SYNCHROTRON X-RAY PAIR DISTRIBUTION FUNCTION: A tool to characterize cement gels

Ana Cuesta[a*], Jesus D. Zea-Garcia[a], Diana Londono-Zuluaga[a], Angeles G. De la Torre[a], Isabel Santacruz[a], Miguel A. G. Aranda[a.b]

[a] *Departamento de Química Inorgánica, Universidad de Málaga, Spain.*

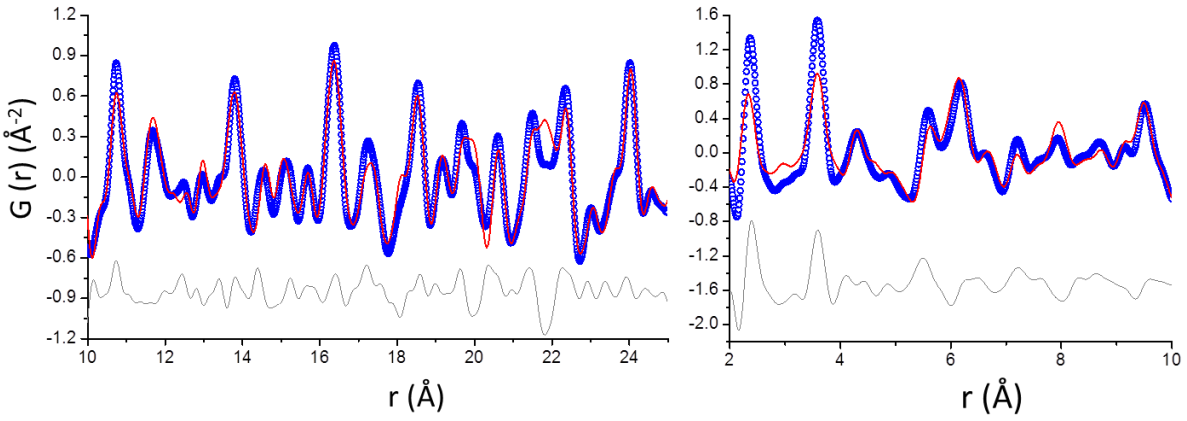
[b] *ALBA synchrotron, Carrer de la Llum, Barcelona, Spain.*

* *a_cuesta@uma.es*

Cement matrices contain large amounts of crystalline phases jointly with amorphous and/or nanocrystalline phases. Consequently, their analyses are very challenging. Synchrotron powder diffraction in combination with the pair distribution function (PDF) methodology is very useful to characterize such complex cement pastes. This work is focused on the study of the short and medium range atomic arrangement(s) in the different nanocrystalline gels which are present in the cement pastes through total scattering Pair Distribution Function quantitative phase analyses.

Powder diffraction data of cement pastes were collected in BL04-MSPD beamline at ALBA Synchrotron (Barcelona, Spain). They have been analyzed by PDF and Rietveld methodologies in order to determine the nanocrystalline and microcrystalline contents, respectively. This methodology allows obtaining a better understanding of the binding gels. Three sets of hydrated model samples have been studied: i) monocalcium aluminate, $CaAl_2O_4$, the main component of calcium aluminate cements [1,2], ii) ye'elimite, $Ca_4Al_6SO_{16}$, the main component of calcium sulfoaluminate cements [2] and iii) tricalcium silicate [2,3], Ca_3SiO_5, the main component of Portland cements. For i) and ii) samples, the influence of some selected parameters were investigated; for instance, different water-to-solid (w/s) mass ratios and temperatures. Moreover, the diameter of the nanoparticles of the aluminum hydroxide nanocrystalline gels have been investigated and compared in these pastes.

For the PDF analyses of Ca_3SiO_5 pastes [2,3], a multi r-range approach was followed: a higher r-range, above 40 Å, was used to determine the microcrystalline phase contents, portlandite and residual alite; then, the intermedium r-range (10-25 Å) allowed characterizing the atomic ordering in the nanocrystalline gels; and finally, the low r-range, below 10 Å, gave insight about the chemical nature of the amorphous component. It is important to highlight that the analysis of the data in the 10-25 Å r-range indicated that a defective clinotobermorite crystal structure fitted the total scattering data of C-S-H gel better than others crystal structures, Figure left. Moreover, the analysis of the PDF data in the 2-10 Å r-range showed that an amorphous component was needed to justify the residual scattering data. This contribution was justified with monolayer calcium hydroxide, Figure right.



[1] Cuesta, A., et al. *Cem. Concr. Res.,* 2017, 96, 1, doi: 10.1016/j.cemconres.2017.02.025

[2] Cuesta, A., et al. *Crystals,* 2017, 7, 317, doi: 10.3390/cryst7100317

[3] Cuesta, A., et al. *Sci. Rep.,* 2018, 8, 8544, doi: 10.1038/s41598-018-26943-y

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