

Pillared Nanosheets for High Level Waste Management and Environmental Remediation in the Nuclear Fuel Industry

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Nuclear energy currently accounts for approximately 10% of the world's fuel consumption, with the United States generating 8% of its annual total energy from nuclear fuel sources [1]. However, with increased environmental and economic pressure to decrease CO₂ emissions and lessen our reliance on foreign petroleum and natural gas, it appears that nuclear power will remain one of the critical energy sources in coming decades. This is further indicated by the approval in February 2012 of the first new reactor in the US since the Three Mile Island disaster [2]. Despite these demands, a solution to the long-term storage of high level wastes generated in the nuclear fuel cycle and reprocessing of wastes has not been well established. Coupled with this issue is an inability to effectively capture radioactive wastes once they have been released to the environment, making the devastating effects of nuclear contamination long lasting and widespread.

Two of the most plentiful nuclear fuel wastes, cesium-137 and strontium-90, are also two of the most difficult to capture. Due to their chemical similarities to potassium and calcium respectively, these isotopes can easily make their way through environments to food and water sources in affected areas. These isotopes also have relatively long half-lives, making the capture and storage of these species of heightened importance.

Pillared nanosheets may offer a potential solution to both the issues of environmental contamination and long term storage of high level nuclear wastes. Pillared nanosheets consist of a hierarchical structure of inorganic layers of approximately 1nm thickness spaced by inorganic pillars; due to the high porosity within and between the nanosheets, high density of ion-exchange sites, controllable thermal stability, and controllable channel width and height these materials may be more efficient at capturing large amounts of nuclear wastes than methods currently employed. Pillaring of nanosheet layers occurs in a three-stage process, consisting of hydrothermal synthesis of the nanosheet precursor, swelling of the material through the introduction of long chain organics and silica containing groups, and calcination. The spaces created between the organic moieties and the layers become a template for the pillaring process, allowing us to create a periodic structure of nanosheets with pore channels of tunable dimensions. The choice of the organic linkers, which have various chain lengths and functional groups, will allow for the physical and chemical control of the pore space.

Aluminosilicate layers of MCM-22 (P), the precursor of the synthetic zeolite MCM-22, will be pillared and tested as a potential solid ion-exchange material for simulated waste streams. The stability of this material in simulated contamination conditions (high sodium/calcium concentrations, low strontium/cesium fractions, mild pH and temperature) as well as simulated fuel reprocessing/long term storage conditions (low pH, high temperature, high strontium/cesium fractions) will be tested. In parallel the creation of a novel nanosheet material, pillared AMH-3, will also be explored. AMH-3, a unique nanosheet precursor containing 3-dimensionally connected diffusion channels within the layers, was developed and demonstrated to swell with various organics by our group [3, 4].

The proposed synthesis of pillared nanosheet materials composed of AMH-3 and MCM-22 layers connected by silicon dioxide bonds with tunable large channel dimensions will be presented. These materials will be tested for their efficacy as cation-exchange adsorbents for radioactive waste streams and contaminated environments.

[1]- DOE/EIA Annual Energy Review 2011

(<http://www.eia.gov/totalenergy/data/annual/pdf/aer.pdf>)

[2]-NRC Combined License Vogtle Electric Generating plant units 3 and 4

(<http://pbadupws.nrc.gov/docs/ML1129/ML112991110.pdf>)

[3]- Choi, S. Coronas, J. *Layered Silicates by Swelling of AMH-3 and Nanocomposite Membranes*, *Angewandte Chemie International Edition* 2008, 47, 552-555

[4]- Choi, S. Coronas, J. *Layered Silicates by proton exchange and swelling of AMH-3*, *Microporous and Mesoporous Materials* 2008, 115, 75-84