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Multi-component testing using HZ-PAN and AgZ-PAN Sorbents for OSPREY Model validation

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Fuel Cycle Technology

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Material Recovery and Waste Form Development

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SUMMARY

In efforts to further develop the capability of the Off-gas SeParation and RecoverY (OSPREY) model, multi-component tests were completed using both hydrogen mordenite-polyacrylonitrile (HZ-PAN) and silver mordenite-polyacrylonitrile (AgZ-PAN) composite sorbents. The primary purpose of this effort was to obtain multi-component xenon and krypton capacities for comparison to future OSPREY predicted multi-component capacities using previously acquired Langmuir equilibrium parameters determined from single component isotherms.

Experimental capacities were determined for each sorbent using two feed gas compositions of 1000 ppmv xenon and 150 ppmv krypton in either a helium or air balance. Test temperatures were consistently held at 220 K and the gas flowrate was 50 sccm. Capacities were calculated from breakthrough curves using TableCurve® 2D software by Jandel Scientific. The HZ-PAN sorbent was tested in the custom designed cryostat while the AgZ-PAN was tested in a newly installed cooling apparatus.

Previous modeling validation efforts indicated the OSPREY model can be used to effectively predict single component xenon and krypton capacities for both engineered form sorbents. Results indicated good agreement with the experimental and predicted capacity values for both krypton and xenon on the sorbents. Overall, the model predicted slightly elevated capacities for both gases which can be partially attributed to the estimation of the parameters and the uncertainty associated with the experimental measurements.

Currently, OSPREY is configured such that one species adsorbs and one does not (i.e. krypton in helium). Modification of OSPREY code is currently being performed to incorporate multiple adsorbing species and non-ideal interactions of gas phase species with the sorbent and adsorbed phases. Once these modifications are complete, the sorbent capacities determined in the present work will be used to validate OSPREY multicomponent adsorption predictions.

Multi-component testing using HZ-PAN and AgZ-PAN Sorbents for OSPREY Model validation

April 24, 2015

CONTENTS

TABLES

Multi-component testing using HZ-PAN and AgZ-PAN Sorbents for OSPREY Model validation

April 24, 2015

ACRONYMS

AgZ-PAN	Silver converted mordenite- polyacrylonitrile
HZ-PAN	Hydrogen mordenite-polyacrylonitrile
GIT	Georgia Institute of Technology
MOOSE	Multiphysics Object Oriented Simulation Environment
OSPREY	Off-gas SeParations REcoverY Model
UNF	Used Nuclear Fuel

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April 24, 2015

1. INTRODUCTION

The release of volatile radionuclides generated during Used Nuclear Fuel (UNF) reprocessing in the US will likely need to be controlled to meet US regulatory emission limits.

A US DOE sponsored Off-Gas Sigma Team has been tasked to investigate emissions and immobilization control technologies for the volatile radioactive species generated from commercial UNF reprocessing.

The physical adsorption process is currently being evaluated for the capture of Kr and Xe at INL. This work has resulted in a novel composite sorbent development procedure using commercially available mordenite powder as the active material bound within polyacrylonitrile (PAN). (1)

Utilizing this sorbent development procedure, INL sigma team members have developed two composite sorbents that have been evaluated for Kr and Xe capacities for a multitude of feed gas compositions and temperatures. Recently, test results supporting modeling efforts including isotherm generation for Kr and Xe have been reported. (2) The model validation for single component systems indicates the Off-gas SeParation and RecoverY (OSPREY) model can successfully be used to predict capacities for Kr and Xe at 220 K. Overall, the model predicted slightly elevated capacities for both Kr and Xe on the sorbents which can be partially attributed to the estimation of the parameters and the uncertainty associated with the experimental measurements. The document herein provides multi-component Kr and Xe capacities that have been determined experimentally for future comparison against OSPREY model predicted values.

2. PURPOSE AND SCOPE

The primary purpose of this testing was to experimentally determine Kr and Xe capacities for both hydrogen mordenite- polyacrylonitrile (HZ-PAN) and silver converted mordenite- polyacrylonitrile (AgZ-PAN) sorbents using multi-component gas compositions at 220 K. These capacities are available for comparison against multi-component OSPREY model predicted values. These comparisons can be used to validate OSPREY output when using Langmuir equilibrium parameters experimentally determined from single-component adsorption isotherms previously generated. (2) The scope of this effort was to perform two capacity tests and document capacity results for each sorbent using two individual feed gases comprised of 1000 ppmv Xe and 150 ppmv Kr in a He or air balance.

2.1 OSPREY Model Review

The OSPREY model has been developed to solve the fundamental transport equations for adsorption in a packed bed in order to obtain a predictive unit operations model for the separation of off-gas constituents. It is a dynamic fully coupled off-gas adsorption model that has been developed in the MOOSE framework to solve off-gas separation systems of equations simultaneously in a fully implicit manner using finite element methods. OSPREY models the adsorption of off-gas constituents for dispersed plug flow in a packed bed under non-isothermal and non-isobaric conditions. Inputs to the model include gas composition, sorbent and column properties, equilibrium and kinetic data, and inlet conditions. The simulation outputs component concentrations along the column length as a function of time from which breakthrough data can be obtained. The breakthrough data can be used to determine bed capacity, which in turn can be used to size columns. In addition to concentration data, the model predicts temperature along the column length as a function of time and pressure drop along the column length.

Currently, the OSPREY model has been modified to allow the user to specify how many components are included in the system and whether or not each component adsorbs. (3) However, OSPREY is currently configured such that one species adsorbs and one does not (i.e. krypton in helium). Introducing multiple adsorbing components requires significant modification to the OSPREY module of MOOSE. Collaboration is ongoing with the Georgia Institute of Technology (GIT) to integrate OSPREY with fundamental level models developed by GIT to describe the equilibria and kinetics of multicomponent adsorption. Modification of OSPREY is currently being evaluated to incorporate multiple adsorbing

species and non-ideal interactions of gas phase species with the sorbent and adsorbed phases. Once these modifications are complete, the sorbent capacities determined in the present work will be used to validate OSPREY multicomponent adsorption predictions.

3. EXPERIMENTAL AND RESULTS

Capacity tests for HZ-PAN using the two test gases were completed using the custom designed cryostat. Capacity tests for AgZ-PAN were performed using these same feed gases in a newly acquired cooling apparatus described previously. (4)

3.1 Capacity Testing

The capacity tests for both sorbents were performed at 220 K with gas flowrates of 50 sccm. These parameters were selected to maintain consistency with the single component testing and OSPREY parameters.

At the conclusion of each capacity test, a sorbent bakeout was performed by flowing UHP He at 50 sccm through the column heated to ~ 360 K to desorb the Kr and Xe from the column. The effluent gas was monitored with a gas chromatograph (GC) to ensure the Kr and Xe had been completely removed prior to initiating the next test.

3.1.1 Capacity Test Results

Table 1 includes the associated capacities obtained for each feed gas composition at 220 K for HZ-PAN.

Table 1. Kr and Xe capacities for both feed gas compositions using HZ-PAN

Feed Gas	Xe Capacity (mmol/kg)	Kr Capacity (mmol/kg)
1000 ppmv Xe, 150 ppmv Kr in He balance	550	8.65
1000 ppmv Xe, 150 ppmv Kr in air balance	264	3.26

Table 2 includes the associated capacities obtained for each feed gas composition at 220 K for AgZ-PAN.

Table 2. Kr and Xe capacities for both feed gas compositions using AgZ-PAN

Feed Gas	Xe Capacity (mmol/kg)	Kr Capacity (mmol/kg)
1000 ppmv Xe, 150 ppmv Kr in He balance	443	3.04
1000 ppmv Xe, 150 ppmv Kr in air balance	254	1.36

These capacities are provided for use in future OSPREY modeling development efforts. The capacities using the feed gas with air balance are provided with the intent that ultimately the model will be capable

of predicting capacities for components in the presence of air, potentially mitigating the need to generate additional adsorption isotherms for air streams.

4. CONCLUSIONS

Experimentally determined capacities for both Kr and Xe using HZ-PAN and AgZ-PAN sorbents with multi-component gas compositions at 220 K have been reported. These capacities are available for comparison against multi-component OSPREY model predicted values. These comparisons can then be used to validate OSPREY output when using Langmuir equilibrium parameters experimentally determined from single-component adsorption isotherms previously generated.

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