Use of Sequential Extractions to Determine the Speciation of $^{226}$Ra in Phosphogypsum

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Phosphogypsum is an impure form of gypsum, CaSO$_4\cdot$2H$_2$O, which is formed during the manufacture of phosphoric acid from sedimentary phosphate ore. Although gypsum is a useful construction material, phosphogypsum is not used for this purpose because of its relatively high content of impurities, including $^{226}$Ra (derived from the U-enriched phosphate rock). This has led to the stockpiling of massive amounts of this material, approximately 30 million metric tons per year in Florida alone.

Our research group has pursued several lines of investigation to determine the speciation of $^{226}$Ra in phosphogypsum—the hope is that an adequate understanding of how radium is incorporated into the material will lead eventually to an economically-viable purification process. We report here on our efforts to use a variation of the standard “Tessier” extraction sequence to separate phosphogypsum into 6 fractions: (1) water soluble; (2) ion exchangeable; (3) bound in carbonates; (4) associated with Fe/Mn oxides; (5) organically bound; and (6) associated with the refractory residue. These fractions are defined by the type of reagent used in an attempt to dissolve the target fraction. The divisions are, therefore, operationally defined and subject to the limitations and artifacts associated with this type of approach. Possible artifacts include: non-specificity of a reagent for a particular species; incomplete dissolution of a target phase; and re-adsorption of an analyte after release from the host material.

Results from several phosphogypsum samples suggest that although artifacts occasionally occur, the sequential extraction approach can provide very useful information. Duplicate results from a typical sample illustrate that radium is frequently distributed between the “water soluble” and “Fe/Mn oxide” fractions (Fig. 1). When compared to the mass loss in these same fractions, it can be seen that while about 90% of the phosphogypsum is water soluble, only about 25% of the radium is released in that fraction. The oxide fraction, on the other hand, with negligible mass loss, accounts for the major fraction of the radium in this sample.

![Chart showing Ra-226 and Mass Loss percentages for phosphogypsum sample gyp-41](chart.png)

Fig. 1 Extraction profile for 2 replicate runs of phosphogypsum sample gyp-41. The mass loss diagram represents the amount of mass loss during each extraction.