

## Abstract

Cement is a well-known infrastructural binder material. Typically, cement is obtained as a powder material which on hydration forms a rock-solid material. Estimation of constituents of the powder (or unhydrated) and hydrated cement is a standard procedure nowadays with XRD/Rietveld analysis. Detailed NMR studies along with XRD have also revealed the molecular structure of each of different components of both unhydrated and hydrated cement. In this study, molecular deformation study has been carried out individually for many of these unhydrated and hydrated cement constituents.

Amongst the unhydrated components of cement, study has been carried out for tricalcium aluminate and calcium sulfates. The evolution of bonds, angles along with the structure of the puckered ring were observed to influence the structure-property relationship of tricalcium aluminate under different loading conditions. For layered structures, such as that of gypsum, the evolution of inter and intra layer separation along with the layer slip influences the structure-property relationship of the gypsum crystal. Water molecules embedded in different hydrated calcium sulfate compounds (gypsum and hemihydrate) was also observed to influence the stress-strain response of the material in comparison to that of anhydrite.

Amongst the hydrated components of cement, structure-property relationship study has been carried out for different phases of tobermorite and jennite (generally considered to be as natural pure mineral analogues of calcium-silicate hydrate), ettringite and portlandite. Atomic charges and bond parameters are observed to change for these materials subjected to large deformation responses. The presence of embedded water in these molecules also influenced the structure-property relationship of these materials.

Stress-strain results obtained from molecular simulations for each of the individual components have significantly higher values compared to results obtained from experimental investigation of hydrated cement. The primary reason for this is presence of defects (voids, inclusions and exclusions) with experimental samples whereas pristine single crystals are considered for molecular simulations. Thereby,

a multiscale homogenization scheme (based on Mori-Tanaka theory) was developed to relate the elastic constants (of each individual components) obtained from molecular simulations to that of experimentally observed elastic constant of the paste.