Annals of Nuclear Energy 69 (2014) 278–291

Contents lists available at ScienceDirect

Annals of Nuclear Energy

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

Review and characterization of best candidate isotopes for burnup analysis and monitoring of irradiated fuel

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ARTICLE INFO

Article history:
Received 24 September 2013
Received in revised form 19 December 2013
Accepted 23 December 2013
Available online 14 March 2014

Keywords:
Nuclear fuel
Burnup credit
Gamma spectroscopy
Half lives

ABSTRACT

This research is an extension of feasibility study of MOX fuel online burnup analysis. A multitude of fission products identified as candidates have been scrutinized for their suitability of burnup analysis and spent fuel analysis. Best isotopes obtained for analysis by investigating half-life, fission yield, branching ratios, production modes, thermal neutron absorption cross section and fuel matrix diffusivity. ^{132}I and ^{97}Nb are identified as good isotope candidates for on-line burnup analysis. ^{132}I is also a good candidate for plutonium/uranium discrimination due to the large difference in the fission yield of the isotope. For interim storage monitoring the well-established cesium isotopes appears to be the best choices unless the data gaps are addressed. Other alternate for cesium for interim monitoring is ^{131}I, ^{140}La, and ^{95}Nb at the present time. Selection of one over the other choice must be made based on application. For the long-term storage monitoring ^{94}Nb is the only attractive candidate. It has a low diffusion rate of $\sim 10^{-11}$ cm^2/s, an almost zero neutron absorption cross section making it burnup history independent and decent gamma yield of 1.44E−09. In addition, the paper also identifies the data gaps for developing a robust burnup analysis tool using gamma spectroscopy.

1. Introduction

Between 1977 and 1981 commercial reprocessing was banned by the United States government, interrupting the U.S. reprocessing industry. The U.S. moratorium on reprocessing did not, however, discourage European countries such as France from developing the technology and incorporating Mixed OXide (MOX) fuel into their nuclear fuel cycle. While other nations pursued reprocessing, the U.S. chose to invest in the Yucca Mountain Nuclear Waste Repository. However, with Yucca Mountain no longer under consideration, the U.S. is reconsidering its fuel cycle options, and taking a second look at reprocessing and MOX fuel (Nuclear waste repository safe for future generations, 2010; Energy Institute, 2010).

Decades of operation in France have demonstrated that MOX fuel can be incorporated into a UO_2 fueled core without risking the safety of the plant or increasing the risk of proliferation (Shaw Areva MOX Services, LLC, 2008). Now the U.S. power industry is investigating the fuel. In 2005, Duke Energy commissioned AREVA to build four MOX fuel assemblies for the Catawba Nuclear Station. The Megatons to Megawatts program is another major commitment by the U.S to incorporate MOX into the fuel cycle. This program is an agreement between the United States and Russia to dispose of approximately 35 metric tons (MT) of weapons-grade plutonium by converting it into MOX fuel to be burned in commercial nuclear power plants. The MOX fuel assemblies will be manufactured at the Department of Energy's Savannah River Site through a contract with Duke COGEMA Stone & Webster. As of July 2012, this program has converted 450 MT of weapons-grade uranium to low-enriched fuel for power reactors (United States Enrichment Corporation, 2013).

Thorium fueled reactors are also under consideration because of the advantages of the thorium fuel cycle. Thorium is abundant in the earth's crust. With roughly four times the concentration of uranium, it can be found across the globe. And because ^{238}U is not usually present in thorium fuel there are fewer transuranic elements in the spent fuel (International Atomic Energy Agency, 2005).

Regardless of how the U.S. fuel cycle changes, better tools and techniques for measuring burnup and monitoring spent fuel will be required. One attractive option for non-destructively examining spent fuel is gamma spectroscopy. In this work, a multitude of fission products identified as candidates for burnup analysis and spent fuel monitoring, as proposed by Dennis and Usman (2006), are scrutinized for their suitability for such a program. Consideration is given to half-life, fission yield, branching ratios, thermal neutron absorption cross section, production modes and fuel

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http://dx.doi.org/10.1016/j.anucene.2013.12.014
0306-4549/

matrix diffusivity. Based on these parameters, the best isotopes for non-destructive burnup analysis and spent fuel monitoring have been identified.

2. Traditional tools, techniques and conventional Isotopes for burnup analysis

A report was published by US Nuclear Regulatory Commission (Bevard et al., 2009) discussing various aspects of spent fuel monitoring and the requirements of the measurements system. There are two broad groups of techniques available; "passive" measurement of delayed neutrons and gamma and "active" interrogation by either a pulsed neutron source or a weak steady state source and subsequently monitoring the gamma and/or neutron emissions. For safe transportation of spent nuclear fuel it is critical to have a reliable and efficient monitoring system capable of on-site, accurate measurement of the spent fuel to ensure compliance with the safety criteria. Gamma measurement is one non destructive technique (NDA) for spent fuel monitoring, analyzing the radiation emitted by irradiated nuclear fuel assemblies to determine parameters such as burnup, cooling time, and a qualitative verification of the irradiation history (Håkansson et al., 1993). These parameters can be calculated with the help of flux measurements in the core and following the path of the fuel assembly as it proceeds with its burnup cycle. These calculations can be experimentally verified by the above mentioned techniques. (Willman et al., 2006a) have investigated ^{137}Cs ($T_{1/2}$ = 30.1 years), ^{134}Cs ($T_{1/2}$ = 2.1 years) and ^{154}Eu ($T_{1/2}$ = 8.6 years) as indictors of cooling time and irradiation history since these isotopes dominate the gamma spectrum of spent fuel after five years of cooling time (Bevard et al., 2009). Moreover, ^{137}Cs has very small neutron absorption cross sections and hence negligible burnup history dependence and its fission yields from ^{235}U and ^{239}Pu is approximately the same. (Willman et al., 2006b) has reported the success of using ^{154}Eu while (Dennis and Usman, 2010) reported the potential of using ^{106}Ru. Because of their relatively long half-lives, these isotopes are most useful for analyzing used fuel after about 10 years of cooling. Focus of this manuscript is to analyze new isotopes for their suitability as a burnup indicator. The conventional fission isotopes (^{137}Cs, ^{154}Eu, ^{106}Ru, etc.) already considered for burnup

Table 1
Reference isotopes for burnup analysis and spent fuel monitoring (Dennis and Usman, 2006).

Simulation energy (keV)	Potential isotope	Energy (MeV)	Photon emission probablity	Simulation energy (keV)	Potential isotope	Energy (MeV)	Photon emission probablity
76.6110	^{243}Am	0.07467	0.66	617.82	^{108m}Ag	0.61437	0.90393
	^{206}Bi	0.074969	0.54146		^{43}K	0.61749	0.80514
	^{61}Co	0.067412	0.85		^{190m}Os	0.61608	0.9862
	^{73}Se	0.067	0.7729601		^{144}Pm	0.61801	0.98597
	^{44}Ti	0.07838	0.97619	626.15	^{43}K	0.61749	0.80514
93.2640	^{49}Cr	0.090639	0.532		^{144}Pm	0.61801	0.98597
101.5900	^{67}Ga	0.93311	0.357		^{148m}Pm	0.62997	0.88998
134.9000	^{99m}Tc	0.14051	0.8907	659.46	^{137m}Ba	0.66165	0.8998
143.2200	^{99m}Tc	0.14051	0.8907		^{132}Cs	0.66769	0.97423
	^{85m}Kr	0.15118	0.75278		^{130}I	0.66854	0.96129
159.8700	^{52}Fe	0.016868	0.966		^{132}I	0.66769	0.987
	^{56}Ni	0.15838	0.98795		^{97}Nb	0.6579	0.9809
176.5300	^{52}Fe	0.016868	0.966		^{126}Sb	0.66633	0.99619
	^{111}Ln	0.17128	0.9024	667.78	^{137m}Ba	0.66165	0.8998
193.1800	^{90}Y	0.20251	0.96631		^{98}Tc	0.65241	0.99745
	^{166m}Ho	0.18442	0.726		^{132}Cs	0.66799	0.97423
	^{190m}Os	0.18673	0.702		^{130}I	0.66854	0.96129
226.4900	^{85m}Sr	0.23169	0.84725		^{132}I	0.66769	0.987
	^{132}Te	0.22816	0.88	692.76	^{97}Nb	0.6579	0.9809
276.4400	^{203}Hg	0.27919	0.773		^{126}Sb	0.66633	0.99619
	^{203}Pb	0.27919	0.768		^{94}Nb	0.70293	1
318.0800	^{192}Ir	0.31651	0.82853	717.74	^{144}Pm	0.69649	0.99492
	^{5}1Ti	0.32008	0.929		^{126}Sb	0.695	0.99619
	^{157}Dy	0.32616	0.938		^{108m}Ag	0.72295	0.90499
326.4000	^{194m}Ir	0.32845	0.929	751.05	^{166m}Ho	0.71169	0.54087
	^{51}Ti	0.32008	0.929		^{126}Sb	0.7205	0.53794
	^{157}Dy	0.32616	09.38		^{244}Am	0.746	0.67
359.71	^{73}Se	0.3611	0.965		^{52}Mn	0.74421	0.9
	^{190m}Os	0.36109	0.9488		^{97m}Nb	0.74336	0.9796
	^{131}I	0.36448	0.81164	759.37	^{98}Tc	0.74535	0.99819
376.36	^{43}K	0.37276	0.87273		^{95}Zr	0.75671	0.55345
	^{204m}Pb	0.89915	0.99164		^{95}Nb	0.76579	0.99808
	^{200}Tl	0.36794	0.873	767.7	^{95}Tc	0.76579	0.9382
434.64	^{108m}Ag	0.43393	0.89881		^{95}Zr	0.75671	0.55345
	^{202}Tl	0.43956	0.915		^{82}Br	0.77649	0.8331
	^{69m}Zn	0.43863	0.94889		^{132}I1	0.77261	0.76196
484.6	^{181}Hf	0.48203	0.825	792.68	^{95}Nb	0.76579	0.99808
	^{194m}Ir	0.48286	0.97		^{95}Tc	0.76579	0.9382
	^{87}Y	0.4847	0.9394	809.33	^{134}Cs	0.79584	0.854
	^{90m}Y	0.47953	0.9099		^{210}Tl	0.7997	0.9896
492.93	^{190}Os	0.50255	0.9778		^{206}Bi	0.8031	0.9889
	^{103}Ru	0.49708	0.889		^{58}Co	0.81076	0.9943
	^{87}Y	0.4847	0.9394		^{136}Cs	0.8185	0.997
534.56	^{130}I	0.53609	0.99		^{166m}Ho	0.81031	0.57136
	^{133}I	0.52987	0.8632		^{56}Ni	0.81185	0.85996
	^{135m}Xe	0.52656	0.80997	1592	^{96}Tc	0.81254	0.81803
601.17	^{134}Cs	0.6047	0.976		^{210}Tl	0.7997	0.9896
	^{124}Sb	0.60271	0.978001		^{140}La	1.5965	0.9549