MOLECULAR MODEL OF GEOPOLYMERS WITH INCREASING LEVEL OF DISORDER IN THE ATOMIC STRUCTURE.

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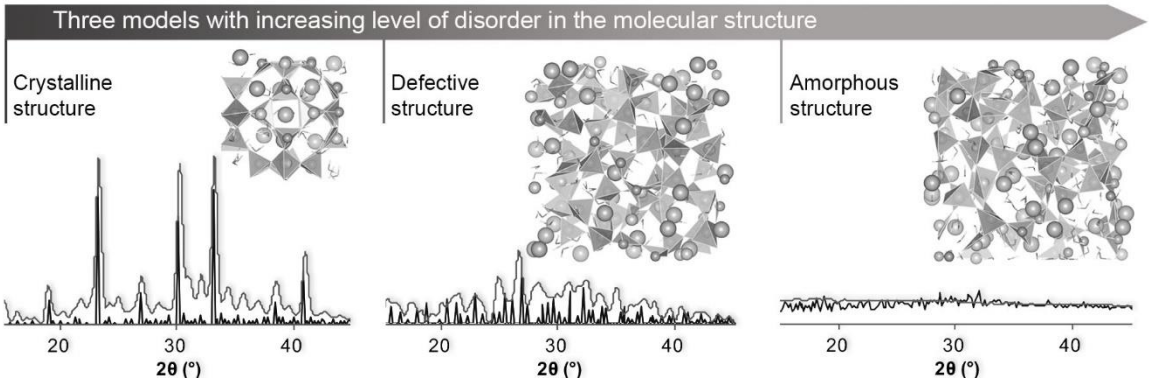

Figure 1 –Three different geopolymers molecular models with increasing level of disorder but with comparable Si:Al. The level of disorder is characterized by the corresponding X-ray diffractions.

Concrete is the most used building material on Earth, but the production of its main binding component, cement, is responsible for 8% of worldwide CO_2 emissions. A greener alternative cementitious material is provided by geopolymers, which can be synthetized from calcined clays and industrial by-products. A key issue, that limits the applicability of geopolymers in the construction sector, is an insufficient understanding of the relationship between their chemistry and development of long-term properties. Reducing these uncertainties requires an integrated approach combining modelling and experimentation. The binding phase of geopolymers often consists of sodium-alumino-silicate-hydrates (N-A-S-H), obtained through the reaction of a sodium silicate solution with an alumino-silicate source. Theoretical models describe this structure at the molecular scale as “pseudo-crystalline” [1] but, the existing models, based on solely amorphous or crystalline structures, are not always in agreement with this definition and with experimental results. For this reason, a defective crystalline structure is proposed here as a baseline geopolymer cell, featuring both amorphous and crystalline attributes (Figure 1). This new structure is created by creating vacancies in a sodalite crystalline cage, which is then stress-relaxed and reorganised to achieve full polymerisation of Al and Si tetrahedra while respecting the Loewenstein's principle. Results are compared with experimental data and with other simulation results for amorphous and crystalline molecular models, showing that the newly proposed structures better capture important structural features with impact on mechanical properties, reconciling experiments with the “pseudo-crystalline” model. Specifically, the comparison with the experiments addresses the effect of Si:Al molar ratio and water content on a range of structural and mechanical properties such as skeletal density, ring structure, bong-angle distribution, X-ray diffraction (Figure 1) and X-ray pair distribution function. The simulation results confirm the necessity of a defective structure able to detect both order and disorder in geopolymers experiments. The proposed defective molecular model provides a starting point for the multiscale understanding of geopolymer cements, as well as for investigating the molecular interactions between geopolymer cements and various adsorbates, e.g. for applications in environmental engineering and nuclear engineering.

References:
[1] J. L. Provis, G. C. Lukey and J. S. J. van Deventer, Chemistry of Materials, 2005, 17, 3075–3085