Progress in Nuclear Energy 72 (2014) 5–10

Contents lists available at ScienceDirect

Progress in Nuclear Energy

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

Thorium fuel production and results from beginning of life irradiation

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ARTICLE INFO

Article history:
Received 20 August 2013
Accepted 21 August 2013

Keywords:
Thorium
Halden reactor
Test irradiation
Fuel production

ABSTRACT

An evolutionary, rather than a revolutionary approach to thorium fueled reactors is proposed with an introduction of thorium as the fertile component in mixed oxide fuel for conventional light water reactors. The utility of thorium as a component in today's light water reactors offers improved accident tolerance due to the superior material properties of thorium fuels over conventional uranium fuels. A few notable advantages include better thermal conductivity, higher melting point, higher oxide stability and superior spent fuel characteristics. Consequently, Thor Energy along with a consortium of industrial partners has established a fuel production and irradiation program aimed to license thorium fuels for use in today's light water reactors. Due to the morphology and physical properties of calcined thorium oxide powder, pellet fabrication used for standard uranium oxide fuels must be altered slightly for thorium bearing fuels to yield a product with acceptable theoretical densities, microstructure, and material integrity. At beginning of irradiation life fuel temperature data demonstrates improved fuel characteristics over standard uranium oxide fuel. Fuel centerline operating temperatures are 30–40 K less with a thorium mixed oxide fuel as compared to standard uranium fuel.

1. Introduction

Since the inception of nuclear power, there has been interest in using thorium as the primary fertile component for nuclear fuel. Whereas the initial motivation for the use of thorium was its natural abundance in the earth's crust (3–4 times that of uranium), the utility of thorium is now being revisited due to demands and constraints brought on by a semi-developed nuclear society.

The semi-developed nuclear society is characterized by a dominance of light water technology with a hold-up in the envisioned deployment of fast-breeder technology. Many nations have chosen to responsibly reprocess nuclear waste with hopes of reutilizing the recovered material in fast reactors. The fast reactor hold-up has prompted these nations to develop vast stockpiles of plutonium (reactor grade, RG) and other trans-uranic elements. Furthermore, stockpiles of plutonium (weapons grade, WG) also exist as remnants of large nuclear weapons arsenals currently being decommissioned. Plutonium management strategies including vitrification followed by disposal and inert/MOX fuel utilization in commercial reactors have been examined. Among the recognized disposal methods includes possibly the most responsible alternative utilizing a thorium based fuel matrix, however, a dearth of knowledge exists for its commercial fabrication and in-reactor operational behavior.

1.1. Background

An intermediate solution to plutonium disposition is being utilized which incorporates the combination of plutonium with depleted uranium to produce a mixed oxide (MOX) fuel and irradiating it in a conventional light water reactor (LWR). Conventional MOX fuel, unfortunately, has many disadvantages over conventional uranium oxide (UOX) fuel, including a reduction in fuel thermal conductivity, higher fuel temperatures, higher transuranic isotope concentration in spent fuel and increased fission gas release. An alternative LWR fuel, in which thorium is used as the fertile substance, helps to eliminate many of these concerns and even provide advantages over standard UOX fuel. This fuel is commonly referred to as Th-MOX.

In addition to beneficial material properties, which will be further discussed below, Th-MOX has the benefit of forming a first step towards a broader introduction of thorium in the nuclear fuel cycle where the ultimate goal is a self-sustaining thorium cycle. In such a cycle, only thorium is mined, fissile ^{233}U is bred from thorium and the only waste produced is fission products with a half-life on the scale of hundreds of years. This ultimate scenario can be achieved using a range of different reactor types (for

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0149-1970/$ – see front matter
http://dx.doi.org/10.1016/j.pnucene.2013.08.008

example the popular molten salt reactor), all of which however share the problem of unsolved technical issues, which place their large-scale commercial introduction several decades into the future.

In the near term, thorium used as an additive (less than 10%) in conventional UOX fuel is seen as a mode for the first adoption of thorium in current reactor fuel. The beneficial aspect of this fuel lies partly in the heightened neutron absorption characteristics of thorium over uranium which can be used to significantly offset the need of gadolinium or other neutron poisons, flatten the core power distribution and thereby improve reactor safety margins (Lau et al., 2012). The addition of thorium also benefits the material properties of the fuel leading to higher thermal conductivities, lower fuel temperatures, reduced fuel cracking and reduced fuel swelling (Karam et al., 2008).

As a first step towards the introduction of thorium in LWRs, Thor Energy is conducting an advanced fuel manufacture and irradiation program in order to assess all relevant material properties of the mixed oxides (Pu, Th)O_2 and (Th, U)O_2. The experiment, which goes under the name of Seven-Thirty, which is a reference to the Halden experimental fuel assembly IFA-730 used in this project, is jointly steered and financed by an international consortium of interested partners from all parts of the civil nuclear value chain.

1.2. Material properties of (Pu, Th)O_2

The main advantage of (Pu, Th)O_2 from a material properties perspective is its thermal conductivity, which is higher than that of UOX for Pu fractions lower than about 14%, i.e. for most Pu fractions that may be considered for use in currently operating LWRs (Cozzo et al., 2011). The temperature dependence of the thermal conductivity of (Pu, Th)O_2 is plotted in Fig. 1 along with that of pure UO_2 (Krett and Cleveland, 1997). The higher thermal conductivity gives a lower fuel temperature, which in turn gives a larger margin to fuel melting and lower fission gas release (FGR). The lower FGR has been confirmed for small Pu fractions (Karam et al., 2008) and will be assessed as one of the key parameters in the ongoing irradiation campaign. The need to limit FGR is one of the most severe restraints, which limit the achievable discharge burnup of nuclear fuel, so a lower FGR may have very direct consequences on fuel economy (Insulander Björk et al., 2013). The larger margin to fuel melting, which is a benefit in transient and accident situations, is further improved by the higher melting point of the (Pu, Th)O_2 ceramic, plotted in Fig. 2.

As a chemical substance, thorium dioxide is known to exhibit a heightened level of chemical stability (Dekoussar et al., 2005). This allows thorium based fuels to exhibit better radiation resistance which further improves the FGR characteristics of the fuel, leading to an order magnitude less FGR (Dekoussar et al., 2005). The chemical stability is also beneficial for the purposes of spent fuel storage and accident scenarios, since it makes the fuel matrix less soluble in water. This property of thorium based fuels has been confirmed by leaching experiments (Verwerft et al., 2011).

1.3. Material properties of (Th, U)O_2

(Th, U)O_2 has been much more extensively studied than (Pu, Th)O_2. One reason is that the ultimate thorium cycle will comprise fuel of Th mixed with ^{233}U. However, there are applications of (Th, U)O_2 also in current LWRs, e.g. for power peaking control and reduction of the need for burnable absorbers as mentioned above (Lau et al., 2012) or for natural uranium savings under certain circumstances (Dekoussar et al., 2005).

(Th, U)O_2 also has the benefit of a higher thermal conductivity, as can be seen in Fig. 1. However, data for mixtures with less than 20 weight percent thorium has not been published to our knowledge, and the Seven-Thirty project aims to fill this knowledge gap. The melting point of (Th, U)O_2 is higher than both that of pure UO_2 and that of (Pu, Th)O_2, as seen in Fig. 2, offering the benefit of increased margin to fuel melting also for this fuel type.

The improvement of chemical stability described above applies to (Th, U)O_2 fuel as well as (Pu, Th)O_2 fuel.

2. Pre-studies

In preparation for the fuel fabrication campaign, Thor Energy has undertaken pre-studies in collaboration with Los Alamos National Laboratory and with the Institute of Energy Technology (IFE) in Norway. Experiments have been carried out with a mixture of Th/U and Th/Ce, since Ce has material properties similar to those of Pu (Kim et al., 2008). Trials were made at LANL comprising different powder preparation techniques and pressing and packing

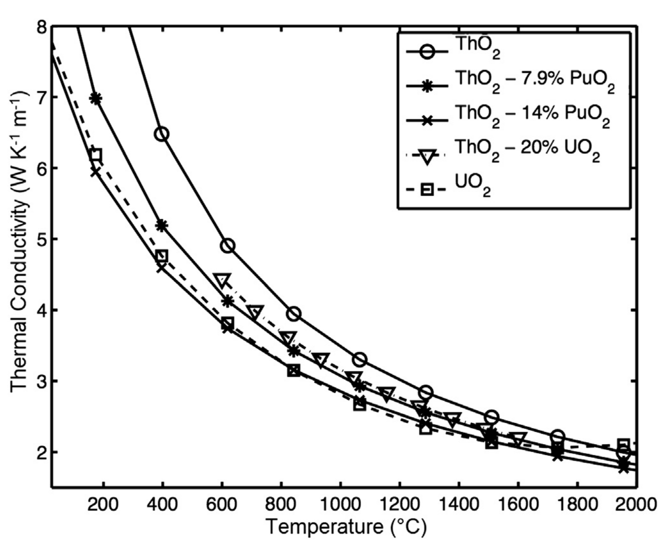

Fig. 1. The thermal conductivity of (Pu, Th)O_2 and (Th, U)O_2. Correlations for (Pu, Th)O_2 are taken from Cozzo et al. (2011) and correlations for (Th, U)O_2 and pure UO_2 are taken from Kim et al. (2006).

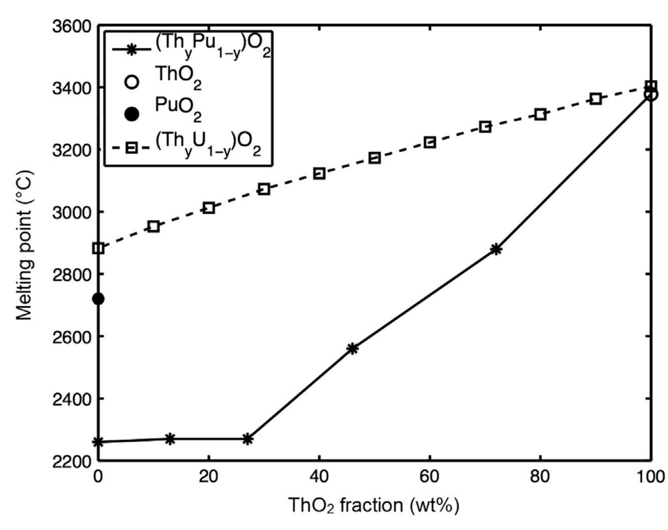

Fig. 2. The melting point of (Pu, Th)O_2 and (Th, U)O_2. Data points for (Pu, Th)O_2 are taken from Katayama et al. (1963) except for the data point for pure ThO_2 which is from Kim et al. (2006) and PuO_2 which is from De Bruycker et al. (2011). Data points for (Th, U)O_2 are liquidus temperatures taken from Belle et al. (1984).