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Review

Roadmap across the mesoscale for durable and sustainable cement paste – A bioinspired approach

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HIGHLIGHTS

- The mesoscale connects molecular details with durability of cementitious materials.
- Features of mesoscale models for cement paste are defined.
- Recent trends on combining multiscale modeling and experiments are reviewed.
- A bioinspired framework for the design of cement paste with additives is proposed.

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ABSTRACT

In recent years, continuum and atomistic modeling of cementitious materials has provided significant advances towards studying the durability of civil infrastructure. An important frontier to understanding structure-property relationships is the "mesoscale", which represents the bridge between underlying (e.g. molecular) processes and bulk macroscale behavior. This review highlights examples of a mesoscale approach within biological materials and emphasizes their applicability to the study and design of sustainable cement-based materials at multiple length scales. We propose a methodology focused on the coupling of computation and experiment for furthering our understanding of the microstructural properties that control the durability of hardened cement paste.

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List of notations

AFM	Atomic Force Microscopy
BET	Brunauer-Emmett-Teller Adsorption
BSE	Back-Scattered Electron Microscopy
B-W-I	Bound Water Index
C-A-S-H	Calcium-Alumino-Silicate-Hydrate
C_2S	Di Calcium Silicate
C_3S	Tri Calcium Silicate
CH	Calcium Hydroxide
C-S-H	Calcium-Silicate-Hydrate
DFT	Density Functional Theory
EDS	Energy Dispersive Spectrum
EIS	Electrochemical Impedance Spectroscopy
FEA / FEM	Finite Element Analysis / Method
FTIR	Fourier Transform Infrared Spectroscopy
GSF	Generalized Stacking Fault
H_2O	Water Adsorption
HF	Hartree-Fock
INS	Inelastic Neutron Scattering
MD	Molecular Dynamics
MEP	Minimum Energy Path
MIP	Mercury Intrusion Porosimetry
M-S-H	Magnesium-Silicate-Hydrate
N_2	Nitrogen Adsorption
NEB	Nudged Elastic Band
NMR	Nuclear Magnetic Resonance
OPC	Ordinary Portland Cement
Q	Scattering Vector
QENS	Quasi-Elastic-Neutron-Scattering
RH	Relative Humidity
SANS	Small Angle Neutron Scattering
SAXS	Small Angle X-ray Scattering
SEM	Scanning Electron Microscopy
SMD	Steered Molecular Dynamics
TEM	Transmission Electron Microscopy
XPS	X-ray Photoelectron Spectroscopy
XRD	X-ray Diffraction
XRF	X-ray Fluorescence Spectroscopy
WAXS	Wide Angle X-ray Scattering

1. Introduction

1.1. Motivation: Design of sustainable and durable cement paste

Cement-based composites are one of the earliest examples of an engineered structural material. The Roman's developed lime-pozzolana cements which were used to construct buildings that exhibit exceptional durability and have survived for more than 2000 years [1,2]. Modern cementitious materials are based on Portland cement, a mixture of limestone and clay that is heated and ground to produce a multiphase clinker that was first used in the early 19th century. Both Roman and Portland cement-based materials were initially engineered through gradual empirical improvements due to the material's complexity and sensitivity to composition and environment. Recent developments in nanotechnology have allowed for significant advances from these primitive manufacturing methods by enabling the investigation of these materials at multiple length and temporal scales [3]. The growth of nanotechnology has also facilitated studies on how nature designs and fabricates advanced functional materials such as bone, nacre and deep sea sponge. These biological materials employ hierarchical structures to achieve remarkable material performance that is adapted to their distinct utilization within an organism. Material scientists have begun to replicate these systems to produce synthetic composites which mimic designs found in nature, however, these concepts have not been widely applied to cementitious materials. Therefore, the motivation for this paper is to develop a framework that combines modern characterization techniques with natural design strategies to improve and engineer the durability of cement paste for specific applications.

Cement paste is a model system for exploration because of its ubiquity in the built environment and adaptability to a wide range of mix compositions. This adaptability provides an opportunity to improve and tailor cement paste using locally available additives, deemed supplementary cementitious materials [4,5]. The availability of local additives to a region depends not only on natural resources, but also on its industrial landscape including energy and manufacturing infrastructure. The use of additives for clinker substitution has also been identified as a low cost and high-impact method of reducing the carbon footprint of a rapidly expanding global concrete industry [6]. Therefore, deriving functioning mixes tailored to local economies is both environmentally and economically sustainable. In many cases these additive candidates, such as fly ash or volcanic ash, are waste products which would otherwise require disposal. Phair presents a tutorial for evaluating and producing sustainable cement, and we refer the reader to this review for the general strategy [7].

In addition to the economic and environmental incentives, additives can be incorporated to produce materials with superior properties compared to Ordinary Portland cement (OPC) pastes. When additives with fine particle sizes are used they act as filler material which provides nucleation sites for additional hydration products, in addition to forming secondary hydration products if the additive is reactive in the alkaline pore solution. These materials have been shown to provide denser microstructures, stronger interfacial bonding and a corresponding higher compressive