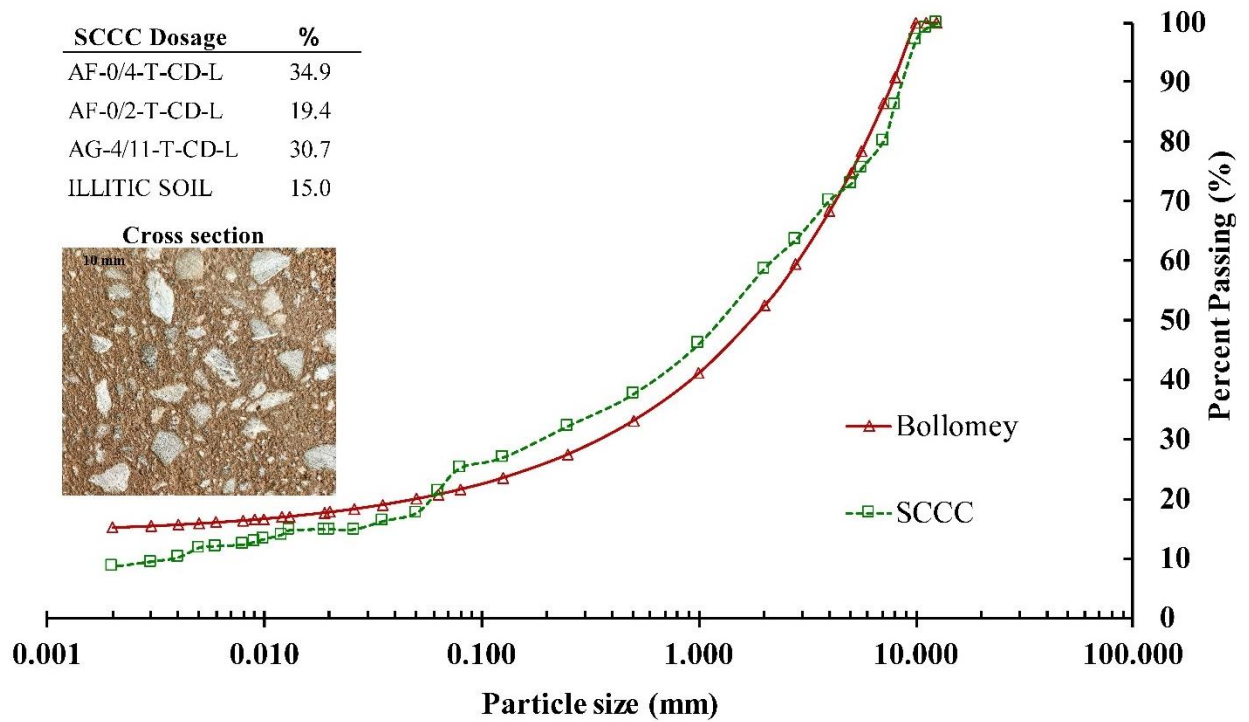
The results of the X-ray diffraction test showed an elevated percentage of K_2O , which is natural in illitic soils. The comparison of the mineralogical composition with the ICDD database revealed a high percentage of illite and kaolinite (Table 3). The phyllosilicate structure of this colloid is appropriate for construction due to its high adherence and dimensional stability under RH% environmental variability. The sedimentation test provided us with the real percentage of clays and fines to complete the fines area of the granulometric curve of our designed concrete. These percentages verify that the soil is silty-clay (CL-ML).

5.2 SCCC preparation

The design of the granular skeleton of our SCCC with the available materials produced a curve similar to the ideal proposed by the Bolomey method (Fig. 2). The distribution of the aggregate in the SCCC can be seen in the cross-section as shown in Figure 2.

For the preparation of the SCCC, the following dosage of additives under investigation was treated as ideal: 0.8% AG/IL and 0.5% NaOH/H₂O both by weight.

Figure 2. Granular skeleton proposal and SCCC cross-section.



5.3 Mechanical tests

5.3.1 Fluidity test and shrinkage test

For the SCCC, with a decrease of the Abrahams cone of 16 cm, the results of linear shrinkage after drying at room temperature and after extraction from the oven are displayed in Table 4.

Table 4. Lineal shrinkage.

| Sample | 24h T 20°C | 7d T 20°C | 24h T 60°C |
|--------|------------|-----------|------------|
| SCCC | 0.15 | 0.72 | 0.76 |

5.3.2 Compressive and flexural strength

The compressive and flexural strength results are presented in Table 5. These values were entered in the calculation simulation to verify that SCCC mechanical properties meet the requirements for the chosen functional unit according the Code on Structural Concrete (EHE-08) [14].

Table 5. *Compressive strength (f_c) and Young's modulus results.*

| Sample | ρ (Kg/m ³) | f_{cm} (MPa) | f_{ctm} (MPa) | E_{cm} (MPa) | Poisson's ratio, ν |
|--------|-----------------------------|----------------|-----------------|----------------|------------------------|
| SCCC | 2,308 | 6.3 | 1.43 | 2,686 | 0.13 |

Flexural ultimate limit state result 0,30 MPa and Punching ultimate limit state 0,120 MPa. These results confirm that a 10 cm SCCC slab complies with the Ultimate Limits State criterion of flexotraction by point load of 2 kN and supports the punching shear limit state for the 2 kN load applied to a 5x5 cm surface, values within the acceptance parameters of the Technical Building Code (CTE SE-AE) for residential use.

Young's modulus and flexural test for the NA specimen did not show representative results given the volumetric inconsistency and irregularity of the specimens and given that the mixing moisture to retain a constant flow rate caused a significant shrinkage of the mixture without additive.

A comparison of the horizontal structural element (the slab) was made, and the requirement was its dead load, its weight, as well as its live load, the structure in operation, of ± 10 KN/m² for a standard single family-house.

5.4 *LCA results*

The LCA calculations were carried out using two highly trusted methods. The first one is the IPCC GWP method, developed the Intergovernmental Panel on Climate Change. The carbon dioxide equivalent emissions are obtained through this method. The second one is the Environmental Footprint method v2, developed by the European Platform on Life Cycle Assessment of the European Commission. This method offers a comprehensive set of results in different impact categories.

5.4.1 IPCC GWP

Networks representing the different processes involving the production of the two kinds of concrete and their contribution to the total carbon emissions are depicted in Figure 3 and Figure 4. In the case of the cement slab, it can be observed that almost 90% of the emissions occur during the production of clinker. In the case of the SCCC, the emissions are mostly related to the extraction of sand and gravel.

Figure 3. Network of cement concrete slab.

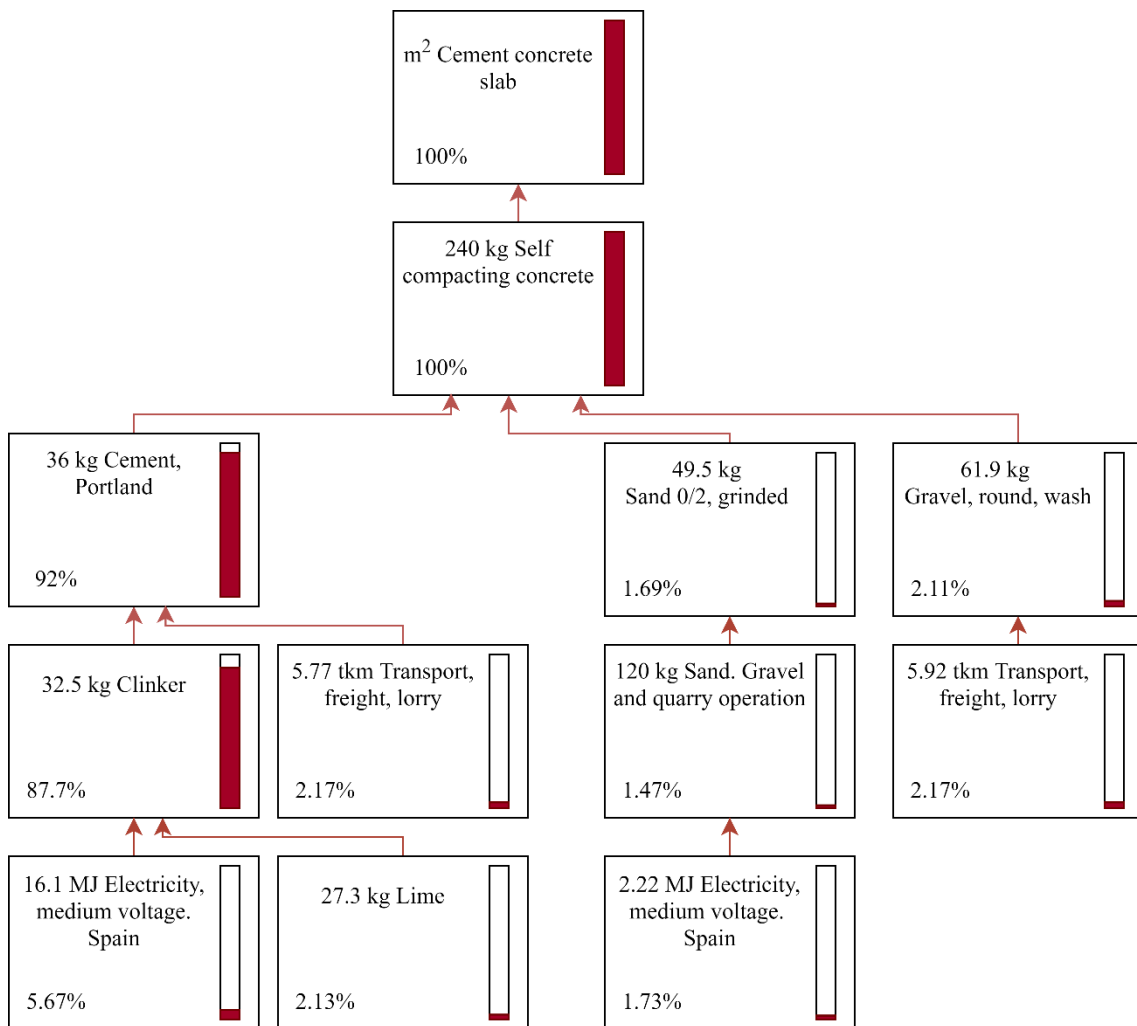
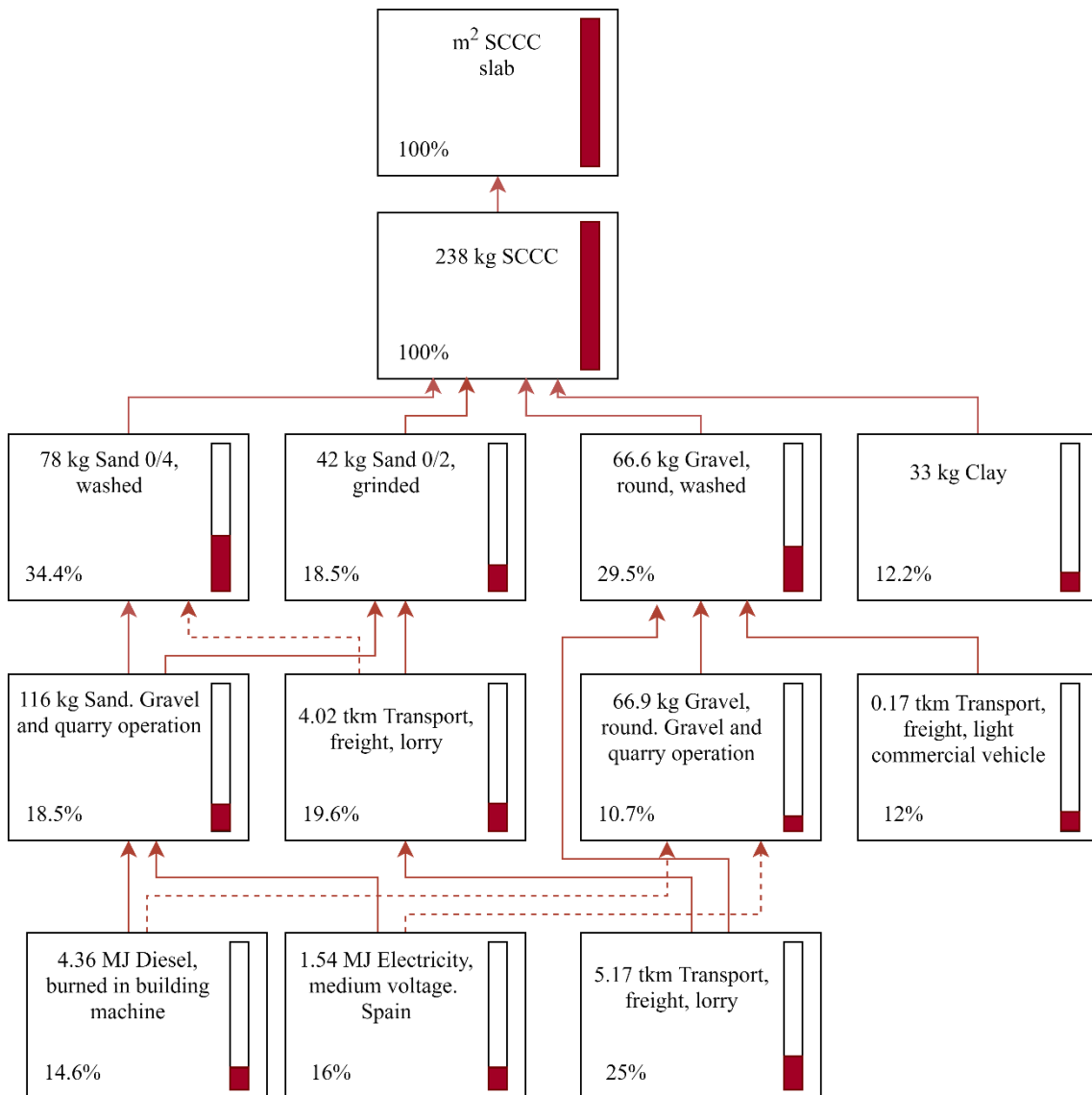
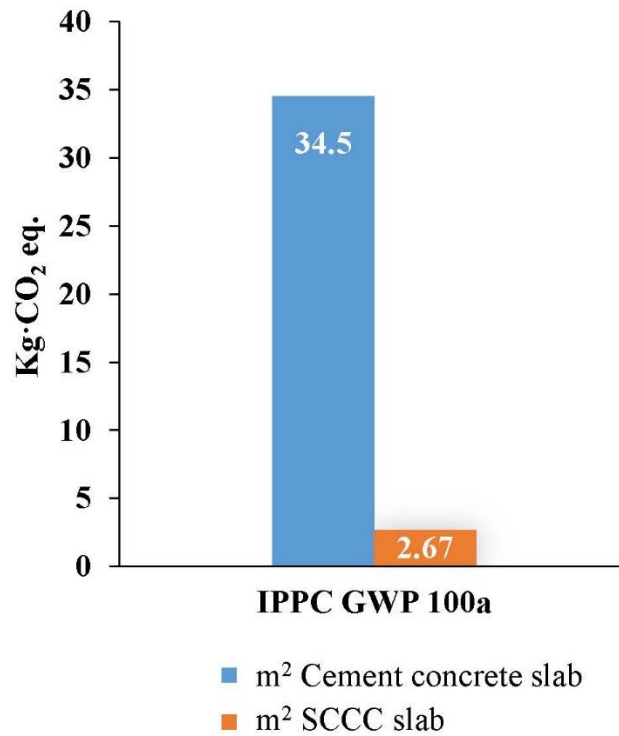


Figure 4. Network of SCCC slab.



The total CO₂ equivalent emissions produced by 1 square meter of each typology are depicted in Figure 5. While building a slab using cement concrete generates 34.5 Kg of CO₂ eq. per square meter, using SCCC to build the slab only generates 2.67 Kg of CO₂ eq.

Figure 5. Comparison between 1m² cement concrete and SCCC.



5.4.2 Environmental Footprint v2 results

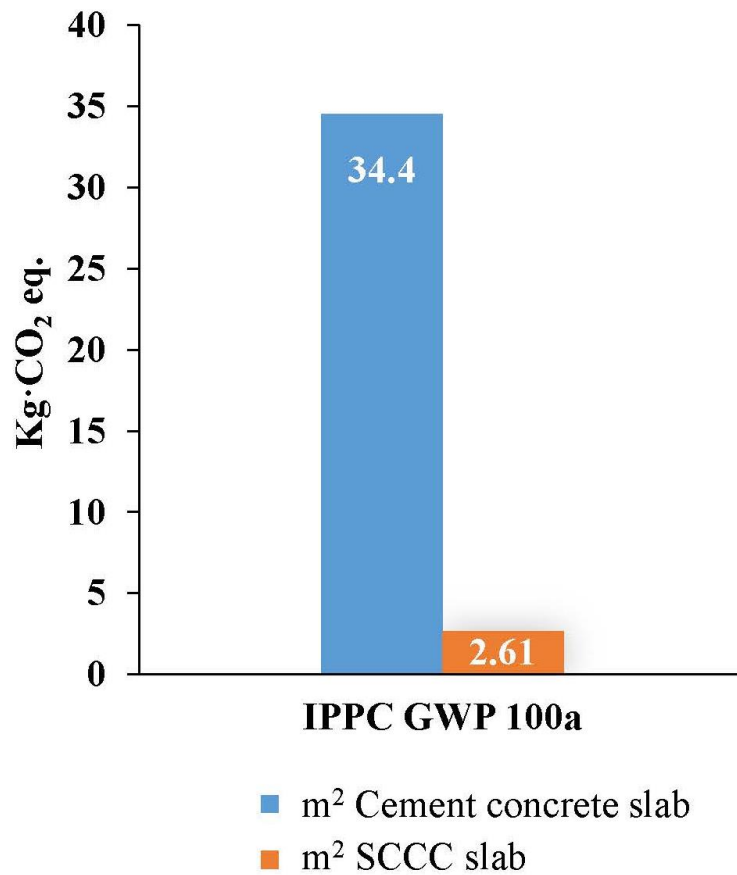
Besides assessing the carbon emissions, other environmental impacts have been analyzed as a part of this study. As said in previous sections, the Environmental Footprint method (EF) has been used to perform the calculations. The characterization results, where the impacts are presented in different impact categories, are reflected in Table 7. As will be discussed in subsequent sections, the results show that the impact generated by the SCCC is lower in every category studied.

Table 7. EF characterisation.

| Impact category | Unit | m ² slab cement concrete | m ² slab clay concrete |
|--|--------------|--|--------------------------------------|
| Climate change | kg CO2 eq | 34.36 | 2.61 |
| Climate change - fossil | kg CO2 eq | 34.33 | 2.61 |
| Climate change - biogenic | kg CO2 eq | 0.0188 | 0.0039 |
| Climate change - land use and transform. | kg CO2 eq | 0.0077 | 0.0025 |
| Ozone depletion | kg CFC11 eq | 1.63E-06 | 4.98E-07 |
| Ionising radiation, HH | kBq U-235 eq | 0.7549 | 0.1638 |
| Photochemical ozone formation, HH | kg NMVOC eq | 0.0787 | 0.0190 |
| Respiratory inorganics | disease inc. | 6.84E-07 | 2.75E-07 |
| Non-cancer human health effects | CTUh | 1.90E-06 | 4.20E-07 |
| Cancer human health effects | CTUh | 1.14E-07 | 6.40E-08 |
| Acidification terrestrial and freshwater | mol H+ eq | 0.1041 | 0.0213 |
| Eutrophication freshwater | kg P eq | 0.0006 | 0.0001 |
| Eutrophication marine | kg N eq | 0.0259 | 0.0060 |
| Eutrophication terrestrial | mol N eq | 0.3166 | 0.0691 |
| Ecotoxicity freshwater | CTUe | 7.00 | 3.52 |
| Land use | Pt | 104.52 | 51.09 |
| Water scarcity | m3 depriv. | 14.36 | 13.46 |
| Resource use, energy carriers | MJ | 182.36 | 37.88 |
| Resource use, mineral and metals | kg Sb eq | 2.73E-05 | 1.60E-05 |

The EF method offers the possibility to normalize the results, which makes it easier to compare the importance that every category has over the total environmental impact. As observed in the normalization results, depicted in Figure 6, the difference between the two typologies in most categories is significant.

Figure 6. EF normalization. Comparison between 1m² of cement concrete and SCCC.



The EF can also weigh the results, increasing the importance that some categories have over the total environmental impact. The weighted results are depicted in Figure 7.

Once the results have been normalized and weighted, the different categories can be added up to obtain a single score result, Figure 8. The impact score obtained by the SCCC slab is 80% lower than the one obtained with the cement concrete slab.

Figure 7. EF weighting. Comparison between 1 m² of cement concrete and SCCC.

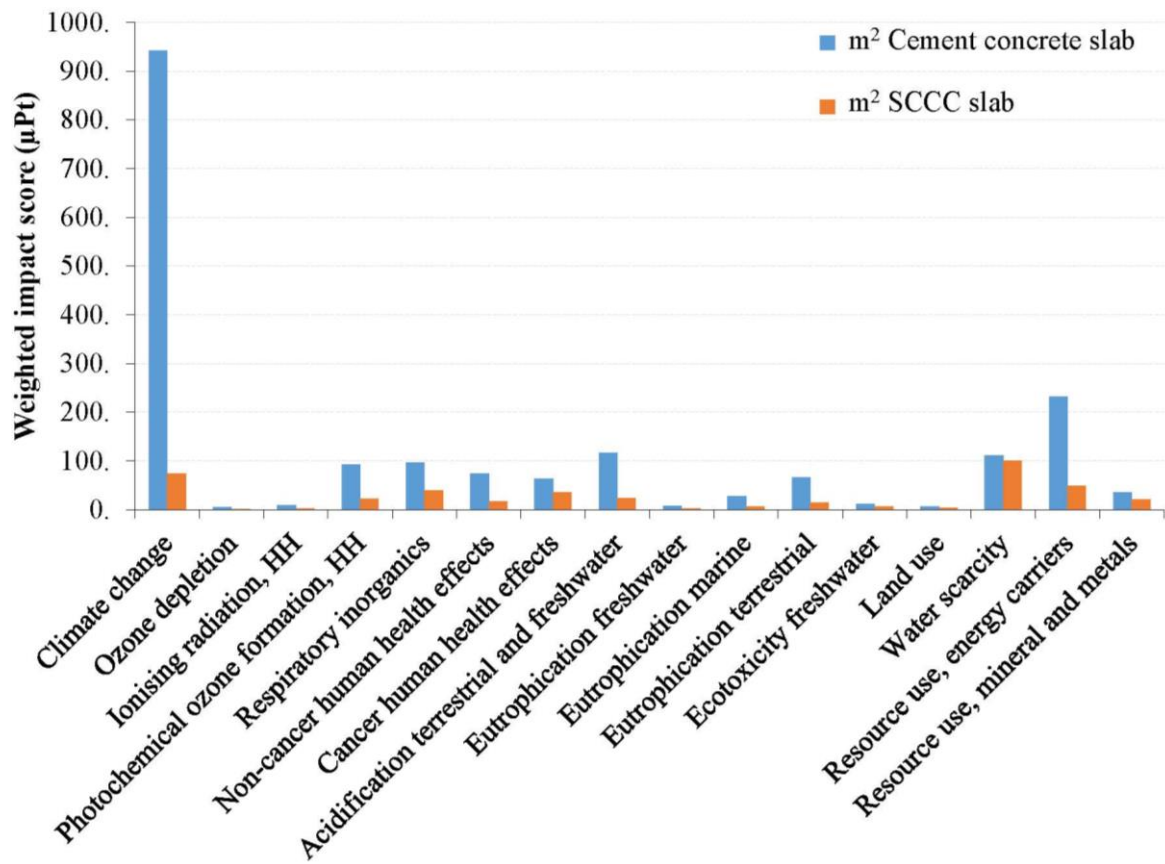
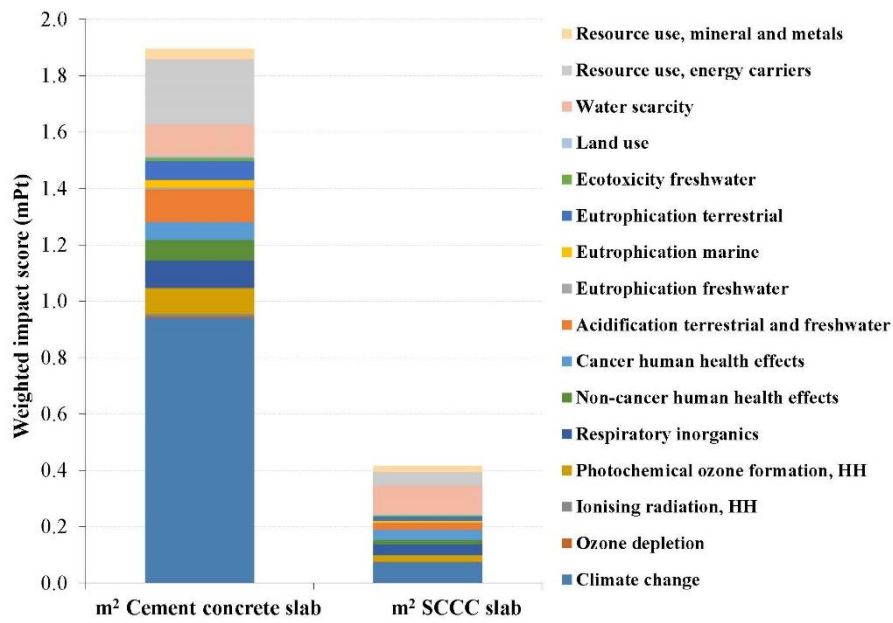


Figure 8. EF single score. Comparison between 1 m² of cement concrete and SCCC.



6 Discussion

After the completion of the study, it is apparent that using SCCC to replace conventional cement concrete is not only better environmentally wise but also adequate in terms of mechanical properties. The results of compressive strength, shrinkage, and fluidity of the SCCC with the natural superplasticizers (GA) studied have been satisfactory for construction purposes in the case of slabs, bearing in mind that the main objective was to achieve liquid concrete without shrinkage for continuous elements. The results are satisfactory because the GA, in conjunction with the chosen illitic soil, at a certain pH, enables clay dispersion with less water such that the concrete becomes highly fluid – and so reduces the amount of mixing water while maintaining the same density as in drier consistencies.

The arrangement of the clay microstructure can also explain the improvement of the compressive and flexural strength. The sorting of the clay changes its electrical charge, as opposed to the case of having an aleatoric arrangement with positions that do not favour the transmission of compressive loads.

In order to obtain conclusive results, a follow-up of the evolution and aging of the material should be carried out in future investigations, also studying the gallic acid reaction in different types of exposure and combination with auxiliary materials.

When it comes to the LCA results, both methods show a big difference between the impacts of the two materials. The results obtained using the IPCC GWP method show a 90% reduction in the carbon emissions when using SCCC to build a square meter of slab. This is due to the massive amount of carbon emitted during the production of clinker. The EF method also shows a big difference in every impact category studied. The results obtained in categories relevant to human health such as cancer and non-cancer human health effects show the positive impact that using SCCC can have on

human health. Other categories related to the ecosystems, for example, resource use and acidification, also reflect the environmental benefits that its use would bring. This becomes apparent when all the categories are summed up after the normalization and weighting process. As it was mentioned in section 5.4.1, the biggest contributor to the environmental impacts of the cement concrete slab is the cement itself, due to the emissions generated during the clinker production. In the case of the SCCC slab, sand and gravel are responsible for almost 90% the total carbon emissions of the slab. These environmental results show consistency with other studies dealing with the environmental impacts of cement [30]. Despite the efforts made by some manufacturers, research indicates that there is a limit to how much the environmental impacts of cement production can be lowered [31]. Using SCCC instead of conventional cement concrete, when the technical conditions are favorable, could have environmental benefits both locally, by attenuating the environmental impacts in places where clinker is produced, and globally, by helping to mitigate climate change.

Conventional cement concrete is an incredibly valuable material but due to its high impact over the environment, finding alternatives to it in some applications is a subject of great relevance. The SCCC presented in this study can be useful to avoid overusing cement in applications where it is not necessary. Furthermore, combining the use of biological materials such as wood, and sustainable geological ones such as SCCC, it is possible to build green constructions.

7 Conclusions

This research aimed to analyze the feasibility of replacing conventional self-compacting cement with a new kind of clay-based concrete. This self-compacting clay concrete

(SCCC) uses soil to replace cement in its composition and chemical additives with natural additives in those scenarios in which the mechanical loads are favourable. The study combined the characterization of the mechanical behavior of the SCCC and a comparative LCA between an SCCC and self-compacting cement concrete slab.

The improvement that this construction technique offered in comparison with its clay-based counterparts is that it maintains a valid compressive strength as a structural element, improving the setting and speed of installation of the material (an important condition in contemporary construction markets) and not shrinking and cracking when used in continuous construction elements such as slabs.

When it comes to the SCCC characterization, the comparison of two types of concrete with different compositions demonstrates the possibility of substituting the cement binder for clay or chemical additives for natural ones in certain scenarios, which emphasizes the technical feasibility of this type of ecological and sustainable construction solutions.

The results obtained in the LCA clearly show that using SCCC instead of cement concrete is hugely beneficial for the environment. By an impact assessment of two comparable typologies, it is possible to identify the existing difference of the materials environmentally wise. In this case, the typology chosen was a concrete slab. The results show a 90% decrease in carbon emissions and an 80% decrease in the overall environmental impact. It can be concluded that using SCCC instead of self-compacting cement concrete to build slabs is not only feasible mechanically wise but also hugely beneficial for the environment and human health.

In future research, the dosages of clay concrete with natural superplasticizers in drier consistencies will be further studied to improve compressive strength instead of workability.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

The authors would like to thank the Geotechnical Department of the UPV (Universitat Politècnica de València) for its vital support in this research. Thanks are due to the bioconstruction cooperative Okambuva Coop, for the practical experimentation spaces, and to the IVE (Valencian Building Institute) for its assistance in carrying out tests. Also, to PYBC S.L. and SILVACHIMICA S.R.L for the materials supplied.

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CAPITULO 3: RESULTADOS Y DISCUSIÓN

En este capítulo se presentan los resultados y la contribución de esta tesis. Principales conclusiones se han extraído directamente de cada documento específico.

En relación al primer objetivo: Identificar un aditivo natural asequible y cercano, susceptible de mejorar las condiciones mecánicas de la tierra, las mayores contribuciones de pueden encontrar en el artículo 1 y artículo 2:

Se observó a la hora de consultar la bibliografía que los aditivos usados a lo largo de la historia en el campo de la construcción con tierra son incontables, y normalmente la metodología de extracción es difusa y poco protocolaria. Se escogió entre todos la *ceratonia siliqua l.* (**Fig 3**) como materia principal de la investigación por su composición y alto contenido en componente reactivo según bibliografía estudiada, además de ser un producto local de amplia explotación y disponibilidad.



Fig. 3 Fruto del algarrobo triturado a 1 cm (izquierda). Fruto del albarrogo pulverizado (derecho).

La complicación en determinar el procedimiento de extracción radicó en la gran variedad de protocolos, y en poder concluir una metodología entre toda la bibliografía consultada, que tuviese en cuenta los criterios prácticos de la arquitectura vernácula y los criterios de la investigación química de la extracción vegetal (artículo 1), sobre todo en el ámbito de la enología por sus propiedades naturales del extracto los polifenoles. Es por ello que el procedimiento de extracción, su caracterización y cuantificación de rendimiento se llevó

a cabo con la ayuda de junto a la Dra. Victoria Lizama Abad en el Dpto. de Tecnología de Alimentos de la UPV (universidad politécnica de Valencia), donde se extrajo el compuesto según el método Pica R. (Pica 2016) (**Fig 5**) de recuperación de antocianos y se identificó y cuantificó el pico de concentración de ácido gálico mediante HPLC (**Fig 4**). (artículo 1 y 2)

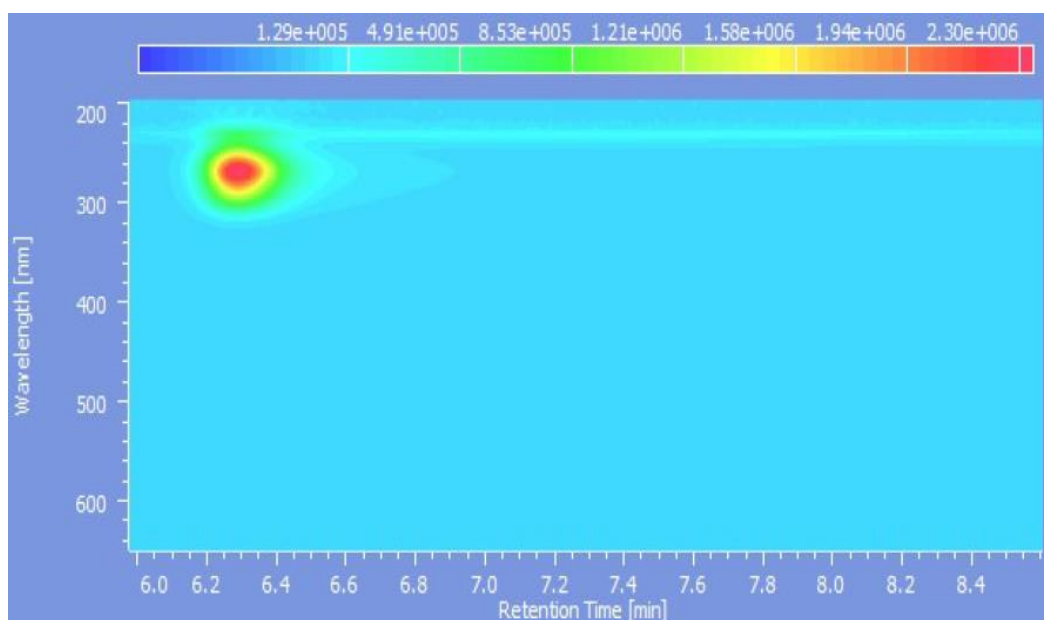


Fig. 4 Determinación del contenido de ácido gálico del extracto de *ceratonia siliqua l.* mediante HPLC.

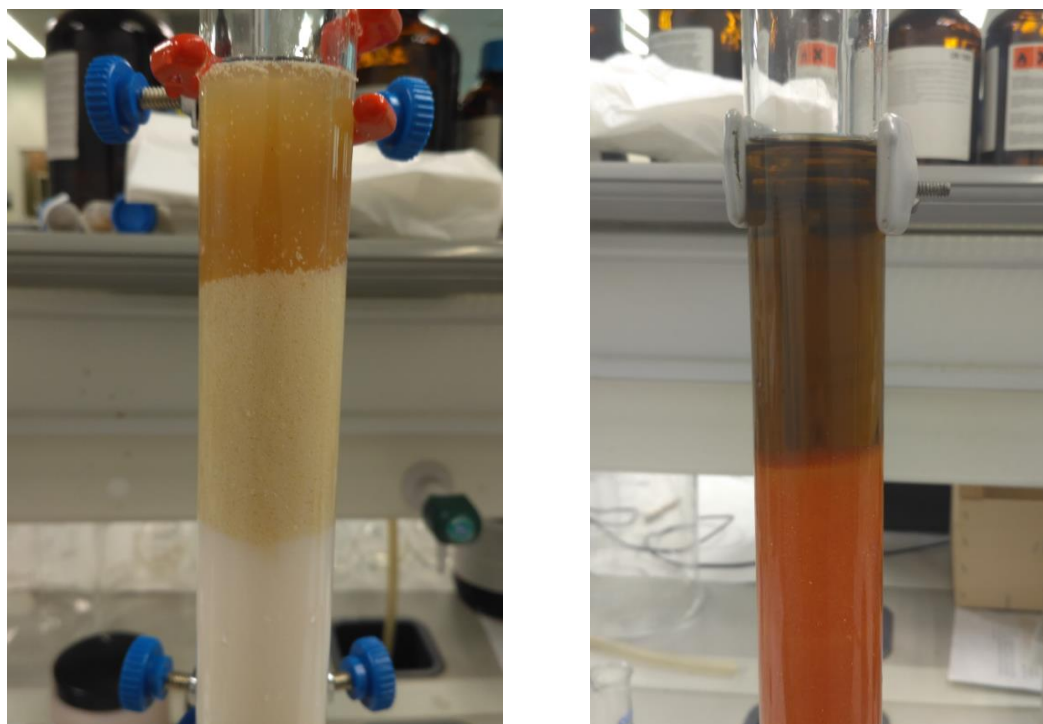
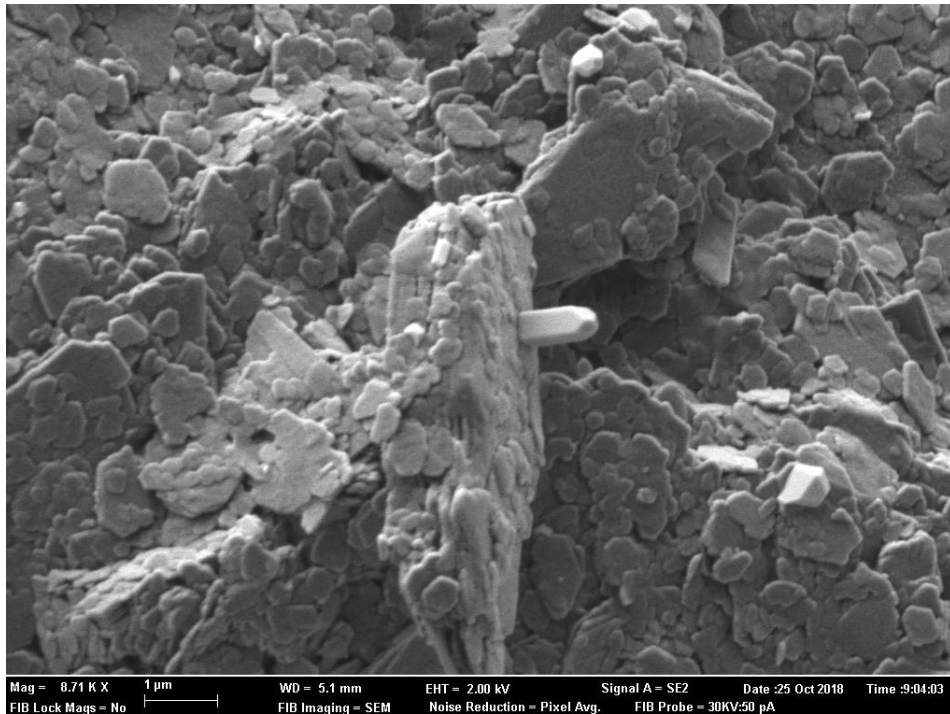


Fig. 5 Extracción de ácido gálico del fruto de la *ceratonia siliqua l.* Resina absorbente a (izquierda) y resina catiónica (derecha).

En relación con el segundo objetivo que era: establecer un protocolo de extracción del compuesto activo reactivo con el suelo, y evaluar las mejoras mecánicas de un conglomerado de arcilla, las mayores aportaciones se realizaron en el artículo 2 y 3:

El extracto de la *ceratonia siliqua l.* ofreció una reactividad específica en unas condiciones ambientales específicos. Según los resultados de protonación realizados por Inclán M. y García-España E. en la Instituto de Ciencia Molecular (ICMol) de la UV (universidad de Valencia), la carga del ácido gálico alcanza -8 a pH alcalino 10-11, lo que aumenta la posibilidad de un intercambio significativo de sus cationes fácilmente disponibles, como el K^+ , por tanto, mediante un estudio de intercambio catiónica del suelo de estudio, y una compensación de su pH mediante adición de componentes básico o ácidos según caso, se pudo alcanzar una reacción ideal con el compuesto de *ceratonia siliqua l.* (artículo 2)

Los resultados de la capacidad de intercambio catiónico de cada suelo reflejaron el potencial que tiene el ácido gálico de ceder los cationes monovalentes K^+ libres a cada suelo. Las arcillas poseen, en mayor o menor medida cationes bipolares interlaminares como el Ca^{2+} que en la mayoría de los casos son coagulantes. Éstos son fácilmente sustituibles por cationes monovalentes disponibles que aporta el ácido gálico como los cationes unipolares de K^+ . Si esta sustitución se produce, un desajuste de cargas provoca modificaciones en la distancia interlaminar de las arcillas, lo que haría debilitar la atracción estática de fuerza variable según naturaleza de la arcilla y sus componentes. Si esta fuerza de Vanderwaals no es suficiente para mantener las láminas unidas (**Fig 6**), la arcilla se dispersa, en este caso el efecto de dispersión del TA en las arcillas sería una dispersión por repulsión electrostática (artículo 2).



*Fig. 6 Reordenación de las plaquetas de arcilla por modificación de carga de las obleas de arcilla.
Mortero de suelo caolínico con AG.*

Como se ha mencionado, la capacidad de reacción del ácido gálico no solo dependió del pH, sino también de su capacidad de intercambio catiónico, es decir, de la propia naturaleza del suelo y sobretodo de sus componentes ferromagnéticos (**Fig. 7**), por ello se realizó estudio en base a distintos suelos de familias radicales lo más distintas posible (artículo 2).

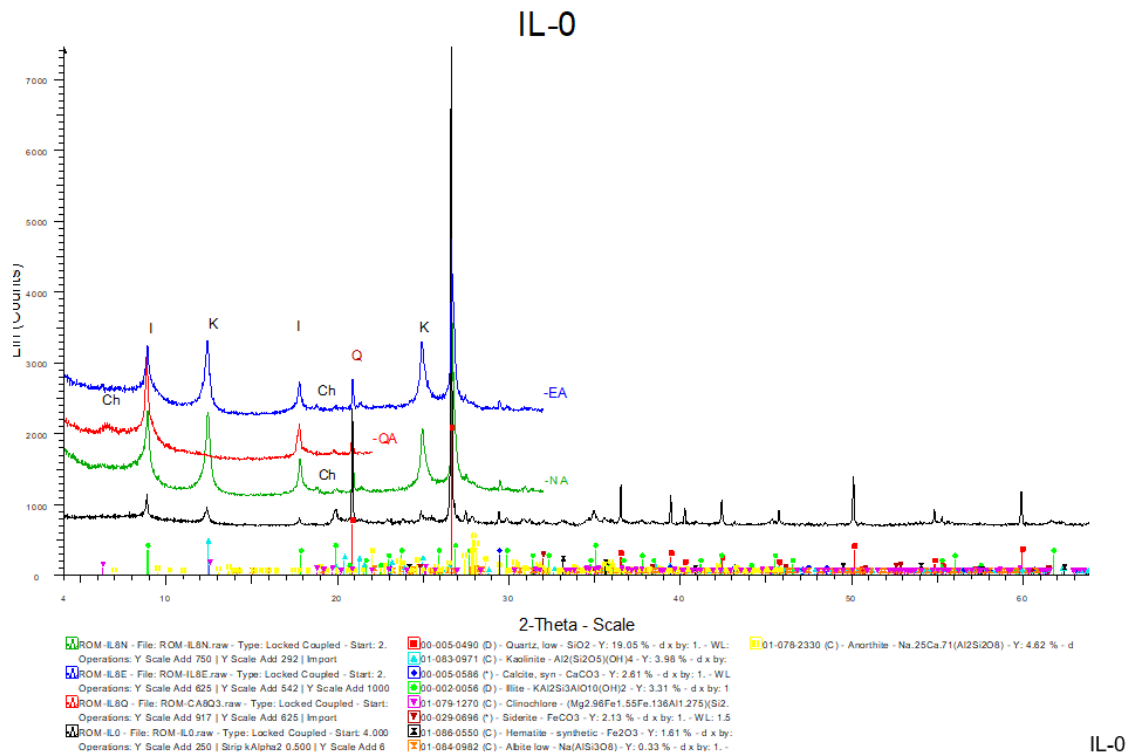


Fig. 7 DRX del suelo illítico con concentraciones importantes en Hematites y Fe_2O_3 .

La reactividad hacia el aditivo GA en cada uno de los suelos es distinta, reconociendo así las posibles aplicaciones en el campo de la construcción, como por ejemplo morteros autonivelantes o de mayor resistencia, siendo esto determinado en última instancia por las propiedades mecánicas del producto resultantes según la reactividad medible (**Fig 8**).



Fig. 8 Cuadro de reacciones en el suelo caolínítico en distintos % de aditivo y pH con el % H_2O demi constante.

Como muestran los resultados de la Tabla 1, en los suelos esmectíticos, se obtuvo reducción de agua mayor, también el caolín como vemos en el reportaje (video 1) (Romero Clausell 2018), no obstante, la aplicación en edificación siempre es mayor en los suelos illíticos (**Fig 9**) combinados dada su composición filosilicática y disposición de sus elementos ferromagnéticos entre láminas, lo que a escala macroscópica representa un comportamiento de adherencia de partículas, anclaje o resistencia mecánica mayor.

| Tipo de suelo | H ₂ O p/p% sin GA | H ₂ O p/p% sin GA | Reducción de agua de amasado (%) |
|---------------|------------------------------|------------------------------|----------------------------------|
| IS | 15 | 11 | 26.66% |
| KS | 24 | 11 | 54.16% |
| SS | 80 | 27 | 66.25% |

Tabla 1 Reducción de agua de amasado según naturaleza del suelo.



Fig. 9 Dispersión de suelo illítico con 10% de agua de amasado, pH9 y %0,01 AG.

Por otro lado, en todos los suelos vemos una mejoría de las propiedades mecánicas (**Fig 10**), siendo por tanto el procedimiento de adición del aditivo ácido gálico, un aporte positivo en diferente medida en dependencia de la composición del coloide arcilla, teniendo en cuenta que la mejoría, y optimización de la reacción para suelos en casos prácticos reales se investigaría en la fase siguiente del estudio.



Fig. 10 Ensayo de abrasión hídrica mediante ensayo SAET.

Comparativa entre mortero con SC sin aditivo (izquierda) y mortero de SC con aditivo (derecha).

Por último, se procedió a extrapolar estos resultados para recrear una curva granulométrica susceptible de poder mantener elementos constructivos de espesor mayor al de un revestimiento, aumentando el tamaño máximo del árido y manteniendo la compacidad del compuesto.

Los materiales utilizados fueron suministrados por empresas del sector de la construcción, y para los procedimientos de ensayos se reprodujeron los protocolos de la construcción convencional, preparando los ensayos correspondientes para poder justificar el material de estudio en la normativa aplicable correspondiente (**Fig 11**) (artículo 3).



Fig. 11 Preparación de especímenes de ensayo para rotura a compresión vertical (F_{cm}).

El tercer objetivo general de la tesis era: Aplicar el aditivo estudiado sobre un material comercializable y compararlo mediante un LCA. Este objetivo se ve representado mayormente en el artículo 3:

El hormigón en base arcillosa fabricado o SCCC (*Self Compacting Clay Concrete*) presentó un escenario comparativo ideal sobre un hormigón de cemento convencional en su aplicación en ciertos elementos constructivos, como la solera en edificación residencial. El ACV comparativo arrojó datos según la metodología de análisis IPCC GWP de una disminución del 90% en las emisiones de carbono (**Fig 12**) y una disminución del 80% en el total impacto medioambiental. Se puede concluir que utilizar SCCC en lugar de autocompactantes. El hormigón de cemento para construir losas no solo es factible mecánicamente sino también enormemente beneficioso para el medio ambiente y la salud humana.

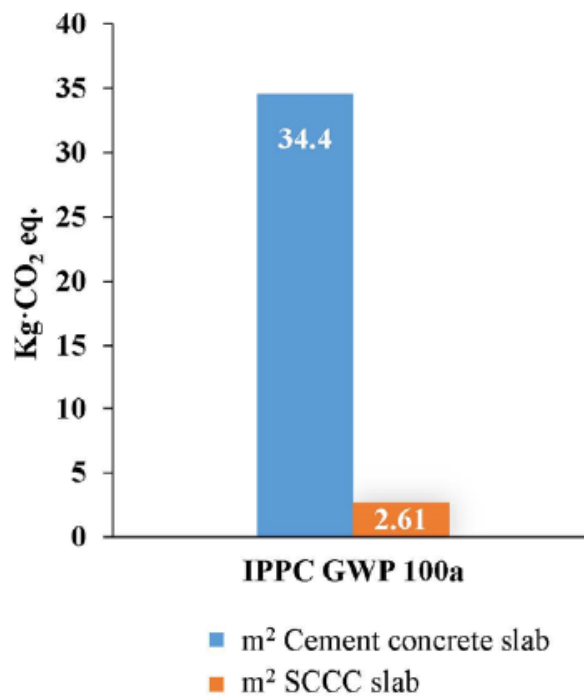


Fig. 12 EF normalization. Comparativa de 1m² de solera de e=10cm entre el SCCC y hormigón de cemento.

CAPITULO 4: CONCLUSIONES FINALES

En la presente investigación se ha tratado de demostrar la viabilidad mecánica del uso de la construcción con tierra dentro del marco constructivo contemporáneo a través de la mejoría y adaptación de su puesta en obra mediante aditivos totalmente naturales como es el ácido gálico extraído de la *ceratonia siliqua l.*

Como conclusiones generales en respuesta a las hipótesis previamente planteadas podemos destacar que:

- *Hipótesis 1: Se puede mejorar la rapidez de puesta en obra del mortero de arcilla, reduciendo la cantidad de agua de amasado y reducir el tiempo de secado:*

La ventaja y mecanismo diferenciador de esta técnica de construcción con tierra radica en la minimización de la cantidad de agua de amasado sea cual sea la trabajabilidad o fluidez que queramos conseguir. Con el aditivo estudiado conseguimos una reducción del agua de amasado de hasta un 66% en suelos esmectíticos. Esta reducción de agua de amasado repercute directamente sobre el tiempo de endurecimiento del SCCC y por tanto de su puesta en obra.

- *Hipótesis 2: Se puede aumentar la resistencia a compresión de los elementos conformados con mortero u hormigón de arcilla:*

Sobre un volumen concreto, según la consistencia sobre la que estemos trabajando o queramos conseguir, ya sea seca, blanda o líquida, al reducir la relación A/A, aumentamos la densidad del material, y por tanto esto repercute directamente en su resistencia a compresión; en la misma línea, la minimización de microfisuras por retracción del material que pueda mermar la resistencia a compresión, y por otro lado la disposición de la microestructura de la arcilla, como vimos en la Fig.8, con el cambio en su carga eléctrica, favorecen la transmisión de cargas compresivas entre las partículas del coloide.

- *Hipótesis 3: El hormigón de arcilla puede cumplir con la normativa aplicable para ciertos escenarios y solicitudes:*

Tanto la resistencia mecánica de los ensayos del material como revestimiento, como los ensayos realizados al material como hormigón, han cumplido con los requerimientos de resistencia a compresión y flexión de la normativa aplicable. Es más, la resistencia mecánica resultada para el SCCC fue superior a la mayor clase resistente como elemento constructivo en la norma UNE 41410 Bloques de tierra para muros y tabiques (UNE 41410 2008) para el BTC5.

- *Hipótesis 4: El hormigón de arcilla puede sustituir al hormigón de cemento en ciertos escenarios siendo mucho más sostenible:*

El escenario planteado como comparativa bajo la unidad funcional de 1m² de solera en e=10cm demuestra una reducción importante, de hasta del 90% en las emisiones de carbono y una disminución del 80% en el total impacto medioambiental.

Es importante subrayar que, dada la tendencia general en líneas de producción y manufacturación de materias primas, se desarrolla un análisis de ciclo de vida de materiales homólogos en cuanto a aplicación, y cuya repercusión en clave medioambiental es muy distinta, no solo en cuanto a emisiones de CO₂, sino en cuanto a posibilidades y ventajas respecto a técnicas constructivas de la misma vertiente de la construcción con tierra, con un ahorro y minimización del uso de agua muy importante.

Esta investigación por tanto deja la puerta abierta a un potencial desarrollo de la tecnología preexistente hacia la incorporación de la materia tierra como un material apropiado y apropiable para el sector de la construcción, el cual siempre debería estar al servicio primero de las personas y en consecuencia con el medio ambiente.

This research has tried to demonstrate the mechanical viability of the use of earthen construction within the contemporary construction framework through the improvement and adaptation of its implementation using totally natural additives such as gallic acid extracted from *ceratonia siliqua l.*

As general conclusions in response to the hypotheses previously raised, we can highlight that:

- *Hypothesis 1: It is possible to improve the installation speed of the clay mortar, reducing the amount of mixing water and reducing the drying time:*

The differentiating function and mechanism of this construction technique with earth lies in the minimization of the amount of mixing water, whatever the workability or fluidity that we want to achieve. With the studied additive, we achieve a reduction of the mixing water of up to 66% in smectitic soils. This reduction of mixing water has a direct effect on the hardening time of the SCCC and therefore its application.

- *Hypothesis 2: The compressive strength of the elements formed with mortar or clay concrete can be increased:*

On a specific volume, depending on the consistency on which we are working or we want to achieve, whether dry, soft or liquid, by reducing the A / A ratio, we increase the density of the material, and therefore this directly affects its resistance to compression. ; Along the same lines, the minimization of microcracks due to retraction of the material that can reduce the resistance to compression, and on the other hand, the arrangement of the microstructure of the clay, as we saw in Fig. 8, with the change in its electrical charge, they favor the transmission of compressive charges between the colloid particles.

- *Hypothesis 3: Clay concrete can meet the applicable regulations for certain scenarios and requests:*

Both the mechanical resistance of the tests of the material as coating, as well as the tests carried out on the material as concrete, have complied with the compressive and flexural strength requirements of the applicable regulations. Moreover, the resulting mechanical resistance for the SCCC was higher than the highest resistant class as a constructive element in the UNE41410 standard, Earth blocks for walls and partitions for the BTC5.

- *Hypothesis 4: Clay concrete can replace cement concrete in certain scenarios, being much more sustainable:*

The scenario proposed as a comparison under the functional unit of 1m² of hearth at e=10cm shows a significant reduction of up to 90% in carbon emissions and a decrease of 80% in the total environmental impact.

It is important to underline that, given the general trend in raw material production and manufacturing lines, a life cycle analysis of homologous materials is developed in terms of application, and whose impact on the environment is very different, not only in terms of CO₂ emissions, but in terms of possibilities and advantages with respect to construction techniques of the same aspect of construction with earth, with very important savings and minimization of water use.

This research therefore leaves the door open to a potential development of pre-existing technology towards the incorporation of earth matter as an appropriate and appropriate material for the construction sector, which should always be at the service of people first and consequently with the environment.

CAPITULO 5: FUTURAS LÍNEAS DE INVESTIGACIÓN

Esta tesis ha abierto una línea de investigación, pero sería necesario una futura profundización para obtener modelos aplicables concretos. La tecnología aplicada de la construcción con tierra evoluciona rápido, y la puesta en uso y variables que pueden suponer trabajar con este material supone un constante estudio y análisis del material

En concreto, en el futuro sería interesante investigar en:

- Obtener dosificaciones específicas para el trabajo en impresión 3D con tierra cruda y superplastificantes.
- Obtener dosificaciones específicas para profundizar en el uso de la tierra como material líquido para la confección de elementos escultóricos o decorativos.
- Estudiar el potencial de esta técnica constructiva en el ámbito de la rehabilitación de patrimonio construido en tierra.
- Estudiar medidas de mejora en los elementos auxiliares de construcción para reducir el tiempo de secado mediante mecanismos transpirables.
- Profundizar en la posibilidad de combinación de otros aditivos complementarios para aumentar la resistencia a compresión creando geopolímeros.

This thesis has opened a line of research, but future research would be necessary to obtain specific applicable models. The applied technology of construction with earth evolves rapidly, and the use and variables that working with this material may entail implies a constant study and analysis of the material.

Specifically, in the future it would be interesting to investigate:

- Obtain specific dosages for 3D printing work with raw earth and superplasticizers.
- Obtain specific dosages to deepen the use of earth as a liquid material for making sculptural or decorative elements.
- Study the potential of this construction technique in the field of the rehabilitation of built heritage on land.
- Study improvement measures in auxiliary construction elements to reduce drying time through breathable mechanisms.
- Deepen the possibility of combining other complementary additives to increase compressive strength by creating geopolymers.

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