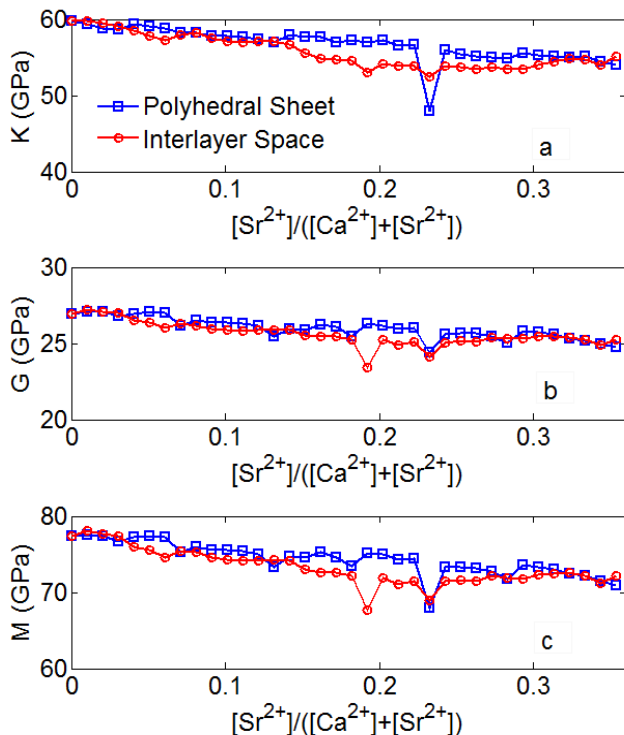


# Locking radionuclides in cement

## Problem

At every stage of the nuclear fuel cycle, cement-based materials play a major role in providing support, shielding and containment. For example, concrete structures are used as storage pools for fresh nuclear fuel. Later on when the nuclear fuel is in operation in the nuclear power plant, the concrete containment, the outermost barrier, shields against any leakage of radioactivity to the biosphere and also is known to be the only irreplaceable component of the nuclear power plant. For decades, concrete structures in nuclear power plants have served as an interim for the spent nuclear fuel. Furthermore, cementitious materials can be used as the waste form and/or container for the whole waste package in the ultimate disposal of radioactive materials in geological repositories. A fundamental understanding of the long-term performance of cement-based materials and developing capabilities to project their lifetimes is of utmost importance in the context of nuclear energy.



The mechanical properties of CSH as a function of  $\text{Sr}^{2+}$  substitution of  $\text{Ca}^{2+}$ . (a), (b) and (c) are the bulk, shear and indentation moduli of C-S-H, respectively.

## Approach

With our recent advances in understanding and modeling the structure of cementitious materials at the nanoscale, we assessed the stability and the mechanical integrity of cement upon cationic exchange between  $\text{Ca}^{2+}$  and  $\text{Sr}^{2+}$  - the former being the host cation in cement hydrate (C-S-H), and the latter is a critical radionuclide found in spent nuclear fuel. We implemented a series of Molecular Dynamics simulation in the NPT ensemble using the core-shell description for inter-atomic interactions and derived the textural and mechanical properties.

## Findings

At the nanoscale, we find that the  $\text{Sr}^{2+} \leftrightarrow \text{Ca}^{2+}$  substitution is energetically favorable in the interlayer void space of C-S-H; but not inside the calcium oxide polyhedral layers. This  $\text{Sr}^{2+} \leftrightarrow \text{Ca}^{2+}$  cationic exchange in the interlayer space does not interfere with stability and coordination of the silicate chains in C-S-H. This trend is more pronounced for low Ca/Si (here, we have considered 9 Å Tobermorite with Ca/Si=0.83 as a reference system) than for high Ca/Si hydrates (normal hydrates with Ca/Si=1.65), which is indicative of nanoscale textural effects. Finally, while we find that the mechanical properties degrade as strontium concentration increases, this degradation is not severe. This suggests that strontium radionuclide waste can be safely locked into cement hydrates for long-term storage. Future work will consider the other important radionuclide found in spent nuclear fuel ( $^{137}\text{Cs}^+$ ).

## Impact

We propose a new approach to investigate environmental and safety issues for the use of cement as a candidate for immobilizing radioactive waste such as strontium. It is based on first-principle statistical physics and allows a quantitative assessment of the impact of locking radionuclides on cement hydrate stability and mechanical properties.

## More

Research presented by Mr Mostafa Youssef under the supervision of Professors Bilge Yildiz and Roland J.-M. Pellenq.



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