Hydrodynamic constraints on relationships between different types of U deposits in southern China

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Four types of U deposits (granite-, volcanic-, carbonaceoussiliceous-pelitic-, and sandstone-type) have been documented in southern China [1, 2]. The majority of them were formed in late Cretaceous-Tertiary (K-E) [2, 3], much younger than the host rocks, suggesting that most of them did not form from magmatic (Triassic-Jurassic) fluids [2]. Based on their association with K-E red-bed basins, it was proposed that the U deposits are genetically related to basinal fluids, like Proterozoic unconformity-type [3]. Alternatively, mantle-derived CO₂-rich fluids, in relation to crustal extention during K-E, were held responsible [4]. It remains poorly understood why U deposits were not well developed before Cretaceous.

It is proposed here that the paucity of U mineralization in Jurassic and Triassic is related to the compressional tectonic environment, which resulted in impermeable basement and dominantly upward fluid flow in sedimentary basins (Fig. 1a). Such a hydrodynamic regime limited oxidizing fluids near the surface and reduced the opportunity for them to extract U from granites at depth. In contrast, the extentional tectonic environment in K-E increased the permeability of the basement, resulting in "leaky" basins and infiltration of oxidizing fluids into the basement (Fig. 1b), forming U deposits in various basement rocks including granites.



Figure 1: Fluid flow patterns of generic basins in southern China. (a) Triassic-Jurassic model with impermeable basement; (b) Cretaceous-Tertiary model with permeable (upper 1km) basement. Basin 50km wide, 5km deep, duration 100 Ma, 50% sand + 50% shale, topographic relief 250 m, computed with *Basin2*.

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CO₂ rich environments under silica confinement

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A potential approach toward the mitigation of the greenhouse effects of CO_2 release into the atmosphere is based on its geological capture and sequestration, a process that relies on the low hydraulic permeability of caprocks resulting from fluid-substrate interfacial and confinement behaviour. Current studies have mainly focused on the interactions of either water-rich CO_2 or pure CO_2 environments with representative caprock substrates, and the concomitant modeling is based on that assumption, *i.e.*, the impact of the CO_2 -contaminants has rarely been addressed. In fact, due to its source, CO_2 always carries small quantities of 'contaminants' including SO_2 , N_2 , NO_x whose impact in terms of fluid-rock interactions must be considered.

Obviously, the presence of SO_2 in aqueous solution may trigger the formation of sulfurous/sulfuric acids [1], whose immediate consequence is the increase in acidity and the potential for dissolution/precipitation of secondary mineral phases that results in significant changes in substrate porosity and permeability [2]. Unfortunately, thermophysical properties for these mixtures at reservoir conditions are scarce or nonexistent [3], and our microscopic understanding of their interaction with caprock is extremely limited.

For that reason, we carried out a molecular-based study of the behavior of $\rm CO_2$ -rich environments containing common contaminants when in contact with and under severe confinement of silica surfaces, to address fundamental issues, including (a) how the degree of surface hydrophobicity affects the interfacial structure and dynamics, (b) how the overlap of interfacial structures affects the confined fluid composition, and (c) how contaminants affect the preferential adsorption and composition of the interfacial fluid layers.

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