Characterization and Modeling of Materials for Kr-Xe Separations

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ABSTRACT

Several radioactive isotopes, specifically $^3$H, $^{14}$C, $^{129}$I, and $^{85}$Kr, have the potential to be released in a gaseous form during reprocessing of spent nuclear fuel. A comprehensive approach to reprocessing requires that these isotopes be captured and stored until they no longer present a radiation hazard. Unfortunately, there are no practical solutions to several gas-phase separations problems and no generally accepted waste forms for $^{85}$Kr. This proposal specifically addresses the issues associated with radioactive $^{85}$Kr and the short lived Xe isotopes that must be captured during the dissolution and processing of fuel. Xe comprises the bulk of the noble gases produced from the fission of $^{235}$U, but the isotopes are either stable or decay to stable isotopes over short time scales. $^{85}$Kr, on the other hand, has a 10.76 yr half-life. Long-term storage of the mixture is expensive, and release of $^{85}$Kr into the atmosphere is unacceptable. An ideal solution would be to separate the Kr for long-term storage (modest Xe impurities would be acceptable), and release or recover the stable Xe. Unfortunately, no technology currently exists to accomplish this task.

This proposal will carry out research aimed at solutions to both issues. For Kr/Xe separations, we will investigate a variety of potential nanoporous sorbents for the pressure swing adsorption process. Specifically, we will determine sorption isotherms for Kr and Xe, heats of adsorption, and estimate the kinetics for adsorption/desorption. As promising materials are identified, we will investigate the nature of the gas/sorbent interaction in order to develop a fundamental understanding of the mechanism(s) involved. This will allow us to target improved materials for this separations process. In addition, we will examine the feasibility of sequestrating of 85Kr in a nanoporous host. Specifically, we will investigate zeolitic encapsulation with the aim of overcoming the moisture sensitivity of current encapsulates. If successful, this may allow for long-term storage within a concrete-like medium with a pedigree as a waste form.