PRODUCTION OF CERIUM OXIDE MICROSPHERES BY AN INTERNAL GELATION SOL-GEL PROCESS

A Thesis

by

JEFFREY JOHN WEGENER

Submitted to the Office of Graduate Studies of
Texas A&M University
in partial fulfillment of the requirements for the degree of

MASTER OF SCIENCE

December 2008

Major Subject: Nuclear Engineering

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Approved by:

Chair of Committee,	Sean McDeavitt
Committee Members,	John Poston
	Miladin Radovic
Head of Department,	Raymond Juzaitis

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ABSTRACT

Production of Cerium Oxide Microspheres by an Internal Gelation Sol-Gel Process.

(December 2008)

Jeffrey John Wegener, B.S., Purdue University

Chair of Advisory Committee: Dr. Sean M. McDeavitt

The experiments performed for this research were completed to produce solid cerium oxide microspheres by an internal gelation sol-gel process. The motivation for this work was to develop a process that would enable the fabrication of a storage or transmutation form for the plutonium and transuranics (TRU) from the Uranium Extraction Plus (UREX+) used fuel reprocessing process. This process is being investigated by the Department of Energy (DOE) and the Advanced Fuel Cycles Initiative (AFCI) through the Nuclear Energy Research Initiative.

The internal gelation production of cerium oxide involves the combination of hexamethylenetetramine (HMTA), urea, and cerium nitrate solutions at ~100^{o}C. Microspheres were produced by injection of a broth solution into a flowing stream of hot silicone oil. The captured microspheres were aged, washed, and then underwent Thermogravimetric Analysis (TGA), Differential Scanning Calorimetry (DSC), and X-Ray Diffraction (XRD) analysis. The process variables examined in this study include the concentrations of HMTA, urea and cerium nitrate, the process temperature, the post-gelation aging time, and the product washing conditions.

Over a series of 70 experiments, it was determined that a broth solution containing a mixture of 1.45 M cerium nitrate and 1.65 M HMTA and urea (1:1 ratio) solutions produced the best cerium oxide microspheres. The spheres were aged for 30 to 60 minutes and then washed in hexane to remove the silicone oil and a subsequent series of ammonium hydroxide washes to remove unreacted product and to fully gel the microspheres.

Through DSC analysis it was determined that excess wash or unreacted product may be removed by an exothermic reaction at approximately 200^{o}C. The XRD analysis of unheated spheres showed the presence of cerium oxide with additional cerium-bearing organics. Following heating, the microspheres were completely converted to cerium oxide.

DEDICATION

This thesis is dedicated to my parents John and Susan Wegener, my brother Michael Wegener, and my sister Sarah Wegener.

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NOMENCLATURE

AFCI	Advanced Fuel Cycle Initiative
DSC	Differential Scanning Calorimetry
DOE	Department of Energy
HMTA	Hexamethylenetetramine
MOX	Mixed Oxide
NERI	Nuclear Energy Research Initiative
ORNL	Oak Ridge National Laboratory
PUREX	Plutonium and Uranium Extraction
RTV	Room Temperature Vulcanizing
TGA	Thermogravimetric Analysis
TRISO	Tristructural-Isotropic
TRU	Transuranic
TRUEX	Transuranic Extraction
UREX	Uranium Extraction
XRD	X-Ray Diffraction

TABLE OF CONTENTS

Page