

# Variable Temperature $^3\text{H}$ Diffusion and $^{237}\text{Np}$ Adsorption with Na- Montmorillonite

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Deep geologic repository (DGR) designs for spent nuclear fuel (SNF) aim to keep radioactive waste emplaced for up to a million years by leveraging multiple barriers to transport. One key part of this design is a clay-based barrier set around the SNF package to prevent groundwater advection and sorb potentially migrating radionuclides. Bentonite clay is a commonly proposed backfill material due to its high swelling capacity and large number of sorption sites. [1] These advantages are due in large part thanks to montmorillonite, a 2:1 smectite clay mineral that makes up the majority of bentonite by weight.

Tritium ( $^3\text{H}$ ) diffusion through Na-montmorillonite has been quantified in duplicate at room temperature via a through diffusion setup (Figure 1). The derived effective diffusion coefficients show excellent agreement both with each other and with literature values derived from similar experimental conditions. [2] Currently,  $^3\text{H}$  diffusion experiments at elevated temperatures are being conducted to evaluate a novel experimental setup using a heating coil to uniformly heat the clay plug to simulate diffusion under temperature conditions expected after closure in a DGR.

The ultimate goal of such experiments has been to validate the experimental setup for eventual through-diffusion experiments with radionuclides relevant to SNF ( $^{99}\text{Tc}$ ,  $^{137}\text{Cs}$ ,  $^{237}\text{Np}$ , and  $^{242}\text{Pu}$ ). Such experiments will be conducted at elevated temperatures as well as under both oxic and anoxic conditions. A full understanding of the transport behavior of these radionuclides requires an understanding of the retardation (*i.e.*, sorption) behavior. To this end, sorption experiments on Na-montmorillonite are also currently being conducted under oxic and anoxic conditions. Already complete are oxic  $^{237}\text{Np}$  adsorption experiments at variable ionic strengths and temperatures. Extended x-ray absorption fine structure (EXAFS) measurements demonstrate that the sorbed Np(V) species do not change as a function of ionic strength or temperature. Thus, one surface complexation species was used to model all batch sorption data.

[1] Zhou, Tong & Yu (2019), In *Nanomaterials from Clay Minerals*, 335-364.

[2] Bestel *et al* (2018), *Applied Geochemistry* 93, 158-166.

