Applied Radiation and Isotopes 105 (2015) 52–59

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

A study on artificial rare earth (RE_2O_3) based neutron absorber

Kyung-O KIM [a], Jong Kyung KIM [b,*]

[a] Korea Atomic Energy Research Institute, 1045 Daedeok-daero, Yuseong-gu, Daejeon 305-353, Republic of Korea
[b] Department of Nuclear Engineering, Hanyang University, Seoul 133-791, Republic of Korea

HIGHLIGHTS

- Quantitative analysis of rare earth elements in PWR spent fuels.
- Extraction of artificial rare earth compound using pyroprocessing technology.
- Characteristic analysis of artificial rare earth elements.
- Performance evaluation of artificial rare earth for criticality control.

ARTICLE INFO

Article history:
Received 10 March 2014
Received in revised form
14 November 2014
Accepted 9 July 2015
Available online 11 July 2015

Keywords:
Artificial rare earth compound
Neutron absorber
Spent fuel storage system
Criticality control
Pyroprocessing technology

ABSTRACT

A new concept of a neutron absorption material (i.e., an artificial rare earth compound) was introduced for criticality control in a spent fuel storage system. In particular, spent nuclear fuels were considered as a potential source of rare earth elements because the nuclear fission of uranium produces a full range of nuclides. It was also found that an artificial rare earth compound (RE_2O_3) as a High-Level Waste (HLW) was naturally extracted from pyroprocessing technology developed for recovering uranium and transuranic elements (TRU) from spent fuels. In this study, various characteristics (e.g., activity, neutron absorption cross-section) were analyzed for validating the application possibility of this waste compound as a neutron absorption material. As a result, the artificial rare earth compound had a higher neutron absorption probability in the entire energy range, and it can be used for maintaining sub-criticality for more than 40 years on the basis of the neutron absorption capability of Boral™. Therefore, this approach is expected to vastly improve the efficiency of radioactive waste management by simultaneously keeping HLW and spent nuclear fuel in a restricted space.

1. Introduction

Designs for spent fuel storage systems need to incorporate safety features to maintain subcriticality, remove residual heat from spent fuel, and provide radiation protection over the anticipated lifetime of the involved facilities. Overall, the fundamental safety objective of these systems is to ensure that subcriticality is always maintained. Thus, additional safety elements such as neutron absorbers have been introduced in the design of these facilities, as subcriticality cannot be maintained in a spent fuel configuration alone. There are currently a limited number of commercially available neutron absorbers that are suitable for use in licensed spent fuel storage systems. Among the available systems, Boral™ (Subramanian et al., 2010) and NeutroSorb Plus™ (Fix et al., 2004) are widely used as supplements to criticality control. However, each of these absorbers has one or more disadvantages with respect to fabrication, mechanical properties, material aging, and cost (Villarreal et al., 1996; Licina, 2000). In the case of Boral™, which has an excellent boron loading capability and is a very effective neutron absorber, its disadvantages include very poor mechanical and thermal aging performance. On the other hand, NeutroSorb Plus™ is easily fabricated and possesses good mechanical properties, but has poor boron loading capability, short of using enriched B^{10}. Furthermore, these two materials are also very expensive; Boral™ costs approximately \$500 per sheet while NeutroSorb Plus™ had a cost of approximately \$100 per pound 20 years ago, which substantially impacts the overall cost requirements of facilities employing these materials. Indeed, current neutron absorbers represent about 25% of the manufacturing cost of storage and transport systems.

In the present study, spent nuclear fuels were considered as a potential source of rare earth elements because the nuclear fission

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* Corresponding author. Fax: +82 2 2294 4800.
E-mail address: jkkim1@hanyang.ac.kr (J. Kyung KIM).

http://dx.doi.org/10.1016/j.apradiso.2015.07.008

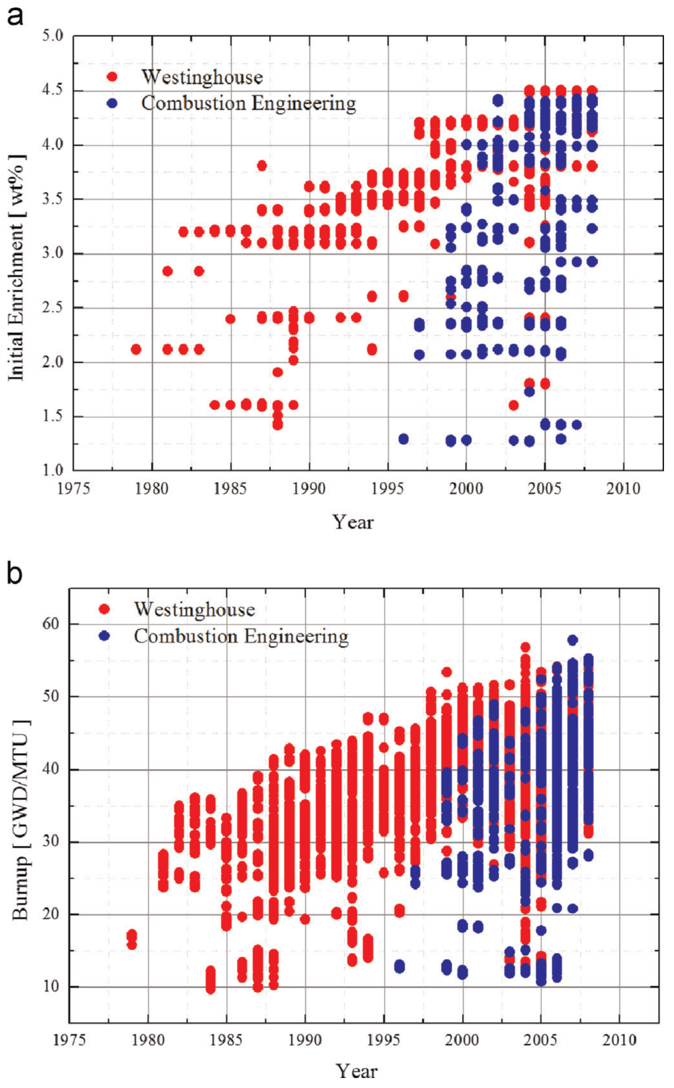

Fig. 1. Initial enrichment and burnup trends of spent nuclear fuels produced from korean nuclear power plants. (a) Initial Enrichment Vs Discharged Year (b) Burnup Vs Discharged Year.

of uranium produces a full range of nuclides. In particular, rare earth compounds (RE_2O_3) are regarded as preferable for this work because these compounds contain a large amount of strong thermal neutron absorbers such as europium (σ_a: ~4600 barn), gadolinium (σ_a: ~49,000 barn), samarium (σ_a: ~5800 barn), and dysprosium (σ_a: ~930 barn). Indeed, if an efficient technique for extracting specific fission products from cumulative spent fuels can be developed, they may sufficiently replace the reactivity control provided by boron-rich materials. Thus, this study is the first to investigate spent nuclear fuels generated by Korean nuclear power plants (except for CANDU reactors). Based on this information, a series of depletion calculations were performed for a quantitative analysis on the material composition of fission products and the total amount of cumulative rare earth elements. In addition, the neutron absorption cross-section of artificial rare earth compounds, derived from the MCNPX code (Denise and Pelowitz, 2005), was compared with those of other boron-rich materials, and the change of their neutron absorption capability was researched as a function of the period of neutron irradiation.

Table 1
SAS2H Calculation parameters for analyzing the material composition of fission products and cumulative amount of rare earth elements

	Westinghouse (17 × 17)	Combustion engineering (16 × 16)
Pellet composition		
U-234	0.034	0.035
U-235	3.82	3.98
U-236	0.018	0.018
U-238	96.128	95.967
Effective radius [cm]		
R_1 (Borated water)	0.562	1.145
R_2 (Clad)	0.602	1.245
R_3 (Borated water)	0.711	1.450
R_4 (Fuel material)	2.417	4.981
R_5 (Borated water)	2.426	5.191
Specific power [kW/kgU]	38.30	38.30
Burnup [GWD/MTU]	40.06	42.08
Cooling time [Year]	11.53	6.09

Table 2
Classification of nuclide inventory in domestic spent nuclear fuels

	Element	Mass [ton]	Total [ton]
Uranium	U	3.16E+03	3.16E+03
TRU	Np	1.97E+00	3.97E+01
	Pu	3.49E+01	
	Am	2.73E+00	
	Cm	1.24E-01	
Noble metals	Mo	9.84E+00	3.00E+01
	Ru	9.13E+00	
	Rh	1.90E+00	
	Pd	5.68E+00	
	Ag	3.03E-01	
	Tc	3.17E+00	
Rare earth elements	Y	1.83E+00	4.11E+01
	La	4.95E+00	
	Ce	9.63E+00	
	Pr	4.54E+00	
	NdPm	1.56E+015.73E-02	
	Sm	3.38E+00	
	Eu	5.49E-01	
	Gd	5.01E-01	
	Others	1.44E-02	
Alkali metals	Sr	3.05E+00	1.32E+01
	Cs	1.01E+01	
Halogens	I	8.04E-01	8.88E-01
	Br	8.43E-02	

2. Quantitative analysis of rare earth elements in PWR spent fuels

In Korea, a total of 21 nuclear power plants are currently operated between four sites (Kori, Yeonggwang, Ulchin, and Wolsong), all of which are Pressurized Water Reactors (PWR) except for the four CANDU heavy water reactors at Wolsong (Feiveson et al., 2011; Whang, 2011). A variety of nuclear fuel assemblies have also been developed for the PWR type of nuclear power plant; however, only two types, namely the WH 17 × 17 and CE 16 × 16 assemblies, are being used at a significant scale in Korea. Hence, these assemblies, manufactured by Westinghouse (WH) and Combustion Engineering (CE), are regarded as representative nuclear fuels in our country.

At the end of 2008, 10,948 tHM of spent fuel had been discharged from domestic PWRs and CANDU reactors and re-located