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The feasibility research of thorium breeding using fluoride salt as a fast reactor coolant

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ABSTRACT

Breeder reactors are considered as a unique tool for fully exploiting natural resources. Fast breeder reactors based on thorium fuel can enhance inherent safety. Fluoride salt has good performance as a coolant in high-temperature nuclear systems. However, there is some doubt about the fuel breeding ability using fluoride salt coolant for fast spectrum due to its moderating ability. The aim of this study was to choose a proper fluoride salt mixture for Liquid-salt-cooled Solid-fuel Fast Reactor (LSFR) based on thorium-uranium fuel and give parametric studies to provide a design window for flexible self-sustaining core design. Infinite assembly model was used to analyze the salt selection from five candidate fluorides for fast spectrum as coolant. Combining neutron balance analysis with linear least squares fitting method based on 0D model, parametric studies at the neutron balance equation unique solution with burn-up for several parameters such as fuel volume fraction, removing fission gases process, total neutron losses and power density were presented in this paper. It was found that BeF_2-NaF was a promising coolant in the five candidate fluoride salt mixture. This study proved that the design of a self-sustaining core for fluoride-salt-cooled fast breeder based on thorium fuel is achievable. A design window was found in the definition of a self-sustaining core for various fuel volume fractions and neutron loss fractions. Core design and fuel management strategy will be given in future.

1. Introduction

In 2002, GFR, LFR, SFR, MSR, SCWR and VHER were selected as Generation IV systems, all of which have the advantage of sustainability, economics, safety, reliability and proliferation-resistance (US DOE Nuclear Energy Research Advisory Committee and the Generation IV International Forum, 2002). In particular, four are fast neutron reactors. Energy sustainably and long-term availability of nuclear fuel are important researching content in Generation IV systems (Locatelli et al., 2013). The fast spectrum system has the ability to breed fuel, allowing nuclear fission reactors to provide a very long-term energy supply. For complete utilization of the fertile isotopes, it is necessary to develop breeder reactors which can transmute the fertile isotopes into the fissile isotopes at a rate faster than the rate of consumption of fissile material for power production. With these unique features, the utilization of uranium resource increases significantly compared to LWRs (Fast spectrum reactors, 2012).

Thorium is 3–4 times more abundant than uranium, widely distributed in nature as an easily exploitable resource in many countries. Thorium fuel cycle is an attractive way to generate long term nuclear energy with low TRU waste (Lung and Gremm, 1998). Application of thorium-uranium fuels have been widely researched in thermal reactors such as PWRs, HWRs, SCWRs, HTGTs and MSRs (Lung and Gremm, 1998; Anantharaman et al., 2008; Jeong et al., 2008). Research has shown that only the high discharge burn-up of thorium-uranium fuel had economy advantage for commercial applications in conventional PWRs and HWRs, but the existing materials couldn't bear the radiation damage of the high accumulated burn-up (Long, 2002; Wang, 2003; Yildiz et al., 2011). SCWRs, HTGRs, and MSRs belong to generation IV reactors and are in a state of research and development. MSRs is the best candidate reactor for thorium-uranium breeding, but research has shown that a self-breeder MSRs (Nagy et al., 2008) requires a fast removal FP and MA online and the extraction of Pa and the re-introduction of the

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formed U. Limited studies have been carried out concerning the use of thorium in Fast Reactors (FRs), historically conceived as breeder reactors, due to the superiority of the uranium/plutonium cycle from this standpoint. The thorium fuel cycle is known to offer a better neutron economy than the uranium fuel cycle at epithermal energies and does not have a large positive reactivity feedback upon spectrum hardening at high neutron energies because (1) η(^{233}U) does not vary as steeply with neutron energy as η(^{239}Pu) does, (2) the fission cross-section of ^{232}Th has a higher threshold energy and smaller magnitude than that of ^{238}U (Fast spectrum reactors, 2012; Lung and Gremm, 1998). In addition, the relatively low mass number of ^{232}Th leads to a characteristically low TRU inventory when a^{232}Th closed cycle is established, with potentially beneficial impacts on the actinide radio-toxicity and decay heat. It is necessary to develop FRs for enhanced inherent safety with thorium-based fuel. Moreover, breeding ^{232}Th leads to a diversification of energy resource bases and allows greater flexibility in the choice of breeder reactor concept and fuel cycle (Fiorina et al., 2013).

Breeder reactors are regarded as a unique tool for fully exploiting natural nuclear resources. Coolant selection for FRs plays an important role in breeding performance and safety considerations. The coolant initially selected for most of existing fast breeder reactors was sodium, which is still considered the best candidate for these reactors (Lafuente and Piera, 2010). However, Na reacts chemically with air and water very easily and requires a sealed coolant system which leads to frequent accidents. Furthermore, Na-cooled FBRs have a positive void reactivity coefficient and this coefficient increases with the reactor size (Ichimiya et al., 2007). Another choice for FBRs coolant is Helium. Because the GFR requires a dense fuel element that can withstand very high temperature transients for lacking of thermal inertia of the system, its fuel element is still under design. Moreover, safety is a major concern for GFR (Anzieu et al., 2009), such as decay heat removal (DHR) without external power in depressurized conditions. As for lead- and lead-bismuth-cooled cores (Generation IV International Forum, 2014), several drawbacks must be overcome, including the need for coolant chemical (oxygen) control for prevention of lead erosion-corrosion effects on structure steels at high temperatures and flow rates, and seismic/structural issues due to the weight of the coolant. Therefore, it seems reasonable to explore new options for FBRs coolant.

In recent years, there has been a rapid growth in research and development on high-temperature molten salts in nuclear systems (LeBlanc, 2010; Serp et al., 2014). Fluoride salt as a coolant has been applied to pebble-bed high temperature reactor successfully. Moreover FHR (Fluoride-salt-cooled High-temperature Reactors) is considered as a better safety Generation IV reactor system. Fluoride salts have many attractive features, such as low pressure, high boiling temperature, optical transparency, high volumetric heat capacities and low chemical reactivity. A new fluoride salt coolant with excellent heat transfer performance perhaps can avoid some safety issues for existing FRs. However there is some doubt about the fuel breeding performance using fluoride salt coolant for fast spectrum due to its moderating ability which result in the chlorine salt as primary coolant for early molten salt fast reactor research. Fortunately, Th-U fuelled FRs still possess the breeding capability for a soft spectrum and this provides the possibility of designing fluoride-salt-cooled fast breeder reactor.

Thorium breeder reactors using fluoride salt as a coolant based on solid fuel already has done some research. The concept of liquid-salt-cooled solid-fuel fast reactor (LSFR) was put forward in 2005 by Forsberg (Forsberg et al., 2007), who has promoted the AHTR concept development in 2003 (Forsberg et al., 2003). Unlike the MSR, an LSFR uses solid fuel and a clean liquid salt as a coolant. This cause two major implications (Forsberg et al., 2005): (1) Salt selection. Existing fluoride salt coolants are based on thermal spectrum and they may not be suitable for fast spectrum. This study will choose a proper fluoride salt mixture for LSFR based on thorium-uranium fuel. (2) Material selection. It is necessary to evaluate the structure materials with the requirements of low neutron absorption rate, high-temperature resistance, irradiation resistance and compatible with liquid salt. Taking fuel elements for existing high-temperature nuclear system as a reference, tristructural-isotropic (TRISO) fuel and the fuel element of helium-cooled fast reactor can be adopted for LSFR. In 2010, Lafuente and Piera gave a design window meeting the requirements of criticality, breeding and safety (a negative coolant void reactivity) with BeF_2 as a coolant using a simple cell model based on thorium fuel (Lafuente and Piera, 2010). But (1) this design window required the high fuel ratio of TRISO pebble which exceeded greatly the engineering manufacturing tolerance; (2) BeF_2 is not suitable as a coolant due to its poor thermophysical properties such as too large dynamic viscosity, high melting point; (3) there is no consideration about leakage rate and burn-up analysis in their calculation. Guifeng Zhu (Zhu, 2015; Zhu et al., 2016) gave similar results for breeding ability research based on LiF-BeF_2 coolant and TRISO pebble. These study indicate that PB-FHR has poor thorium fuel breeding ability because graphite matrix in coated particle fuel has strong moderating capability which results to high parasitic neutron absorption rate of coolant and low effective fission neutron number produced per absorption. In order to improve this situation, Guifeng Zhu adopts new fuel pellets of coated fuel particles dispersed in SiC matrix. Pilot study shows that this new PB-FHR has a better breeder capacity, the maximum discharge burn-up is more than 20% FIMA with self-sustainable thorium-uranium fuel cycle and negative coolant void reactivity coefficient. However there are two major challenge for the fuel element design based on the new SiC matrix: (1) existing fuel dispersed particles based on SiC matrix is tubular not spherical; (2) dispersed particles based on SiC matrix has a greater inconvenience in the post-processing. With the incentive to minimize the specific fissile in the breeder reactor, a “tight” fuel element that maximizes fuel volume fraction is desirable (Fast spectrum reactors, 2012). Moreover the higher fuel volume fraction minimizes fissile loading mainly by reducing reactor leakage. It's obvious that TRISO fuel is not suitable for LSFR. Taking GFR as a reference (Perkó et al., 2015), the design of LSFR in this study use carbide ceramic fuel and numerous favorable properties SiCf/SiC as cladding material and liner material with a cylindrical arrangement of hexagonal fuel assemblies. Actually the fuel element of GFR has good high temperature nuclear radiation performance and SiCf/SiC is compatible with fluoride salt (He et al., 2014).

In this study, the research content mainly includes: (1) choose a proper fluoride salt mixture for LSFR based on thorium fuel; (2) investigate the effect on the thorium breeding capacity and the average batch discharge burn-up that an equilibrium cycle of a fast reactor can be operated at of several parameters such as fuel volume fractions, removing fission gases, the effect of neutron loss and power density. In the fast neutron spectrum, because ^{232}Th is less fissile than ^{238}U and has a higher fission threshold energy, this study require design of thorium-based fuel self-sustaining core with BR slightly >1.0 for multiple recycling of recovered thorium and uranium without actinides. This paper is divided into five sections. Section 1 is the introduction. Section 2 presents the methodology, first introduces the calculation tools and then describes the calculation procedure of fluoride salt mixtures selection with infinite assembly model, finally focusing on the calculation procedure of parametric studies for long term with burn-up analysis based on a simplified 0D cores. Section 3 presents the results, firstly, investigated the BR, fluoride salts nuclide absorption share