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TOMOGRAPHIC ANALYSIS OF REACTIVE FLOW INDUCED PORE STRUCTURE CHANGES IN COLUMN EXPERIMENTS

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Summary. Dissolution followed by precipitation is a major mechanism in the formation of secondary precipitates in most porous sediments - the precipitates existing as surface coatings or cementing material between grains. Secondary precipitation is of interest as a structure modifying mechanism with the potential to control contaminant transport in the subsurface environment. Quantification of structural change is also a necessary component for the construction of predictive models for effective reaction rates at field scales. We have employed synchrotron X-ray computed microtomography in combination with flow-column experiments *to capture and quantify snapshots in time* of dissolution and secondary precipitation changes in the microstructure of Hanford sediments exposed to simulated caustic waste (bulk Peclet number ~0.3, Peclet-Damkohler ~10^{-5}). Our goal was to document the accompanying structural changes and to identify preferential areas, if any, of dissolution and secondary precipitation at conditions that approach field parameters. The dominant reactions occurring in the flow column were dissolution of quartz, producing silicic acid, and precipitation of the sodium aluminosilicates: Linde type A zeolite, nitrate-sodalite and nitrate-cancrinite. Flow rate in the column decreased by 64% over the course of 106 days. The data show clear decreases in the number of pores and throats accompanied by significant shifts in the distribution of pore and throat sizes. Dissolution induced changes included an increase in the number of larger pores. Precipitation induced changes included reduction in the number of small pores and closure of small throats, with accompanying reduction in pore coordination numbers and reduction in the number of pore pathways. Pathway tortuosities however, were not dramatically affected.