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Development of solid state MAS NMR techniques for the characterization of alteration products formed during the dissolution of spent nuclear fuel.

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The migration of radionuclides from underground nuclear waste repositories will involve the formation of uranium secondary minerals when ground water reacts with the spent nuclear fuel.

During oxic or anoxic spent fuel dissolution, amorphous phases can precede the development of well-defined crystalline mineral phases. Nuclear magnetic resonance (NMR) can probe the structure of these amorphous alteration products and trace the evolution of secondary phases.

To develop the technique, potential secondary phases of ^{17}O-enriched uranium minerals have been fabricated to establish the oxygen environments of becquerelite, $Ca(UO_2)_6O_4(OH).8(H_2O)$, andersonite, $Na_2Ca(UO_2)(CO_3)3.6H_2O$ and grimselite, $K_3Na(UO_2)(CO_3)_3.(H_2O)$. These contain calcium ($Ca^{2}+$), carbonate ($CO_3{}^{2+}$), and other ubiquitous ions.
Solid state ^{17}O MAS-NMR spectra of uranium minerals have been obtained in a 9.39T magnetic field.

Detailed differences in oxygen local environments such as uranyl bond lengths and interlayer OH and H_2O can be distinguished by analyzing the spectra, which can elucidate the alteration of UO_2 by ^{17}O water.

Summary

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