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In Situ Imaging of Ra-226 Carriers in Phosphogypsum Waste from the Phosphate Industry

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The wet process treatment of phosphate for the manufacture of phosphoric acid produces industrial waste phosphogypsum (PG) at a rate of five tons of PG per ton fertilizer yielded. Florida produces approximately 70% of the phosphate for the United States and 20% for the world and has accumulated more than one billion tons of phosphogypsum. This phosphogypsum is stored in large stacks because it is mildly radioactive (≤ 20 pCi/g). These stacks pose a major environmental hazard, but the remediation is limited by the presence of radium. A critical step for valorization is to isolate and remove the source of radioactivity, principally Ra-226, within phosphogypsum waste. Previous work examined distributions of radioactivity via physical partitioning of phosphogypsum grain sizes, but results were inconclusive. In this study, we use real-time alpha autoradiography (AI4R BeaQuant-s) to locate *in situ* minerals that emit alpha particles. This novel technique generates quantitative heatmap images of alpha particle counts with a 20 µm spatial resolution. The imaged radioactive hotspots were then overlaid above a reflected light image and compared against an EDS chemical map of the same region. The EDS mapping revealed that the radioactive hotspots coincide with prominent CaF_2 grains present within the gypsum matrix. Raster scanning by laser ablation ICP-MS enabled identification of the trace element content of calcium rich regions that were not $CaSO_4$. Fluorine has a first ionization potential too high (17.4 eV) to be significantly ionized by the ICP source and is, therefore, not determinable by laser ablation ICP-MS. We filtered the data using the Ca/S ratio to identify CaF_2 regions by difference with $CaSO_4$. Based on this, CaF_2 is found to be an important carrier of Ba and rare earth elements. Because Ba is a divalent cation with the same charge and similar ionic radius to that of Ra, the presence of Ba corroborates that CaF_2 is a major Ra host within the phosphogypsum. This knowledge allows us to target removal of CaF_2 from phosphogypsum as a means of remediation of Ra-226 with the ultimate goal of valorizing the gypsum.