

INSIGHTS ON THE EVOLUTION OF THORIUM FUEL

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Abstract- Eight decades of literature on thorium fuels ranging from 1940s to the present are categorized and analyzed. Various literatures are divided into 12 categories to simplify them. The US comes first in publishing a huge number of publications and literature for an approximate of 950 publications made so far. India holds the second place in number of publications made including research and studies. Most of the research and publications has been done on waste management and reprocessing techniques, which is necessary for the application of thorium fuel in reactors. A few numbers of publications were made on physics and nuclear data. The study of thorium fuel has started in early 1940s. In the overall review, the study of thorium fuel cycles has been at peak by 1970s and literally stopped by 1980s. Up until 1980s, uranium-thorium mixed fuel study has provided enough data to operate this mixed fuel powered nuclear reactors. India is the only country that didn't stop the use of thorium mixed fueled nuclear reactors in between the decline period of thorium studies - 1980s to 2000s. After 2000s the number of papers published has gone higher than 1980s which is to be considered. Now the interests have revived in the 21st century, the research of thorium fuel has made its way go higher than ever. This paper provides an overlook on those publications which made effective progress in each category. Thus, stating that thorium fuel may be the next Generation Nuclear Fuel.

Keywords-waste management, reprocessing techniques, reactor, thorium fuel.

I. Introduction

Thorium is considered to be an effective alternative for uranium. After a lot of fabrication, irradiation tests and multiples of fuel cycle facilities including hundreds of literatures states the effective significance of thorium. Even then, yet, thorium is not a commercial success as its uranium based counterparts. Since various research and studies are yet to be done on thorium, we have some valid points on thorium. Thorium fuel can enhance inherent safety. It is 3-4 times abundant than uranium (Yu Peng, Guifeng Zhu, et al., 2017). Th²³² has higher threshold energy (lung & Gremm, 1998). Thorium has an incredible life time. The half-life of thorium is 1.9 years (Belle & Berman, 1984).

Vanderbilt university and Oak Ridge National lab has provided support for the revival of discussion on thorium under various aspects since 2012. From the database available at them, they have published a review on the insights from the eight decades of thorium fuel cycle research and literature. (Timothy Ault, Steven Krahn, 2017). India has 24% of thorium of the earth. Adding the present data to it, this paper will provide a complete review on thorium research yet in a categorized way.

II. Quantitative Literature Assessment

A. Resources and recovery

During the 1950s, publications regarding the study of thorium as a chemical and electrolyte mineral were

published. In 1960s the recovery of thorium from these minerals were improvised with further studies. As of 2010, Thorium can be obtained as a by-product of existing products thus avoiding the need of opening new mines.

Cuthbert (1958) presented the summary of thorium along with its chemical and physical properties, separations, recovery, ores and safety methods of processing [21]. **Crouse and brown (1959)** presents extraction of thorium from monazite with reagents from early to 1960s [20]. **Tennery(1978)** presented many thorium deposits across US and also an ore extraction process [74]. **Keni(1990)** consolidates thorium recovery in India up to 1990 and provides various alternative production methods [46]. **Gupta and Krishnamurthy (2005)** presented accounts of virtually all commercially viable thorium bearing ores including bastnasite and monazite [33]. **Ault (2015)** shows a shift change in thorium recovery paradigm from direct to by-product recovery systems and also provided major thorium deposits by region based on capacity[5].

B. Fuels

The main research was in fuels from the beginning as the tests, fabrication, irradiation and all necessary steps prior to the largescale production were based on it. As of course the early publications were all based on fuels. The fuel research has a sharp decline at 1970s. Now after 1990s, the emphasis increased on mixed fuels like

thorium-uranium and thorium-plutonium. All these fuels emerged as oxide fuels. Early research had the study of irradiated fuel in a limited irradiation experiments. Even now the presence of uranium plays an important role since the study of uranium is well settled. Since thorium has an expanded life time, a deep study of thorium fuel and its containment has to be studied yet. Primary tests and demonstration of thorium has been studied from late 1950s.

Batch and Snidow(1960) states the irradiation efforts at the consolidated thorium reactor along with various physical properties which hasn't been previously verified[7]. **Olshen(1979)** consolidates a variety of fuel fabrication development for a variety of fuel types including oxides, carbide, metallic and even on mixed fuels [61]. **Ringel and Zimmer (1979)** present techniques of radiation protection requirements for reprocessing fuel [68]. **Belle & Berman(1984)** varieties a remarkable difference in the physical properties of thorium dioxide which also includes high-level irradiation summaries [10]. **Clayton (1987)** describes corrosion experiments in metallic and mixed fuels [17]. **Balakrishna(2012)** reviews the sintering, extrusion and gel fuel fabrication of thorium based fuel types of India[6]. **Bjork (2015)** gives one of the most recent thorium fuel fabrication and irradiation testing program which happened in Norway which leaves to be the only one in 21st century [11]. **Gyorgy and Czifrus(2016)** presents thorium as an effective fuel in nuclear core other than uranium and plutonium with various data charts and analysis. **Michel Lung and Otto Gremm(1998)** provides the perspectives of thorium fuel along guidelines that thorium is future nuclear fuel [53].

C. Physics

The physics data available for thorium is no way sufficient when compared to uranium or plutonium. In 1960s, BARC, B&W conducted some independent resonance integral and cross section experiments. The number of such experiments raised in 1970s and now in 2000s efforts are being sought to bid a gap between uranium and thorium data.

Gore (1978) bids the gap between critically data and physical parameters from various experiments and also identifying other data gaps [31]. **Abbondanno (2001)** took a multinational level effort to improve data quality for thorium related isotopes [1].

D. Light water reactors

Only a few publications were presented on thorium based LWRs. Only in 1970s experience with LWR for thorium based fuel is found. In 1980s the LWRs started to fade off as other categories showed up. However, there are only 10 publications between them. After

1980s, the numbers increased more than a hundred. Since the lack of universities and government based laboratories, very few publications were made.

Farrel (1960) presents the consolidated Edison Thorium Reactor summary which is probably the first publication of thorium based LWR [26]. **Shapiro (1977)** states the assessment of alternative thorium fuel cycle option along with recycle in conventional PWRs [72]. **Atherton (1987)** summarizes the characteristics of LWBR along with the fuel development, operational procedures, necessary modifications in the plant [4]. **Kazimi (1999)** presents an overall review till date on LWRs based on thorium from the perspective of neutronics, thermal hydraulics, safety and waste management [45]. **Todosow (2005)** represents the missed uranium-thorium fuel lattices in PWRs without reprocessing which thrusts the thorium research in 21st century. **Lindley (2014)** summarizes both the PWRs and BWRs which use thorium to achieve transuranic recycle, which complements reprocessing [50]. **Liu and Cai (2014)** reviews the prospects of using thorium in supercritical water reactors [51]. **Hong yeop Choi, Chang Je Park (2017)** provides a concept of epithermal spectrum nuclear core in LWR for thorium fuel along with data showing a change in lattice structure of fuel rods.

Table 1 Thorium fuel -description by category

Category title	Description/embedded topics
Resources & Recovery	Thorium resources from deposits or from chemical separation processes from natural occurring minerals.
Fuels	Fabrication, re-fabrication, chemical and physical properties of thorium fuel in both fresh and irradiated forms and also in post-irradiation forms.
Physics & Nuclear Data	Descriptions of data, experiments to measure nuclear data such as cross sections and resonance integrals.
Light Water Reactors	Studies of pressurized water reactors, boiling water reactors, and supercritical water-cooled reactors utilizing light water as coolant with reports
Heavy Water Reactors	Studies of pressurized heavy water reactors as well as other reactor system

	concepts that use heavy water as a moderator with reports.
Liquid-Metal-Cooled Reactors	Studies of systems with liquid metal coolants, such as sodium-cooled fast reactors (SFRs) and lead-bismuth-cooled reactors
Gas-Cooled Reactors	Studies of both “block type” and pebble bed gas reactors, including both thermal- and fast-spectrum options which uses heavy gases as coolant
Molten Salt Reactors	Studies of liquid-fueled salt-cooled reactors (both fluoride- and chloride-based) and solid-fueled salt-cooled reactors
Externally-Driven systems	Studies of both fusion-fission hybrids and accelerator-driven systems which use external systems to start functioning.
Reprocessing & Waste Management	Reports comprising the “back-end” of the fuel cycle, such as fuel dissolution, aqueous reprocessing methods, non- aqueous reprocessing methods, waste characteristics, and waste disposal approaches.
Safeguards	Non-proliferation and safeguards aspects of managing and measuring U-233 and all safety measures.
Multi-Topic/Other	Reports that span combinations of other topical categories, or address different topics altogether.

E. Heavy Water Reactors

From the 1950s the HWRs started out with some collaboration among United Kingdom and Babcock and Wilcox. By 1960s, the thorium based PHWRs emerged slowly. In 1970s they started in Canada, India and then followed by other countries. Studies went down totally except India and Canada in-between 1970s and 1980s.

Cason and Landrum(1955) present the most comprehensive review of the aqueous Homogeneous Reactor, which turned to be a major project in 1950s [16]. **Redman (1961)** summarizes results from a series of experiments conducted on Zero Power Reactor facility [67]. **Dormuth and Lidstone(1977)** contains the study of pressurized heavy water reactors including a significant emphasis on the insights at the fuel cycle level. **Jagannathan(2001)** provides insights from the PHWR technology in India as part of the three stage Nuclear powerplant [41]. **Bozcor(2002)** reviews a decade of research on thorium in CANDU reactors along with a huge perspective on a variety of fuel recycle strategies and fuel cycle missions. **Sahin(2008)** provides 20th century papers on thorium in HWRs which use plutonium and minor actinide inventories [71]. **Bromley (2014)** strategies in improved use of thorium in PHWRs along with safety, fuel ratio and geometry [13].

F. Liquid Metal Cooled Reactors

This section is the less studied when compared to other reactor technologies in thorium based LMRs. One study from before 1977 has been found from India which provides the fundamental physics aspects of the use of thorium in sodium cooled systems. Other than that, only 3 publications have been made till 2006. After 2007 the remaining papers have been published. Even though interests have been improved, unless India starts to study further, no proceedings seem to happen.

INFCE WG5(1978) reviewed a lot of papers based on thorium based fueling options in LMRs and also comparing their performance in breeding, safety and reprocessing. **Atefi(1979)** did evaluation of thorium blanket fuels in LMR from a most extensive pre-1980s study in thorium on LMR. Okawa and Sekimoto, 2011 summarizes few attempts to link thorium fuel to a lead cooled system while checking for fast spectrum thorium fuel cycle options [3]. **Fiorina (2013)** states the important findings which provide a comparison of thorium’s and uranium’s performance in SFR [27].

G. Gas cooled Reactors

GCRs have the most studies on thorium fuels. The first was G1 test reactor from France in 1950s which was then followed by United Kingdom in 1960s. The subject of thorium in GCRs increased widely in 1970s. The interests were high in this time and due to parallel research and studies, there was a significant development in this period. The thorium High temperature reactor was operated in 1983 to 1989. Even after these, the post irradiation has been a little trouble. Sharply after 1970s there is a huge decline in the study of GCRs. Only 4 publications have been found

in between 1983 to 2001. Netherlands, US, Sweden and Canada have a significant portion in recent studies.

De Rouville(1958) documented the earlier use of thorium in reactor in France along with the experiences of the plant operations, reliability and the use of thorium-uranium mixed fuel. **Lotts and Coobs, (1976)** represents one of the high temperature gas reactor reports in 1970s along with the implications of thorium fuel and also the bed type systems. General Atomic, 1980 gives information on licensing, setup and early operations of the thorium-uranium GCR. **Bultman 1995** states the prospects of thorium use in GCR's from a high-level perspective along with examples of use of thorium fuels in GCRs. **Wols** presents the most generalized of the numerous 21st century studies on modular bed GCRs using thorium fuel. **Amr Ibrahim (2017)** published the analysis of thorium fuel in GCR at a large scale by using MCNPX code along with safety measurements to decay the waste.

H. Modern Salt Reactors

The MSR holds majority in the number of publications published since the modern salt reactor experiment and projects in that period. After 1970s, by the end of ORNL, MSR publications have a huge downfall in numbers. Now with LPSE, MSR has come back in 21st century.

Haubenreich and Engel(1970) summarized development and operational experience with salt water reactor along with corresponding conclusions [35]. **Robertson (1971)** consists of comprehensive reviews of a single fluid design variant of salt water reactor with considerations on hydraulic, thermal and physics [69]. **Engel (1980)** provides info on MSBR with more safety characteristics including once-through refueling and denatured u^{232} [25]. **Kamei (2010)** provides a collection of papers across several decades of research on thorium based MSRs in this instance considering the potential rate of deployment [43]. **Merle-Lucotte(2011)** summarizes a set of papers on fast spectrum thorium based MSR fuel cycle projects which anticipate future challenges with deployment of thorium MSR fleet [57]. **Holcomb (2013)** consists of extensive writeups on salt cooled MSR systems, and considers various designs and fuel cycle options [36]. **Yu Peng (2017)** shows that fluoride salt serves to be the fast reactant coolant which also provides data for providing techniques which help waste management of this method.

I. Externally Driven Systems

The earliest of EDS were published from 1970s. more papers were published in the recent times when compared to other category. Research on accelerated

systems are not available. Only a few studies in the late 1990s are available.

Maniscalco(1978) states the series of LLNL publications on the prospects of breeding u^{232} in a fusion-fission hybrid along with design allotments. **Takahashi (1983)** represents the only assessment of u^{232} breeding by thorium in conjunction with accelerator driven system [73]. **Rubbia (1995)** shows the concept of energy amplification and considers thorium as a plutonium disposition strategy [70]. **Ubeyli, (2006)** constitutes the most generalized of fission- fusion hybrid to emerge from a consortium. **Fratoni(2012)** presents a review on thorium based fission-fusion hybrid including potential and design strategies. **Coates and Parks(2012)** assessed the thorium based accelerator driven systems including discussion of safety and fissile material management[18]. **Nefinecker(2001)** summarizes basics of accelerated subcritical reactors.

J. Reprocessing and Waste

Reprocessing of thorium fuels were found to be apublished from 1950s. Most publications on this topic have been published in the 1970s. Aqueous reprocessing of thorium-uranium fuel and GCR fuel were emerged during this time period. India took part in providing reprocessing techniques of aqueous fuel in between 1970s and 1980s. Only India and BARC initiated this reprocessing of fuel in the earlier times. In general, reprocessing of fuels is tougher. Even much documentation have been made on the dissolution rates, availability of reactants, process efficiencies and shielding requirements which show that reprocessing has its difficulty. On the other hand, reprocessing of fuels seems to be an effective way than waste management. Reprocessing has given significant amount of thorium and uranium than waste management processes.

Rainey and Moore (1962) described the development and refinement of THOREX process along with the achievements in aqueous thorium reprocessing including flowcharts and descriptions of waste streams [66]. **Baxter (1976)** published one of the most comprehensive discussions of atomics process to recycle GCR thorium based fuel and about some challenging stuff [9]. **Orth(1979)** state the purity of thorium and uranium being reprocessed at Savannah river bank which is one of the largest recover of uranium. This also includes the data regarding the quantities of uranium reprocessed [62]. **Pickett (1982)** relates the dissolution of thorium and thorium-uranium dioxide along with its catalog results [63]. **Gruppellar and Schapira(2000)** summarized a variety of information on waste management of thorium fuel cycles along with the management of other inventories

[32]. **Dey and Bansal(2006)** states the experiences in reprocessing the thorium fuels along with various technologies used for reprocessing them [23]. **Croff and Krahn(2016)** studied the radiotoxicity if both thorium and uranium based fuels stating their effects over time [48]. **Wymer(2014)** provides a conclusion on the underlying physical characters which lead to the difficulty of reprocessing thorium when compared with uranium fuel [79]. **Timothy Ault (2017)** discussed the uranium and thorium fuel recycling processes in various types along with data charts [56]. **Roger Barlow (2016)** summarizes the use of ADSR system as a better option of waste management of thorium fuel in between many other options. **Felipe Sotelo (2017)** published a paper on solubility of uranium and thorium with cement by processing it in fuel waste management system.

K. Safeguards

The earliest publications on safeguards have been dated by 1977 which was a lead to 11 other publications within 1979. As starting, the publications included denaturing of uranium, material accountability, nondestructive assay and some analytical techniques. All publications on safeguarding show that uranium will have similar safeguarding to plutonium.

Hakkila (1978) describes a variety of chemical techniques regarding the safeguarding of thorium based fuels [34]. **Forsberg (1998)** provides a calculation to support the definition of stable uranium [28]. **Bathke(2009)** provides a discussion to figure calculations along with multiple fissile materials which was a uranium and plutonium mixed fuel [8]. **Worral(2016)** described the challenges in implementation of thorium fuel cycle till date and also discussing on various techniques for safeguarding [77].

L. Other Studies

Some papers can't be categorized under the above topics. Therefore, they are set to be in general, described below.

Wymer(1968) documented the entire second international Thorium Fuel Cycle symposium and presents the state-of-the-art for many fuel and reactor types [78]. **BNL (1969)** presents the thorium resource options, aspects of thorium fuel cycle, prospects of use of thorium in every reactor type technology along with the advancement in fuels and reprocessing. **MacDonald and Nair(1979)** studies on the radiological aspects of thorium fuel cycle on handling, reprocessing, waste disposal along with figures and data for safe operations [54]. **IAEA (1987)** consolidates the proceedings of thorium fuels and their projects from the early to the decline in 1970s [37]. **Lung(1997)** summarizes the European prospective on

thorium fuels along with their reactors, fuels, resources, reprocessing and waste management [53]. **Andreev(2013)** states the conspiracy of uncertainties on economic assessment of thorium fuel cycle [2]. **NEA(2015)** presents the entire thorium fuel cycle with modern perspectives, integrating the knowledge over a dozen of international publications in energy level portfolios [59]. **Martin and Girieud (2016)** provides midterm theories on effective application of thorium fuel in PWR which also circumscribes the reprocessing of thorium. **Peegs(2012)** published the necessary and surety of thorium as the future nuclear fuel by analyzing various types like LWR, HWR, FWR, ADS and others. **Vijayan (2017)** presents the role of thorium in Indian nuclear powerplants since its wide availability in India.

III. Conclusion

This literature has documented the publications on thorium fuel cycle, spanning across the globe. Several hundred organizations and governments have taken part in the research and studies on thorium although in which certain like ORNL, IAEA has done particularly great works in the peak times and succeeded with their work. Well, nowadays universities and journals have nominated the study on thorium fuel cycles due to the availability of online literature and journals. This has increased the interest in this study that shows the reason behind the number of publications which are published lately in 2017 as higher than 1970s.

There are lot of uncertainties in the study on thorium based fuels, reprocessing, resources, waste management, containment of radiation, constructions of beds, LWR, GCR, HWR and so on. Yet the research is meant to provide the favorability of understanding whether or not to pursue this thorium fuels. Though the researchers don't expect the results to be favorable, this study is considered to be necessary. As the present nuclear reactors are said to be Gen-2 type and those available are Gen-3. Thorium based nuclear reactors are considered to be Gen-4 reactors which is mainly due to the life time of thorium, (Belle & Berman, 1984).

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