U.S. Department of Energy

Zeolite Membranes for Krypton/Xenon Separation from Spent Nuclear Fuel Reprocessing Off-Gas

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Program: MS-FC-1: Fuel Cycle R&D

ABSTRACT:

In this 3-year Mission Support project, we will develop a high-performance, low-energy intensity, lower-cost zeolitic membrane process for Kr/Xe separation during spent nuclear fuel (SNF) processing; and investigate the underlying molecular adsorption and transport processes in both idealized and realistic operating conditions to develop reliable membrane synthesis-structure-property relationships. Advanced, cost-effective, and near-complete SNF recycle and waste minimization are critical national needs in nuclear energy production. SNF processing off-gas capture and separation is an important component of these technology requirements. In this context, the present project deals with capture and purification of the long-lived radioisotope ^{85}Kr and stable ^{136}Xe components of process off-gas. There are multiple advantages of sharp ^{85}Kr/Xe separation: large reduction in storage volume and cost, revenue from sale of purified Xe, and compliance with EPA regulations limiting ^{85}Kr emissions and dose.

Existing Kr/Xe separation methods (cryogenic distillation, absorption, solid adsorption) are not considered economical. Membrane processes can have relatively low operating costs, smaller equipment size, and ambient operating temperature. However, Kr/Xe separation with membranes is challenging due to the small physical difference (0.036 nm) in molecular size of the two gases, as well as the lack of radiation robustness of conventional polymeric membranes. Our Georgia Tech-ORNL collaborative team recently obtained preliminary findings that molecular sieving SAPO-34 (silicoaluminophosphate-34) zeolitic membranes show attractive Kr/Xe selectivity >10 and Kr permeance ~20 GPU (0.04 mol/m^2/min/atm). Given this baseline performance, we hypothesize that a multistage zeolite membrane process would be an attractive way to obtain highly pure Kr and Xe streams. However, there are several scientific challenges to be overcome before a well-understood and high-performance SAPO-34 membrane process can be realized for Kr/Xe separations in the nuclear fuel cycle. This project is a targeted effort focused upon four key challenges that require new advances: **(1)** Understanding and control of intrinsic Kr and Xe adsorption/diffusion properties of a well-defined set of SAPO-34 materials of tuned compositions. **(2)** Controlling SAPO-34 membrane thickness and defects in tubular geometry, *via* methods for growth of thin (~1-2 μm) SAPO-34 membranes, as well as characterization and mitigation of membrane defects. **(3)** Quantifying the impact of radiation exposure on SAPO-34 membrane performance through permeation experiments with radioactive off-gas streams with controlled radiation dosing. **(4)** Detailed binary transport measurements with SAPO-34 membranes to validate transport models, and membrane process calculations to determine the estimated system size and cost.

Our project deliverables are: **(1)** Demonstration of a selective (Kr/Xe selectivity > 15), highly Kr-permeable (permeance > 50 GPU), and radiation-resistant zeolitic tubular membrane module; **(2)** New understanding and comprehensive database of Kr and Xe transport and radiation resistance properties of SAPO-34 (CHA) materials and membranes; **(3)** New synthesis methods for thin/submicron SAPO-34 membranes; and **(4)** Accurate process model-based estimates for membrane size and cost for different levels of Kr and Xe purity. Successful completion of this Mission Support project will form a sound basis for further development of a transformative separation technology for the nuclear fuel cycle.