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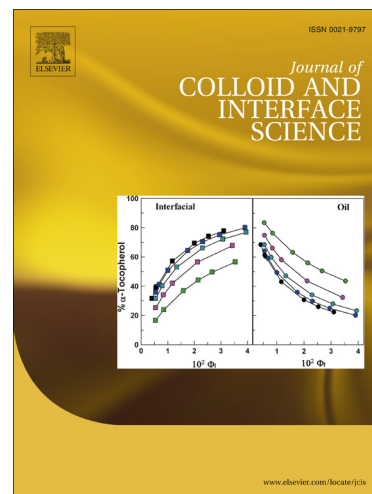
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Precipitation mechanism of amorphous silica nanoparticles: a simulation approach.

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Hypothesis: Despite its importance in numerous industrial and natural processes, many unsolved questions remain regarding the mechanism of silica precipitation in aqueous solutions: order of the reaction, role of silica oligomers, existence of an induction time and characteristics of the particle population. This may be traced back, in past models, to the lack of account of the first stages of nucleation, size dependence of the growth law, and full particle population.

Computational method: A microscopic description of the nucleation and growth of amorphous silica nanoparticles is achieved which reproduces a large set of experimental measurements, under various thermodynamic conditions. The time evolution of the solution supersaturation and of the precipitate characteristics is established.

Findings : A growth law of order 6 allows reproducing experimental results, without being correlated to the presence of silica oligomers in the aqueous solution. The saturation plateaus are shown not to be due to an induction period. The characteristics of the particle population are more complex than assumed by simple precipitation models (Johnson-Mehl-Avrami-Kolmogorov or Chronomal models) and strongly depend on how supersaturation is reached. Such a microscopic approach thus proves to be well suited to elucidate the mechanism of nanoparticle formation in natural and industrial contexts.

Keywords: nucleation and growth, computer simulation, NANOKIN code, amorphous silica, nanoparticles

I. INTRODUCTION

The precipitation of silica nanoparticles occurs in numerous industrial as well as natural processes. For example the formation of silica scales in deep geothermal wells represents a strong limitation to the possibility of extracting heat from high-temperature geothermal waters¹. It also impacts subsurface contaminant remediation, underground radioactive wastes disposals, biomineralization and silicification processes among others².

For these reasons, the mechanism of amorphous silica precipitation has been the subject of intensive research. Results obtained in laboratory have yielded precious information on the role of temperature, pH, ionic strength and initial supersaturation²⁻¹⁰. The whole precipitation process is usually described as a succession of steps. First, silica monomers bind to each others to form small oligomers (dimers, trimers, tetramers, etc) in the aqueous solution, before, eventually, condensed spherical nanoparticles precipitate¹¹⁻²¹. The first stage of monomer association is promoted by increasing the total aqueous $(\text{SiO}_2)_{aq}$ concentration and by decreasing temperature. For the formation of solid particles, in some

cases, the existence of an induction period is invoked, function of temperature and initial supersaturation^{2,4,7,8}. It is followed by a stage, during which the degree of silica supersaturation strongly decreases, which allows the determination of global-reaction rates. Despite intensive studies, conflicting results have been obtained regarding the order of the global reaction, which varies from 1 to 8, depending on the authors and the conditions of precipitation²². In an ultimate stage, Ostwald ripening and aggregation may take place after $[\text{H}_4\text{SiO}_4]$ activity has reached the amorphous silica solubility limit.

It is noteworthy that, in most studies, only the time variation of the total silica aqueous concentration $(\text{SiO}_2)_{aq}$ is recorded, making it difficult to fully characterize the precipitation mechanism, since information on the size distribution of the particles are missing. Thorough experiments performed on a time scale of 20-100 days,⁴ point out that the average molecular weight of the final particles increases from 10^5 to 10^9 molecular units as temperature is increased from 5 to 180°C, which corresponds to an increase of the average particle-radii from 10 nm to ≈ 220 nm. Experiments performed on much smaller time scales, rather find average radii of the order of a few nm^{7,8}, in agreement with earlier speculations². Only recently has Small Angle X-Ray Scattering (SAXS) been used to in-situ determine the time evolution of the mean particle sizes in precipitation experiments induced by neutralization of a high-pH silica solution or fast cooling, allowing a more complete description of the amorphous silica precipitation characteristics^{9,10}.

It is thus timely to apply an advanced simulation technique to decipher the microscopic mechanism of amor-

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