Understanding the Synergetic Interaction of Graphene-Oxide (GO) and Calcium-Silicate-Hydrates (C-S-H) Nanocomposite: *Ab Initio* Study

Art Anthony Z. Munio[a], Alfredo Q. Liboon Jr.[b], and Leo Cristobal C. Ambolode II[ac]

(a)Physics Department, Mindanao State University – Iligan Institute of Technology, 9200 Iligan City, Philippines
(b)College of Arts and Sciences, Jose Rizal Memorial State University – Tampilisan Campus, 7116 Tampilisan, Philippines
(c)Premier Research Institute of Science and Mathematics (PRISM), Mindanao State University – Iligan Institute of Technology, A. Bonifacio Avenue, 9200 Iligan City, Philippines

Abstract

Recently, cementitious materials (C-S-H) and nanocarbon composites have gained a significant amount of attention owing to their superior mechanical properties and diverse functionalities. Aside from having robust mechanical properties of the composite, which can withstand extreme conditions, breakthroughs in the enhancement of thermoelectric and piezoelectric properties of the C-S-H/nanocarbons composites are reported revealing potential applications as an energy harvester for sustainable infrastructure. Despite several experimental investigations of the C-S-H/nanocarbon composites, very little was known about the synergetic interaction at the nanoscale, which basically dictates the overall performance of the composite material. This study systematically investigates the bonding mechanism and electronic redistribution of the C-S-H/graphene-oxide nanocomposite using *ab initio* density functional theory. The high-throughput calculations revealed that van der Waals and hydrogen bonding dominate the interaction of the C-S-H/graphene-oxide nanocomposite. These claims are supported by careful inspection of the charge density difference isosurface and the 2D contour Electron Localization Function of the nanocomposite. The findings of this study explain the successful incorporation of the properties of the nanocarbon in the cementitious material. We anticipate that this work will serve as the basis for developing smart-cement materials for future smart cities.

I. Introduction

Cementitious (C-S-H) materials are one of the most used and important construction materials. The development of human society relies heavily on the usage of cement. Although, as society advances, construction materials need to meet advanced properties such as superior mechanical properties, multifunctionality, low-cost, self-healing, self-sensing and a self-cleaning mechanism that allows the creation of sustainable and smart infrastructure. These aforementioned properties are not achieved by conventional cementitious materials. Recently, the composite of C-S-H and nanocarbons has been fabricated, revealing robust mechanical properties with advanced properties such as electromagnetic shielding capabilities, piezoelectric and thermoelectric properties[1].

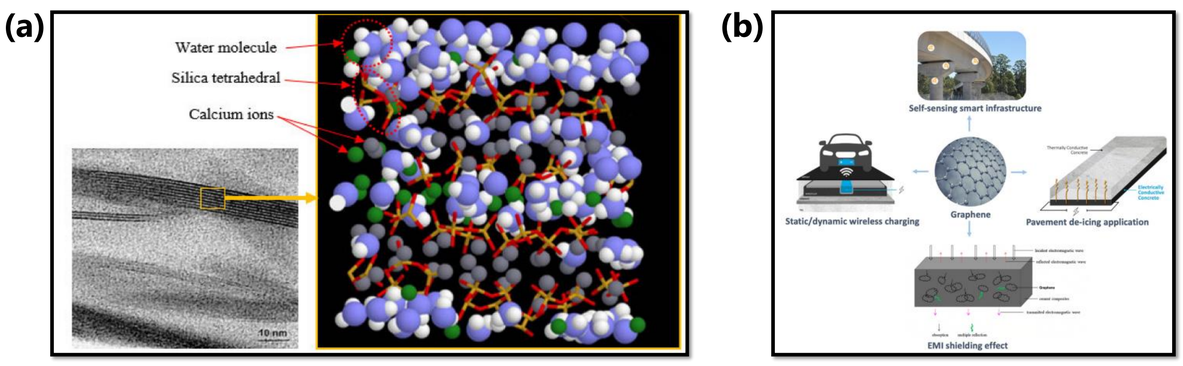


Figure 1. (a) TEM image (left) and atomic configuration (right) of C-S-H layered structures. (b) Potential applications of C-S-H/nanocarbon nanocomposite[1].

Motivation

Despite breakthroughs in cement/nanocarbon composites, the fundamental operating bonding mechanism at the interface remain unexplored.

Objectives

Using high-throughput *ab-initio* density functional theory, we calculated (i) the binding energy per sq.nm; (ii) Charge Density Difference; (iii) Bader charge transfer, and (iv) Electron Localization Function of C-S-H/graphene-oxide nanocomposite.

II. Methodology

All DFT calculations are conducted using *Quantum Espresso* within generalized gradient approximation employing Perdew-Burke-Ernzerhof parametrization and Grimme-D3 dispersion correction[2].

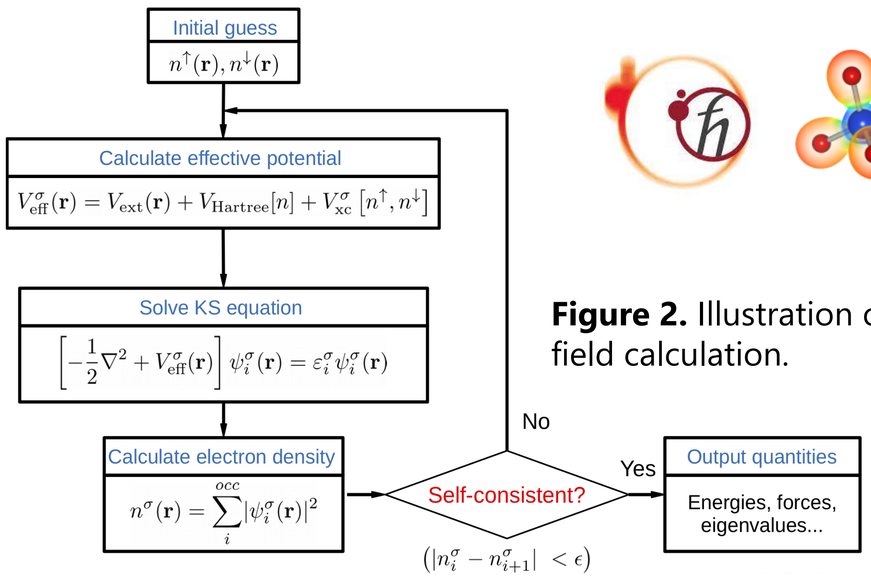
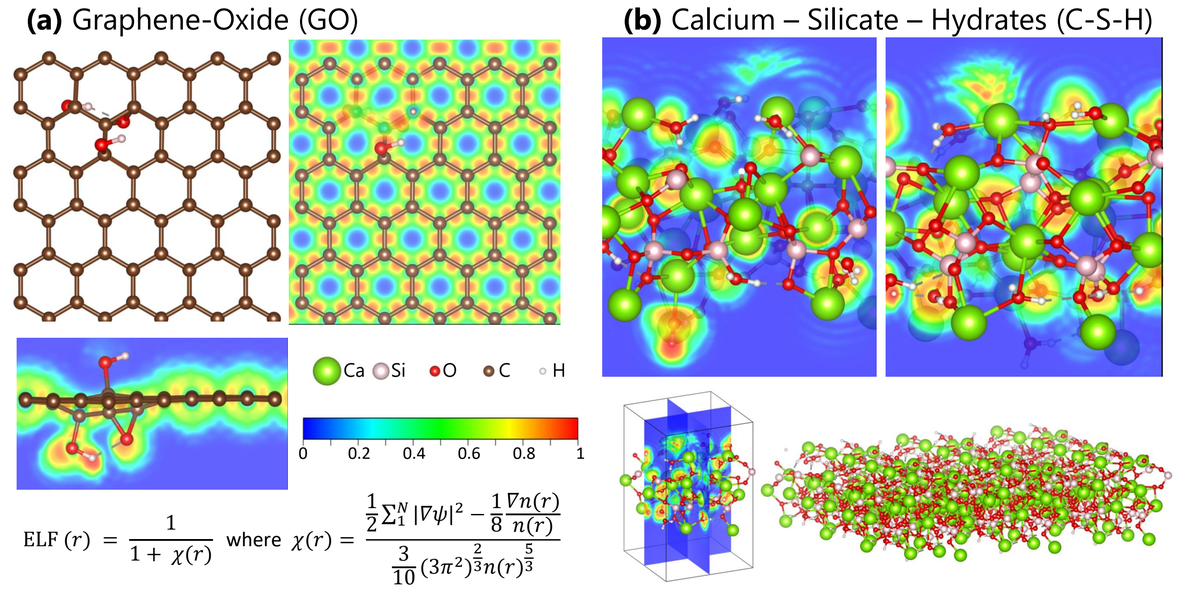


Figure 2. Illustration of self-consistent-field calculation.

III. Results

Graphene-Oxide & Calcium-Silicate-Hydrates



ELF = 0 (No Localization of Electrons)
0.2 < ELF < 0.7 (Metallic Bonding)
ELF > 0.7 (Chemical Bonding)
Deformed ELF (Induced Dipole)

Figure 3. Electron Localization Function of (a) Graphene-Oxide and (b) Calcium-Silicate-Hydrates.

Nanocomposite

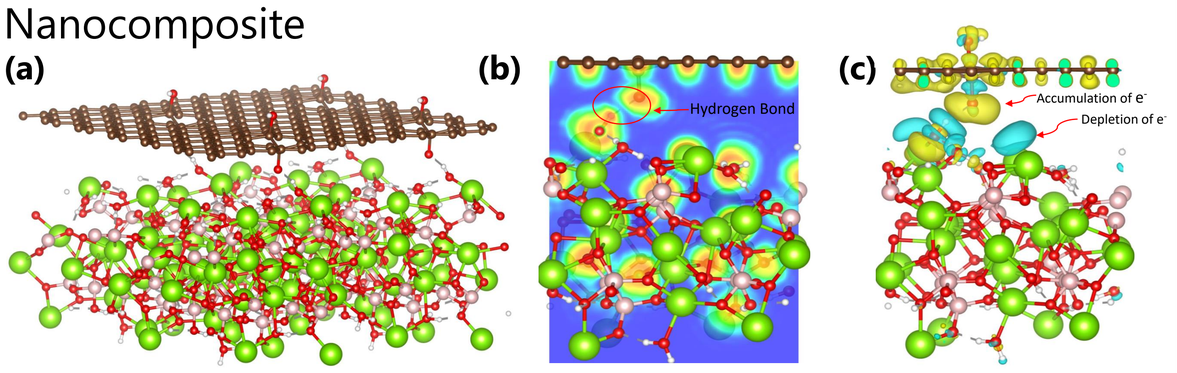


Figure 4. (a) Optimized configuration of C-S-H/Graphene-Oxide nanocomposite. (b) and (c) depicts the Electron Localization Function and Charge Density Difference of the nanocomposite.

Binding Energy = -2.32 eV/sq.nm (Exothermic)
Charge Transfer = 1.35 e/sq.nm (From C-S-H to Graphene-Oxide)
Equilibrium Distances (Hydrogen Bond; Ca-C)= 1.74 Å; 5.12 Å

IV. Conclusions

High-throughput density functional theory calculations are conducted to reveal the fundamental bonding mechanism of the C-S-H/graphene-oxide nanocomposite. The simulations revealed the following:

- The bonding mechanism of C-S-H and graphene-oxide nanocomposite is mainly dominated by strong hydrogen bonding, induced dipole, and van der Waals interaction.
- The process of adsorption is exothermic in nature, where the nanocomposite can form spontaneously and the structure can keep thermally stable.

References

[1] Jayasooriya, D.; Rajeev, P.; Sanjayan, J. Application of Graphene-Based Nanomaterials as a Reinforcement to Concrete Pavements, *MDPI Sustainable,* **2022**
[2] Giannozzi, P.; et al. Quantum ESPRESSO toward the Exascale. *J. Chem. Phys*. **2020**

Acknowledgement

The author acknowledges the support of Jose Rizal Memorial State University, Mindanao State University – Iligan Institute of Technology, and Department of Science and Technology – Science Education Institute.