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Investigations on enhanced nuclear fuel utilization in light water reactors by mixing of uranium and thorium based heavy metals

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Diplomarbeit

Hiermit erkläre ich, dass ich die vorliegende Diplomarbeit selbstständig und ohne unzulässige fremde Hilfsmittel angefertigt habe. Die verwendeten Literaturquellen sind im Literaturverzeichnis vollständig angegeben.

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Zusammenfassung

Der erste Teil dieser Arbeit, die im „Institut für Neutronenphysik und Reaktortechnik“ des Forschungszentrums Karlsruhe gemacht wurde, betrifft Untersuchungen von Kernbrennstoffen. Die Aufgabe war es die Machbarkeit eines auf Thorium basierten Brennstoffkreislaufs mit vereinfachten reaktorphysikalischen Berechnungen zu untersuchen. Thorium stellt hierbei eine mögliche Alternative zum klassischen Uran Brennstoff dar. Die Hauptpunkte, die untersucht worden sind, sind mögliche Einsparungen von Uran, die Radiotoxizität der erzeugten Abfällen, und die Sicherheitseigenschaften des ganzen Brennstoffkreislaufs. Der zweite Teil der Arbeit wurde teilweise im „Rensselear Polytechnic Institute“ (USA) durchgeführt. Das Thema war ein neues Modell für die Neutronenstreuung im Resonanzenergiebereich, das in dem FZK entwickelt wurde, experimentell zu bestätigen. Dafür wurden Messungen der Neutronenstreuung an Thorium durchgeführt. Der Einfluss dieses neuen Modells auf die Ergebnisse von Brennstoffkreislaufuntersuchungen wurde betrachtet.

Die weltweit verfügbaren Thorium-Ressourcen werden auf das Dreifache der Uran-Ressourcen geschätzt. Thorium kann, sowie Uran, als Brennstoff für Kernkraftwerke benutzt werden. Seit dem Anfang der nuklearen Energieerzeugung wurde Thorium untersucht, aber noch nicht zu einem kommerziellen Brennstoff entwickelt. In dieser Studie soll der Einsatz von Thorium in heutigen und zukünftigen Druckwasserreaktoren betrachtet werden. Die Nutzung von Thorium ist aber in diesen Reaktoren technisch komplizierter als Uran. Thorium enthält keine spaltbaren Isotope wie natürliches Uran, das 0,7% spaltbares U-235 enthält. Thorium muss deswegen mit Uran oder Plutonium gemischt werden, um als Brennstoff in einem Reaktor eingesetzt werden zu können. Das Ziel dieser Arbeit ist es zu bestimmen inwiefern es möglich ist, Thorium als Brennstoff in Druckwasserreaktoren (DWR) zu benutzen.

Die Untersuchungen wurden mit dem Code KAPROS durchgeführt. Dieser neutronenphysikalische Code existiert seit 40 Jahren und wurde im Kernforschungszentrum Karlsruhe kontinuierlich weiterentwickelt. KAPROS ist gut geeignet für Brennstoffkreislauf Untersuchungen, und insbesondere für innovative Reaktorkonzepten. In dieser Arbeit wurde der DWR Kern als einfache 2D-Zelle mit KAPROS dargestellt. Da in dieser Studie im Wesentlichen nur Vergleichsrechnungen von verschiedenen thoriumhaltigen Brennstoffen durchgeführt werden

erlaubt diese vereinfachende Modellierung kurze Rechenzeiten. Ein Vergleich zwischen KAPROS und Monte-Carlo Rechnungen für Thorium Brennstoffe wurde durchgeführt und hat befriedigende Ergebnisse geliefert.

Es wurde zuerst festgestellt, dass es nicht günstig ist, Thorium in einem klassischen „Once-Through cycle“ zu benutzen. Bei diesem Zyklus werden die Abfälle nicht recycelt. Ein Vorteil von Thorium ist, dass es im Reaktor U-233 produziert. Dies ist ein sehr gutes Spaltmaterial, im Vergleich zu Pu-239 oder U-238. Um dieses Potenzial des Thoriums zu benutzen, muss der Spaltstoff U-233 im sogenannten „Closed Cycle“ recycelt werden. Dies benötigt eine Wiederaufbereitung des abgebrannten Brennstoffes. Die zurückgewonnenen Spaltstoffe werden dann in einen frischen Brennstoff eingesetzt. Dies führt zu einer Einsparung von Uran.

Zwei Typen von thoriumhaltigen Brennstoffen wurden untersucht: Uran/Thorium und Plutonium/Thorium Mischungen. Uran/Thorium Brennstoffe wurden für industrielle Anwendungen betrachtet, während die Plutonium/Thorium Brennstoffe eine sehr effiziente Verbrennung von Plutonium darstellen.

Eine erste Optimierung des Thorium Brennstoffzykluses wurde durchgeführt. Hierzu wurde der Einfluss der folgenden Parametern studiert: Thorium Inhalt im Brennstoff, das Verhältnis Wasser Volumen zu Brennstoff Volumen und die Anreicherung von dem Uran.

Die Rechnungen haben gezeigt, dass je mehr Thorium der Brennstoff enthält, desto besser die Bilanz des Uranverbrauchs ist. Allerdings ist der Thorium Anteil im Brennstoff auf ca. 73% durch die Wiederaufbereitung begrenzt. Die Berechnung des Uran Verbrauchs hat gezeigt, dass ein geschlossener Thorium Brennstoffzyklus bis zu 23% weniger Uran verbrauchen kann als ein geschlossener Uran Brennstoffzyklus, und 60% weniger als ein klassischer, offener Uran Brennstoffzyklus. Es wurde ebenfalls gezeigt, dass Thorium bessere sicherheitsrelevante, neutronphysikalische Eigenschaften hat als Uran (Doppler-koeffizient, Moderator Dichte Koeffizient, Void Effekt). Ein wichtiger Aspekt der Kernbrennstoffen wurde auch untersucht: die Radiotoxizität der Abfälle. Die Ergebnisse zeigen, dass Thorium ca. 25% weniger minore Aktiniden und ca. 50% weniger Plutonium als Uran produziert. Dies ist eine deutliche Verbesserung. Durch die Produktion von U-232, hat Thorium eine gute Proliferationresistenz.

Die Verbrennung von Plutonium gemischt mit Thorium wurde auch untersucht. Plutonium wird in einem Reaktor durch Neutroneneinfänge von Schwermetallen produziert. Thorium produziert deutlich weniger Plutonium als Uran. Plutonium wird deshalb gemischt mit Thorium und stärker verbrannt als mit Uran. Plutonium selbst kann nicht pur gebrannt werden. Es

wurde gezeigt, dass die Verbrennung von militärischem Plutonium in einer Thorium Matrix doppelt so effizient ist wie in einer Uran Matrix. Dabei werden 30% weniger minore Aktinide erzeugt. Dieser Prozess produziert U-233, das als Brennstoff benutzt werden kann.

Der zweite Teil der Arbeit war hauptsächlich experimentell. Ein neues Modell für die Neutronenstreuung wurde von Dr. Dagan im FZK entwickelt. Dieses Modell beschreibt die sekundäre Energie eines Neutrons nach einer Streuung an einem Atomkern unter Berücksichtigung von Resonanz-wirkungsquerschnitten. Es liefert deutlich unterschiedliche Ergebnisse für Neutronenreaktionen im Resonanzenergiebereich im Vergleich zu dem standard Modell, welches in gängigen Codes benutzt wird. Das Standardmodell vernachlässigt Resonanzeffekte und teilweise Temperatureffekte. Dies kann Einfluss auf die Kritikalität und die Neutronenabsorptionsrate von schweren Metallen haben, und die Ergebnisse von Brennstoffkreislaufuntersuchungen verändern.

Das resonanzabhängige Modell wurde in der Vergangenheit teilweise durch Streuung an U-238 bestätigt. Im Rahmen dieser Arbeit wurde ein Experiment mit Th-232 durchgeführt. Dieses Experiment fand im „Gaertner LINAC Laboratory" in Troy, USA statt. Durch eine„time of flight“ Messung wurde die Energie von Neutronen gemessen, die unter einem bestimmten Winkel an einer Thorium Probe gestreut wurden. Das Experiment wurde mit dem MCNPX Code für zwei Fälle simuliert: mit dem standard und mit dem neuen Modell. Diese Rechnungen wurden dann mit den experimentellen Ergebnissen verglichen. Es wurde festgestellt, dass die experimentellen Daten viel besser mit dem neuen Modell als mit dem alten übereinstimmen. Für eine vollständige Bestätigung des neuen Modells müssen weitere Untersuchungen gemacht werden.

Mögliche Einflüsse von diesem neuen Modell auf die in dieser Arbeit durchgeführten Brennstoffkreislaufuntersuchungen wurden untersucht. Die Ergebnisse zeigen, dass der Einfluss der verbesserten Behandlung der Streuung an Thorium gering ist.

Die durchgeführte Arbeit hat gezeigt, dass Thorium prinzipiell als Brennstoff betrachtet werden kann. Der Einsatz von Thorium ermöglicht Einsparungen von Uran. Die Nutzung von Thorium muss aber technische Probleme lösen. Ein neues Modell der Neutronenstreuung wurde durch ein neues Experiment bestätigt.

Abstract

The current renewal of the nuclear industry worldwide, due to its cost effectiveness and its ability to mitigate the CO2 emissions, will probably lead to the construction of a considerable amount of light water reactors in the near future. The uranium resource will therefore be a very important topic for the world nuclear industry. Thorium is a potential fuel, and has been studied in the past, but was not yet implemented at commercial scale. Its utilisation could be an effective way to save the uranium resources before the implementation of IV-generation reactors.

The present work shows how thorium could be used as fuel in a Light Water Reactor. Investigations have been made on simple pin cell calculations with the reactor code KAPROS developed at the “Institute for Neutron Physics and Reactor Technology” of the “Karlsruhe Institute of Technology”.

The ability of different thorium-based potential fuels to save uranium in modern LWR without major modifications has been analysed. The investigations showed that a Th/enriched uranium fuel cycle with multi-recycling is feasible and could lead to savings of uranium, lower minor actinides production and improved safety parameters. A plutonium/thorium fuel could be more efficient for plutonium incineration than a plutonium/uranium fuel, leading as well to lower minor actinides production.

Participation to a neutron scattering experiment on thorium confirmed a theoretical improvement of the neutron scattering model used by reactor physics codes, which was developed in FZK. First estimates of the consequences of this improvement on the burnup simulations are presented.

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1.1 Introduction

The majority of commercial nuclear reactors in the world use uranium as fuel. In the context of a growing energy demand combined with an urgent need to mitigate the anthropic CO_2 emissions, large scale nuclear reactors utilisation in the next future is an interesting option for the main energy consuming countries (USA, China, Europe and India).

Uranium is a renewable resource, which based on the current consumption might be available only for an approximate time of a hundred years [1].

There are numerous plans to build new kinds of reactors, which would drastically lower our consumption of natural uranium. These are the so-called generation-IV reactors, like Fast Reactors, Molten Salt Reactors, and High Temperature Reactors. The implementation on a wide scale of these reactors could save our resources and help us to provide CO_2-free energy for a long time.

However, it is expected that these generation-IV reactors will not be available at commercial scale before 2050, for the most optimistic forecasts. In addition to this, the countries concerned by these new implementations have taken the decision to reduce drastically their CO2 emissions in the near future. Most of them will continue to produce electricity with the actual LWRs (Light Water Reactors), which represent 356 from the 436 existing reactors [2] or they will implement the latest III-generation reactors, like the EPR (European Pressurized Reactor), which is a LWR. These reactors have a lifetime of about 60 years. They will significantly impact the uranium resources consumption in the next hundred years.

Therefore, there is a need to control our use of the uranium resources. The incentive of this study is to discuss the possibility to use thorium as fuel in conventional light water reactors. The world thorium resource is about 3 times more abundant than the uranium resource. Mined thorium is composed at 100% of the Th-232 isotope. In a reactor, Th-232 undergoes neutron captures, which lead to the production of fissile U-233. This behaviour is comparable to U-238, which produces fissile Pu-239 in a reactor. Thorium has also a great potential for the reduction of the production of minor actinides, which are long-lived wastes produced in the conventional nuclear fuel cycles, and are responsible for the majority of the long-term radiotoxicity of the wastes.

This study performs an evaluation of possible fuel cycles using thorium mixed with uranium, relative to uranium consumption, reprocessing of the spent fuel and safety aspects. These fuel cycles will be simulated for conventional light water reactors.

1.2 Front end of a thorium cycle

1.2.1 Thorium Resources

Natural thorium contains only the Th-232 isotope. The resources of thorium are quite widely distributed in the world, and are supposed to be around 3 times larger than the uranium resources. But these resources have not been investigated so intensively like for uranium so far, due to the low demand. Indeed, thorium has only been used for research purposes by the nuclear industry. Consequently there is an uncertainty in the estimations. No industrial scale production process was developed. Thorium can be found in general in association with rare-earth elements and uranium. Until now, thorium has essentially been a by-product of the extraction of zirconium and titanium from monazite.

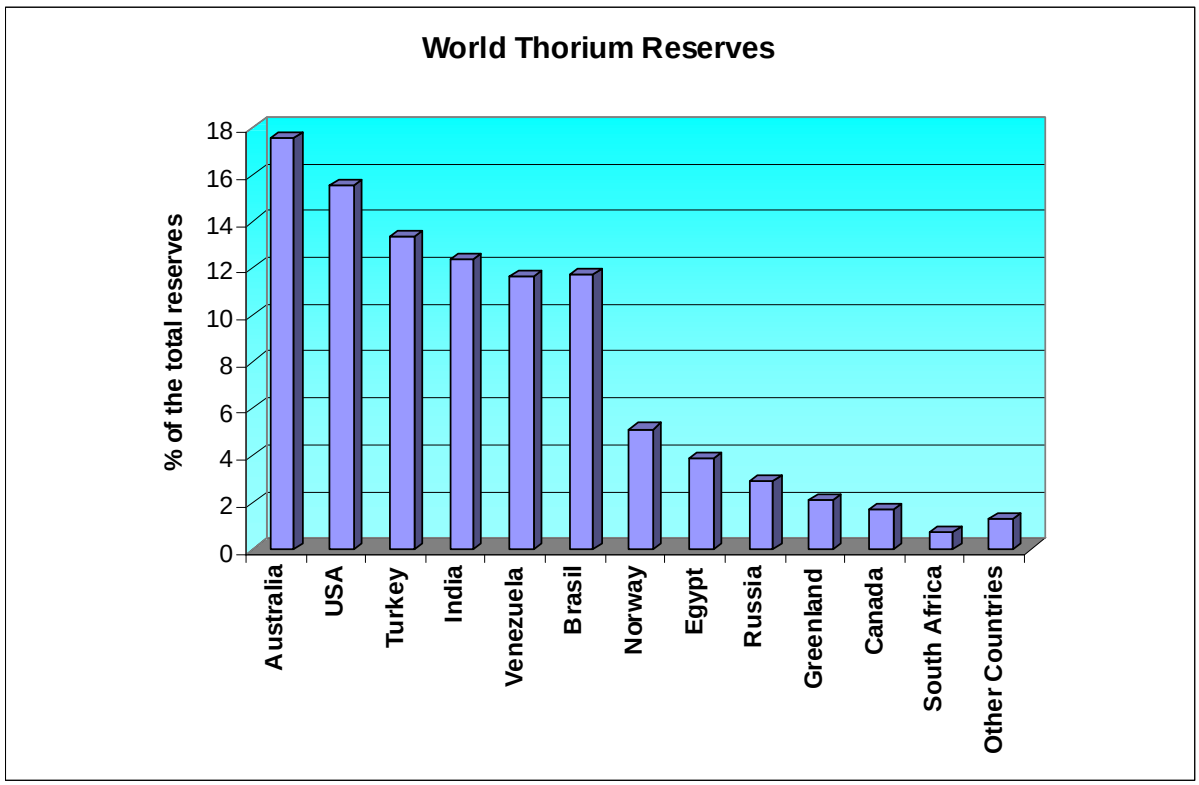

fig. 1-1 World Thorium reserves [3]

As can be seen in fig. 1-1, India has large thorium reserves, which represent around 13% of the world reserves. On the other hand, India does not have large uranium resources. India has therefore shown a big interest in developing a thorium fuel cycle. In case of success, the thorium fuel cycle could allow a self-sustainable nuclear industry in this country, based on local resources.

1.2.2 Mining and fuel fabrication

The mining of thorium and the fabrication of thorium based fuels is relatively easy and is well developed at industrial scale. After extraction and concentration of sands, the ore is converted into monazite, which is then converted to thorium concentrate. This thorium concentrate is the basis of nuclear grade ThO2. Thorium is easy to extract and the amount of radioactive wastes created by the extraction is about 2 orders of magnitude lower than in the case of uranium extraction [13].

Different thorium fuel elements have been tested in many research reactors. Their fabrication is based on reliable techniques. All these fuels are listed in the table below, taken from reference [13].

REACTOR TYPE	COMPOSITION	FUEL SHAPE	FUEL ELEMENT
High temperature Gas cooled reactors	ThO_2, $(Th,U)O_2$, ThC_2, $(Th,U)C_2$ (^{235}U or ^{233}U)	Microspheres 200-800 µ coated with multiple layers of buffer & pyrolytic carbon and SiC	Mixed with graphite and pressed into large spheres (~60 mm) for Pebble-Bed Reactor or fuel rods for HTGRs with prismatic fuel elements
Light water reactors Heavy water reactors	ThO2, (Th,U)O2, (Th,Pu)O2 (<5%Pu, 235U or 233U)	• High-density Sintered Pellets • High-density Microspheres	• Zircaloy clad Pin Cluster encapsulating Pellet-Stack • Zircaloy clad 'vi-pac' Pin Cluster encapsulating fuel microspheres
PHWR	ThO_2 for neutron flux flattening of initial core		
AHWR	$(Th,U)O_2$ $(Th,Pu)O_2$ (<5%Pu, ^{235}U or ^{233}U)	High-density Sintered Pellets	Zircaloy clad Pin Cluster encapsulating Pellet-Stack
Fast reactors	• ThO_2 blanket • $(Th,U)O_2$ & $(Th,Pu)O_2$ (~25%Pu, ^{235}U or ^{33}U) fuels • Th metal blanket • Th-U-Zr & Th-U-Pu-Zr fuels	High-density Sintered Pellets Injection-cast Fuel Rods	Stainless steel (SS) clad Pin Cluster encapsulating Pellet-Stack SS clad Pin Cluster encapsulating Fuel Rods
Molten salt breeder reactor	$Li^7F + BeF_2 + ThF_4 + UF_4$	Molten salt liquid form	Circulating molten salt acting as fuel and primary coolant

fig. 1-2 Types and geometry of thorium-based fuels and fuel elements

1.3 History of the Thorium Fuel

Thorium fuel has been considered as a fuel since the early years of the nuclear industry, but has never been used at commercial scale like the uranium fuel. This is mainly due to the fact that Thorium does not contain fissile isotopes and requires additional fissile material, unlike uranium. There was an interest for thorium fuel in the 60's for many countries like USA, Germany, India, the UK, France and Russia. A lot of work has been conducted at this time considering mining, fuel fabrication, and irradiation in test reactors. The thorium fuel has been studied mainly for Light Water Reactors. However, the possibility to implement thorium fuel was studied in important programs in High Temperature Gas Cooled Reactors too, for example in Germany and the USA.

1.3.1 Light Water Reactors (LWR)

One of the most significant examples of thorium fuel utilisation is the Light Water Breeding Reactor (LWBR) of Shippingport in the USA. The Shippingport power station began its commercial operation in 1957 as the first commercial nuclear reactor dedicated to electricity production. The LWBR program began in 1965 investigations for a thorium fuel to be tested in the Shippingport nuclear reactor. From 1977 to 1982, a slightly modified core produced electricity with a U-233/Thorium fuel. At the end of operation, investigations showed that the amount of fissile material was 1,39% higher after the cycle than at the beginning. It demonstrated that it is possible to achieve net breeding of fissile isotopes and to use thorium as fuel in a Light Water Reactor[4]. The fuel arrangement was a seed/blanket concept: each assembly contained a mix of $^{233}UO_2$ and ThO_2 with an enrichment of 5-6% in U-233 in the seed, and 1,5-3% in the blanket. The reflector was pure thorium at the beginning of the cycle.

1.3.2 High Temperature Gas cooled reactors (HTGR)

In Germany, a first experimental (15MWe) pebble-bed gas cooled reactor was tested in the Forschungszentrum Jülich during the time period 1967-1988: the AVR (Atom Versuchs Reaktor). The fuel was spherical fuel elements, containing coated particle in a matrix of graphite. The fuel elements were continuously unloaded, recycled and reloaded until their final burnup, so that there was no excess reactivity needed. This successful operation showed that a thorium fuel cycle was technically feasible in pebble-bed reactors. A scale up of this reactor has been realised: the Thorium High Temperature Reactor (THTR). This prototype reactor (300 MWe), located in Hamm-Uentrop in Germany, was in operation between 1983 and 1989. The THTR fuel elements were composed of mixed UO2 (93% enriched in U-235) and ThO2. The operation was successful. Because of an incident and growing public concern after the Chernobyl accident, the authorities decided to shut the reactor down in 1989.

1.3.3 India

As said before, India has a great interest in using its large thorium resources to assure its energetic independence. Indian is running a research program, which involves a three-stage concept, to efficiently use the thorium resource:

-Stage1: Production of plutonium in a Pressurized Heavy Water Reactors (PHWR) fuelled by natural uranium

-Stage2: A fast breeder reactor (FBR) fuelled by uranium and plutonium produces a larger amount of plutonium and U233 is also produced in a blanket of thorium

-Stage3: The U233 and the plutonium produced in the previous stage will be burnt in an Advanced Heavy Water Reactor

This concept has been tested at a pilot scale, but the implementation at demo scale has not been tested. The construction of the AHWR is planned in the next few years. More information on the Indian thorium fuel cycle program can be found in reference [5].

1.3.4 Summary

The thorium fuel cycle has undergone a lot of R&D efforts in the 60's and the 70's. Pilot scale and demonstration scale reactors have been successfully constructed and operated. However, the interest on thorium cycle fell down at the end of the 70's for more than 20 years. Indeed, the price of the uranium, which reached a very high level in the 70's giving an incentive for a thorium fuel cycle, declined to a low level a few years later, and remained stable until now. Thus, alternatives such as the thorium fuel cycle were not interesting any more, as the uranium cycle was economically competitive and was developed very successfully. But now, as there is growing concern relative to the world uranium resources, and as the renewal of the nuclear energy because of its ability to mitigate our CO_2 emission will affect the world consumption of the uranium resource, there is a renewed interest in the thorium cycle. Moreover, the thorium cycle has a great potential to reduce the amount and the radiotoxicity of the long-lived radioactive wastes. Depending on how fast the next IV-Generation reactors will be in operation at industrial scale, the thorium fuel cycle can play a role in the transition period, even in conventional Pressurized Water Reactors.

1.4 The Thorium fuel cycle

1.4.1 Review of physics properties

The basic process on which all the thorium fuel cycles are based is the production of fissile U233 in the following reaction:

$$^{232}Th + n \rightarrow {}^{233}Th \xrightarrow{22.3\,\mathrm{min}} {}^{233}Pa \xrightarrow{26.95\ \mathrm{day}} {}^{233}U$$

This production process of the U-233 is comparable to the production of Pu-239 from U-238, which takes place in all LWR's: this reaction has a significant impact on the energy production. At the end of a cycle, around 40% of the energy comes from the fission of Pu239.

$$^{238}U + n \rightarrow {}^{239}U \xrightarrow{23\text{min}} {}^{239}Np \xrightarrow{2.3\ \text{day}} {}^{239}Pu$$

It should be noted that the thorium ore contains only the fertile isotope Th-232, and contains no fissile isotope. Natural uranium contains 0.7% of fissile U-235, which can be enriched. Thorium (Th-232) as fuel must therefore be mixed with a fissile isotope like U-235 or Pu-239.

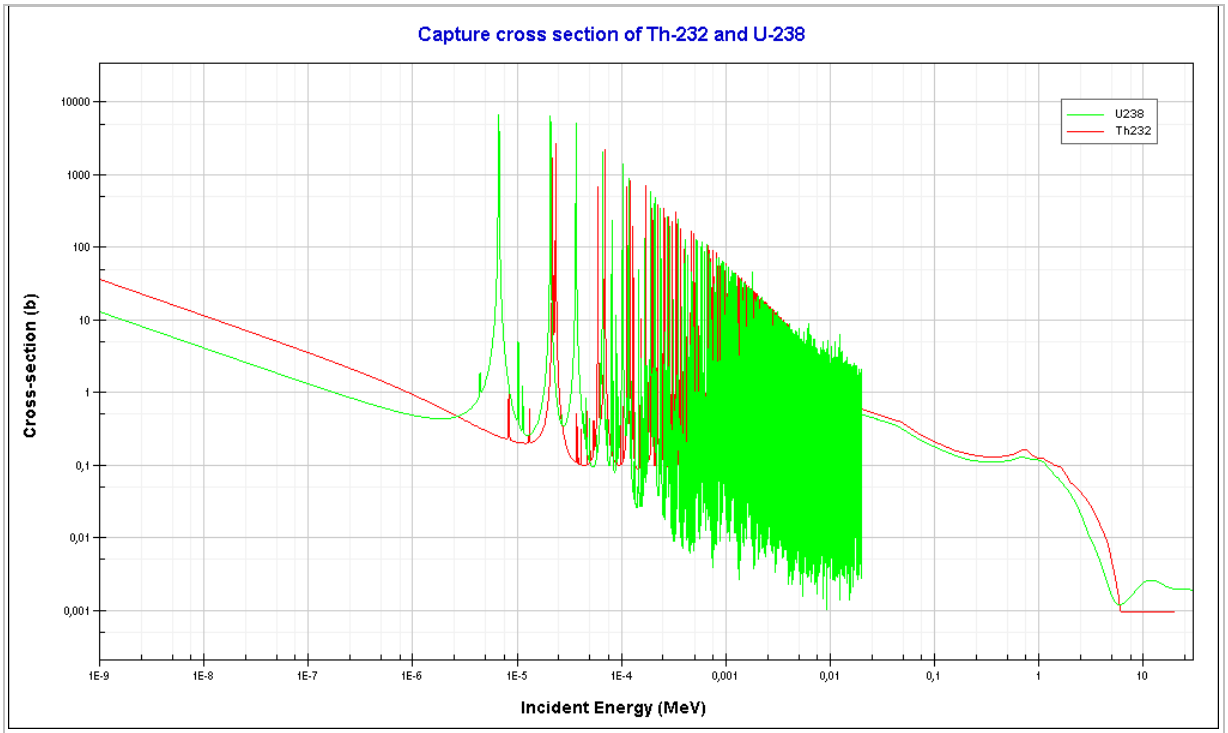

fig. 1-3 Capture Cross Section of Th-232 and U-238

The absorption cross-section (fig. 1-3) of Th-232 is almost three times higher than the one of U-238 in the thermal range until 1eV. This should lead to a better conversion of Th-232 into the fertile nucleus U-233, compared to the conversion of U-238 into Pu-239 for thermal reactors. However, a greater absorption in the thermal spectrum requires a higher enrichment in fissile material to achieve the same burnup. It must be noted, that unlike Np-239 which decays to Pu-239 in 2.3 days, Pa-233 decays to U-233 in 27 days. This could cause a reactivity increase a long time after the shutdown.

Properties of the fissile nucleus U-233

The fission cross-section of U-233 is comparable to the fission cross-section of the usual fissile material U-235 in the thermal range, as can be seen in fig. 1-4. In the epithermal range, the first resonances of U-233 have higher cross-sections than U-235, which can play

a role in the dynamic behaviour of the reactor in case of important shifts of the neutron spectrum.

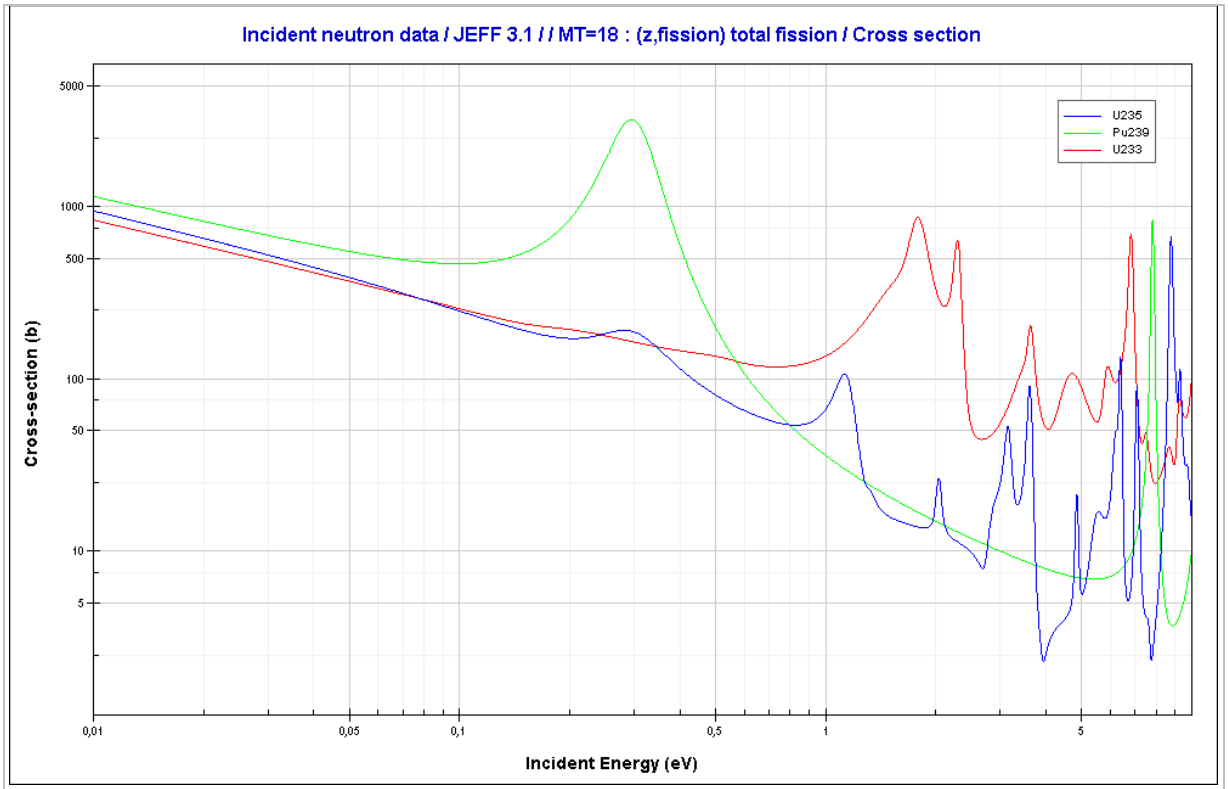

fig. 1-4 Fission cross-section of U-233, U-235 and Pu-239

	E_f (Mev)	σ_f (barn)	σ_c / σ_f	η (thermal)	β (pcm)
U-233	190	531,1	0,09	2,27	276
U-235	192,9	582,2	0,17	2,06	650
Pu-239	198,5	742,5	0,36	2,1	210

Tab. 1-1 Fissile elements properties in the thermal region

We can see in Tab. 1-1 that U-233 is a very good fissile material in a thermal spectrum because it has the best average neutron production per neutron absorbed (η). Moreover, it has the lowest capture/fission ratio for thermal neutrons, which should limit the production of heavier nuclei. In comparison, the capture/fission ratio of Pu-239 is 4 times higher.

As can be seen in Tab. 1-1, the fraction of the delayed neutrons in the fission of U-233 is about half of U-235. This can have consequences on the dynamic behaviour of the reactor. Even if U-233 has very good fissile properties, this must be put in parallel with the other neutron physical properties of all other elements of the thorium fuel cycle. Only reactor calculations, which take into account all effects, can determine if a thorium fuel cycle has good properties in terms of breeding, safety, etc…

1.4.2 Fuel cycle options

As for a classical uranium cycle, there are two main types of cycles: the once-through cycle (or open cycle) and the closed cycle. In the open cycle, a fuel containing thorium and another fissile material is loaded in the reactor, but the spent fuel will not be reprocessed at the end of the cycle. In the closed cycle, the fuel will be reprocessed at the end of the cycle, offering better performances of the cycle, because no fissile material is lost. However, it involves a lot of technical issues associated with the reprocessing and the refabrication of the fuel, which are significant from a commercial point of view.

Once-through cycle

The main interest of the Once-Through Then Out (OTTO) cycle (or OT cycle) is the absence of the reprocessing and refabrication of the fuel.

In a OT cycle, thorium can be mixed with fissile material like enriched uranium or plutonium. To optimize the in-situ burning of U233, a solution can be the seed-blanket concept (the Radkowski concept [7]), where the seed contains fissile material providing neutrons to the blanket containing thorium. This concept offers great conversion ratios, but has a poor energy distribution, and the uranium produced in the thorium blanket is very high enriched in U233, which should be avoided. Without seed-blanket concept, the breeding and fissioning of the U233 is limited for LWRs, and the fissile material must be more enriched than in a conventional fuel cycle. A thorium OT cycle offers similar performances to the OT uranium cycle. For this reason, a thorium OT cycle should not be considered, because of the higher costs involved.

However, the OT cycle has a great potential to burn plutonium coming from the spent fuel of PWR or existing military plutonium from dismantled weapons. This will be analysed in part 4.1.

Closed Cycle

In a closed cycle, the spent fuel will be reprocessed to recycle the U233, which will be used as fissile material for a new fresh fuel. The main interest of the closed cycle is that it avoids the loss of the fissile material present in the spent fuel, providing a lot better utilisation of the resources. However, reprocessing and refabrication are associated with numerous engineering difficulties, and additional costs. In addition to the costs associated with reprocessing for the uranium cycle, the thorium closed fuel cycle involves even higher costs. Indeed, the reprocessing of a spent fuel containing thorium requires shielding because of the production of U-232, which has hard gamma emitting decay products [6]. The closed cycle allows a reduction of the waste production, which represents a reduction of the radiotoxicity of the waste per energy produced. Combined to the reduction of minor actinides inherent to the thorium cycle, this represents a great advantage of the thorium closed fuel cycle over the classical uranium cycle.

The closed cycle is the most interesting way of using the thorium resource to save the uranium resource. There are many possibilities of fuel cycles in Light Water Reactors, depending on the topping fuel, which will be used at the beginning to start the cycle, the fuel which will be mixed to the recycled U233 after each processing, and many other parameters.

The purpose of this study is to determine the influence of determined parameters on the efficiency of the thorium fuel cycle concerning key aspects such as natural uranium consumption, wastes and proliferation resistance.

As explained before, thorium does not contain any fissile material, because the only isotope contained in natural thorium, Th-232, is not fissile. Thorium must therefore be mixed with a fissile material. This fissile material can be:

-Enriched uranium (Medium Enriched until 20%, or High Enriched above 20%)
-Plutonium coming from the spent fuel of a reactor (reactor grade Pu)
-Plutonium coming from a dismantled weapon (military grade plutonium)

This mix of thorium and a fissile material will be used as fuel in a LWR in a normal cycle, for a burnup of 45GWd/tHM (Giga Watts day per tonne of Heavy Metal), as can be seen in the left side of fig. 1-5. U-233 will be produced by the irradiation of Th-232 in the reactor. A part of this U-233 will be burnt in-situ in this first cycle, providing a part of the energy, but an amount of U-233 will be left in the spent fuel. This U-233 will be recycled and mixed with thorium and one other fissile material, as can be seen in the right side of fig. 1-5. U-233 will be reprocessed at each end of cycle, lowering the amount of other fissile material needed for each new cycle in reactor.

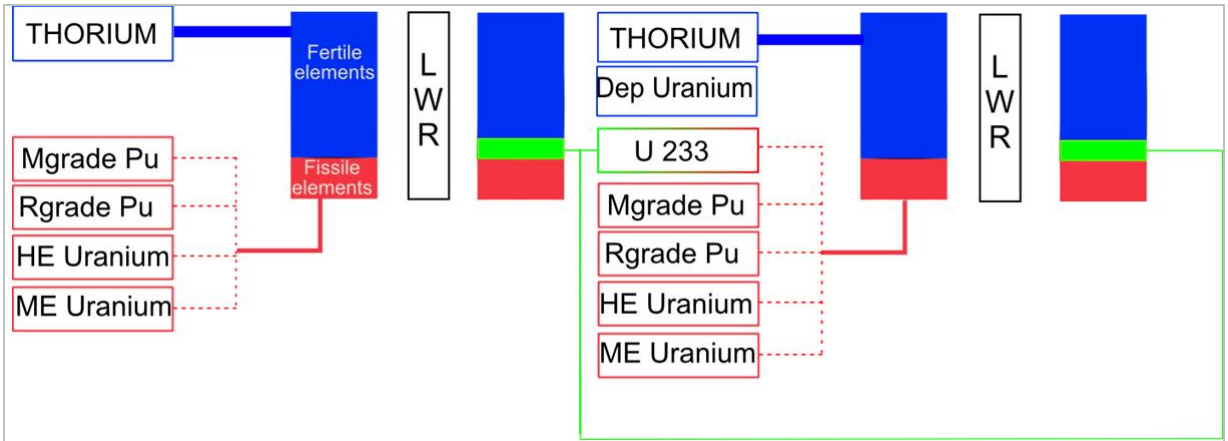

fig. 1-5 Closed fuel cycles options

Each possibility will be analysed as described in section 1.5, and compared to a standard uranium fuel cycle.

1.5 Objectives and parameters of the analysis

All the possibilities of thorium cycles represented in fig. 1-5 have their own advantages and drawbacks, and different objectives can be assigned to each one. They all have numerous parameters that must be analysed. This section will list the main objectives that have been applied to the investigations of this work, and give the related parameters influencing the results.

1. Natural uranium consumption

The production of U-233 is a key aspect because it strongly influences the amount of fissile materials that is needed at each refuelling of the reactor. The saving of fissile materials is the base idea of using thorium as a fuel and this aspect will be carefully assessed. To determine the potential savings, one needs to compare the consumption of fissile materials of the thorium fuel cycle to a reference. As explained before, a once-through cycle with thorium should not be considered because it is not more efficient that a uranium once-through cycle. The potential of the thorium can only be exploited within a closed fuel cycle. A closed fuel cycle is technically a lot more complicated to realize and involves higher costs. For this reason, it would not make sense to compare a closed thorium fuel cycle with a uranium OT cycle: a closed fuel cycle always offer better performances. The thorium and the uranium closed fuel cycles will therefore be compared to each other. For this comparison, the aspect of the consumption of natural uranium per unit of electrical energy produced was chosen. Indeed, the main objective of all innovative reactor or fuel concepts is to save the uranium resource. The main objective of our calculations will be to minimize the consumption of natural uranium of our reactor.

There are numerous parameters influencing this consumption of natural uranium. The thorium proportion in the fuel is one of the most important parameters. The more thorium is present in the fuel, the more U-233 will be produced. Together with the thorium concentration, the moderation ratio has a great influence on the U-233 concentration. Indeed, it influences the neutron spectrum, on which the capture cross-section of Th-232 is strongly dependant. The number of recyclings will as well be a key parameter in the estimation of natural uranium savings. It might be expected that there is an optimal number of recyclings relative to the consumption of uranium. Indeed, if a recycling should always save fissile resources, it reintroduces in the reactor a fuel that is contaminated with undesirable nuclides.

2. Acceptable safety coefficients

The introduction of thorium and the production of U-233 will inevitably change the behaviour of the core. However, it must be guaranteed that the safety coefficients remain at least as good as in the case of a classical uranium fuel. The concerned safety coefficients are the Doppler coefficient (or fuel temperature coefficient), the moderator density coefficient and the void coefficient. These coefficients describe the reaction of the reactor to a perturbing event. Their estimation results of a complex superposition of effects that mainly depend on the spectrum and on the composition in fissile nuclides in the fuel. The parameters that must be investigated are the moderation coefficient, and the composition of the fuel.

3. Reduction of the spent fuel radiotoxicity

As for the safety coefficients, it will not be acceptable to have a spent thorium fuel with a higher radiotoxicity than for the standard uranium case. The radiotoxicity depends on the fission products concentration, the plutonium concentration and the minor actinides concentration in the fuel. There is no major changes in the fission products from U-233 compared to U-235. However, thorium has potential to reduce the minor actinides and plutonium production. This potential must be quantified. The parameters influencing the radiotoxicity will be the thorium and the uranium content, in combination with the moderation ratio. Attention will as well be paid to the production of the radioactive U-232, which is specific to the thorium fuel cycle.

In addition to these three main points, other aspects are studied, in particular concerning the particular ability of thorium to burn plutonium efficiently. This aspect will be quantified and the main parameters influencing this will be identified.

2 Reactor and fuel cycle simulation

This section will introduce the simulation tools used in this work for the fuel cycle analyses, whose objectives and parameters were described in the last section. The physical models used in the neutron physical simulation of nuclear reactors will be introduced. Details will be given on burn-up calculations, which are the base of fuel cycle investigations. The computer code KAPROS that was mainly used for the reactor simulations will be briefly described. A validation work of KAPROS in the specific context of thorium fuels has been conducted and is presented in this section.

2.1 Reactor physics basics

The main task of reactor physics calculations is to determine the criticality of a reactor. A reactor must contain enough fissile material such that the core can become critical and can produce energy. Practically, the reactor must be designed in a way that the effective criticality is greater than one for many reasons. The main reason is the fuel depletion, which will lower the criticality of the reactor. There must be an excess of reactivity at the beginning of the irradiation that is sufficient to maintain the reactor critical during a determined period of time. During operation in a light water reactor, neutron poisons must be introduced into the core to compensate the excess of reactivity and bring the effective criticality to exactly 1. These neutron poisons are then slowly removed during operation to compensate the loss of criticality due to the depletion of fissile materials and the buildup of fission products. How much and what kind of neutron poisons, as well as the changes with time is another task of reactor physics calculations. All these results can be obtained when the neutron flux is known. Indeed, all the calculations are based on computation of reaction rates of the form:

$$R_{i,j}(\vec{r}) = N_j(r)\int dE f(\vec{r},E)s_{i,j}(E) \tag{2.1}$$

where the subscript I indicates the type of nuclear reaction is involved (scattering, absorption, fission, etc.) The behaviour of a nuclear core relies on the description of the neutron population represented by the neutron flux Φ (density multiplied by the velocity) and its dependence on time, position, direction and energy. The neutron population can be described by the Boltzmann equation, which can be found in its integro-differential form in equation (2.2).

$$\underbrace{\frac{1}{v}\frac{\partial f(\vec{r},\vec{\Omega},E,t)}{\partial t}}_{\text{Flux variation}} = \underbrace{-div(\vec{\Omega}f(\vec{r},\vec{\Omega},E,t))}_{\text{Leakage}} \underbrace{-\Sigma(\vec{r},\vec{\Omega},E,t)f(\vec{r},\vec{\Omega},E,t)}_{\text{Removal}} + \underbrace{\int dE'\int d^2\Omega'\Sigma_s(\vec{r},\vec{\Omega}'\to\vec{\Omega},E'\to E,t)f(\vec{r},\vec{\Omega},E,t)}_{\text{Transfer}} + \underbrace{Q(\vec{r},\vec{\Omega},E,t)}_{\text{Source}} \tag{2.2}$$

v: velocity of the neutron with the energy E

$f(\vec{r},\vec{\Omega},E,t)$: angular flux (nv) at the point $\vec{r}$, in the direction $\vec{\Omega}$, for the energy E ant the time t.

Σ_t: total absorption cross section

$Q(\vec{r},\vec{\Omega},E,t)$: source of neutrons (external sources, spontaneous and neutron induced fission, (n, 2n) reactions etc…

$c(\vec{r},E)$: fission spectrum of the neutrons at the point r and the energy E

This equation expresses the balance of the number of neutrons in the elementary volume of the other variables between t and t+dt:

There are 4 different contributions to this $\frac{\partial n(\vec{r},v,\vec{\Omega},t)}{\partial t} d^3rdvd^2\Omega dt$ balance of neutrons:

1. Leakage
   The leakage term arises form the fact that the neutrons are in motion. The neutron density is very high in the reactor. Consequently, more neutrons will leave the reactor than enter the reactor.
   The number of neutrons crossing a surface ds at the velocity $(v,v+dv)$ and the direction $(\vec{\Omega},d^2\Omega)$ is: $\vec{\Omega} f(\vec{r},v,\vec{\Omega},t)dvd^2\Omega dtd\vec{s}$. Integrating over the surface and using the Green-Ostrogradsky formula, the number of neutrons of direction $(\vec{\Omega},d^2\Omega)$ and velocity $(v,v+dv)$ leaving a volume d³r in dt is: $\vec{\Omega} f(\vec{r},v,\vec{\Omega},t)dvd^2\Omega dtd\vec{s}$ ("Leakage" term)

2. Removal
   The neutrons of the element $d^3rdvd^2\Omega$ can disappear by absorption or transfer to another direction or velocity. This is represented by a total macroscopic cross-section $\Sigma(\vec{r},v,\vec{\Omega},t)$, depending in general on the 4 variables. This gives the "Removal" term: $\Sigma(\vec{r},v,\vec{\Omega},t)f(\vec{r},v,\vec{\Omega},t)d^3rdvd^2\Omega dt$

3. Transfer
   When the neutron hits a nuclei and is not absorbed, it is transferred to another velocity and another direction. This can be represented by a differential scattering cross-section: $\Sigma(\vec{r},v\to v',\vec{\Omega}\to\vec{\Omega}',t)$ which gives the transfer term by summing over all initial velocities and all initial directions.

4. Source
   New neutrons can be produced by disintegrations, fissions or other nuclear reactions. It is usually separated in two terms: external sources and fissions:

$$Q(\vec{r},\vec{\Omega},E,t) = \underbrace{Q_{ex}(\vec{r},\vec{\Omega},E,t)}_{\text{external sources}} + \underbrace{\frac{c(\vec{r},E)}{4p}\int dE'n\Sigma_f(\vec{r},E')\int d^2\Omega' f(\vec{r},\vec{\Omega}',E,t)}_{\text{fissions}}$$

These four terms are summed, with a negative sign for the term 1 and 2, as they represent a negative contribution to the overall balance of neutrons.

For practical calculations, number of assumptions can be made to facilitate the solving of the equation.

The simulation of the behavior of a reactor core is generally done for fast transients or quasi-static burnup calculations. In transient calculations, the dynamic behavior of the core is studied. The purpose of such calculations is to estimate all the feedback effects and to determine the overall reaction of the core to an perturbing event, like a loss of coolant, a sudden rise of power…Indeed, an augmentation of the neutron flux will lead to higher fission rate and energy release in the fuel. As a consequence, temperature of the fuel and of the coolant will rise, causing different feedback effects. The rise of the temperature of the fuel has a negative effect on the reactivity because of the Doppler effect (increase of the effective resonance integral due to the broadening of the resonance). In the fuel, the rise of temperature will decrease the moderator density, which lowers the moderation but decreases the absorption, leading to positive or negative feedback depending on the spectrum. These are the main feedback effects, but there are other feedbacks that must be taken into accounts to have a precise estimation of the behavior of the reactor in transients. The feedback effects have different time constants, which have significant impact depending on the goal of the calculation. Thus, Doppler feedback is immediate compared to moderator temperature feedback. The calculation of these effects requires the solving of the time dependant Boltzmann equation. The present works focuses on the long term behavior of the reactor, by performing burn-up calculations. The goal of these calculations is to estimate the evolution of the isotopic composition of the fuel with the burn-up. The only modifications in a static production phase in a reactor are the slow motion of the control rods, the slow modification of the boron concentration in the primary coolant and the slow depletion of the fissile material in the fuel. In this case, the variation of the flux with time is very slow. For these reasons, we can make the following assumptions:

-The variation of the flux with time is small enough to be treated like a succession of static states.

-The external neutron source can be neglected when considering that the reactor is in steady state.

-If the scattering is considered to be isotropic, the angular dependency can be resolved by integration over the full angular range. This approximation leads to the *Diffusion Equation*.

Energy discretization

To simplify the solving of the Boltzmann equation, the energy variable has to be discretized. For deterministic calculation like in our case, the full energy range is divided in groups. In this case, the flux and the cross sections are calculated for each group in the following manner, where g is the group index and x the neutron reaction type:

$$f^g(\mathbf{r}) = \int_{E_g}^{E_{g-1}} dE\Phi\ (\mathbf{r}, E) \tag{2.3}$$

$$\Sigma_x{}^g(\mathbf{r}) = \frac{1}{f^g(\mathbf{r})} \int_{E_g}^{E_{g-1}} dE\Sigma_x(\mathbf{r}, E) f\ (\mathbf{r}, E) \tag{2.4}$$

With this formalism, after integration in energy, the Boltzmann equation can be expressed by a system of G coupled equations if G is the number of energy groups. However, for solving this equation, the neutron flux $f\ (\mathbf{r}, E)$ needs to be known. The problem is that the calculation of the flux is one of the final aims of a reactor calculation and it is unknown at the moment of group cross section calculation. For this purpose, an approximation of the flux has to be done.

The KAPROS calculations used for this work use 69 energy groups. KAPROS can use up to 350 groups, which gives finer results, in particular because of a better description of the resonances and of the outscattering, but this requires higher calculation time. Further assumptions are usually made concerning the position and direction dependence of the Boltzmann equation, to facilitate the solving of the equation. Details can be found in reference [7]. Further information on reactor physics can be found in the references used for this part: [15], [16] , [17], and [28].

2.2 Burn-up calculations

The calculation methods used in the present work are so-called burn-up calculations. The main objective of this kind of calculations is to describe the evolution of the criticality and of the isotopic composition of the fuel during the energy production. These two results are the base of fuel cycle investigations. Indeed, the evolution of the criticality with the burnup, in particular the loss of criticality, will help to determine the needed concentration in fissile isotope at the beginning of a cycle. The fissile isotope concentration determines the surplus of criticality present at the first fission in the fuel, and then if we know the loss of criticality at each time step, we can find the burnup that it is possible to reach. Inversely if a final burnup is set as reference (45GWd/tHM in our calculation), it is possible to determine the needed fissile isotope concentration in the fresh fuel. A burn-up calculation can as well determine the precise isotopic composition of the fuel after irradiation. This important result allows the simulation of a closed fuel cycle with recycling of the spent fuel.

The base of burn-up calculations is the solving of the time-independent Boltzmann equation. The neutron flux and its dependence on energy and positions and the reaction rates will be calculated at each time step. The build-up and depletion of nuclides will then be estimated between two time steps. In particular, the depletion of the fissile nuclides initially present in the fuel (U-235 for example) and the building of new fissile nuclides (U-233 from Th-232 or

Pu-239 from U-238) will determine the criticality of the fuel. The mathematical methods used in this kind of calculations will be briefly described in the following.

The general equation of formation and disappearance of a nuclide may be written as follows [18]:

$$\frac{dX_i}{dt} = \sum_{j=1}^{N} l_{ij}\, l_j X_j + \Phi \sum_{k=1}^{N} f_{ik} s_k X_k - (l_i + \Phi s_i) X_i \text{ for } i = (1,...N) \qquad (2.5)$$

X_i: atom densities of nuclide i

l_i: decay constant of nuclide i

s_i: spectrum averaged neutron absorption cross-section of nuclide i

l_{ij} and f_{ij} are the fractions of radioactive decay and neutron absorption by other nuclides which lead to the formation of nuclide i

Φ is the position and energy averaged neutron flux.

The system of equations formed by the equation (2.5) applied to each nuclide needs to be solved. In this form, the system is non linear since the flux depends on the fuel composition which varies between two time steps. However, the flux variation is very low with the time. It can be considered to be constant between two time steps, making the system linear and easier to solve. The time steps have to be chosen carefully to take into account different nuclides production that have an impact on the neutron flux, but with different time constants. For example, the xenon build-up at the beginning of the operation has a great influence on the neutron flux, the time steps at the beginning must therefore be small enough to deal with this effect. In the context of thorium fuel cycle investigations, where Pa-233 plays an important role, the times steps are shortened at the beginning of operation, until stabilization of the Pa-233 concentration after ca. 100 days.

With these assumptions, the system of Eq. 1 is a homogeneous set of first-order differential equations with constant coefficients, which may be written in matrix notation as:

$$\dot{X} = AX \qquad (2.6)$$

This equation has the simple solution:

$$X(t) = \exp(At) X(0) \qquad (2.7)$$

X(t) is the vector which contains the atom densities of all nuclides

X(0) is the vector of initial atom densities

Depending on the input parameters, further simplifications may need to be made for computational purposes. Details can be found in the reference [18].

2.3 The deterministic code system KAPROS

As explained above, the analysis of the fuel cycles relies on a good estimation of the needs of fissile material for each fuel and for each cycle, and on reliable calculation of the isotopic compositions of the spent fuels. The calculation of the loss of reactivity during a cycle of energy production allows the determination of the required content of fissile material in the fuel to reach a defined burn-up.

These calculations have been performed with the module KARBUS (Karlsruhe Reactor BUrnup System) of the modular system KAPROS (KArlsruhe PROgram System) developed at the FZK. KAPROS is a code capable of criticality, flux- and power-distribution calculation especially in innovative reactors. It is used for fuel cycle investigations, and in recent international projects like for Accelerator driven Systems (ADS) and Sodium or Lead cooled Fast Reactors (SFR).

The KAPROS code was initially developed for fast reactor investigations and has undergone many major modifications since the seventies. During the work for tight lattice water reactors in reference [7] the code was extended for reliable calculations for thermal, epi-thermal and fast spectrum systems. Detailed information about the models and solutions of the current version may be found in the references [7] and [12]. Considerable efforts were devoted to the validation of the calculation methods and data libraries for various applications. For light water reactor burnup validation in reference [11], a dedicated experiment in the German LWR at Obrigheim [30] was re-analysed with the current version of the KAPROS/KARBUS code system. Good agreement between the results of the KARBUS calculations and the experimental data could be observed. Recently, the code system was applied for fuel cycle analysis in reference [14], including Th/U-233 components. In the framework of the ongoing qualification of the code system KANEXT, as follow-up version of KAPROS, also this type of fuel is under investigation. Comparison of the results of burnup simulations, based on JEFF3.1 nuclear data, for Th/U-233 fuel with the Monte Carlo codes MCNPX [31] and MCB [29] and with the module KARBUS with final group constant libraries indicate good agreement for fresh fuel characteristics. The calculations in this work were performed with a preliminary JEFF3.1 group constant library, giving slightly lower criticality values at end of cycle, compared to current best estimate results with KARBUS. Details of the validation work conducted can be found in section 2.4. In addition to this validation work specific to the investigations on thorium based fuel cycles in light water reactors, a previous validation work had been conducted for ADS with fast spectrums, which showed a very good agreement between KAPROS and MCB. This work can be found in reference [26].

The KARBUS module for burnup calculations uses a part of the modules present in KAPROS to realise the following tasks:

- Determination of the atomic number densities from the defined fuel and geometry
- Calculation of effective multi-group cross-sections
- Three-zone cell transport calculations

- Standard one-group cross section collapsing
- Burnup and depletion calculations
- Evaluation of time dependant inventories from archived burn-up data.

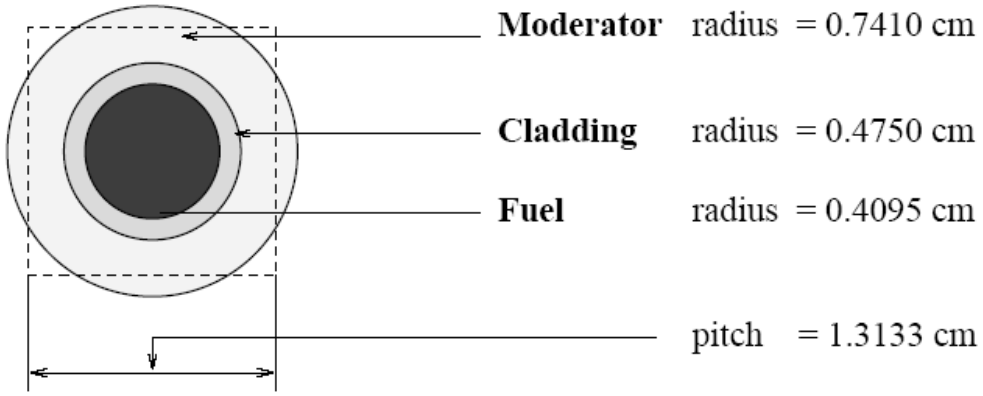

fig. 1-6 Wigner-Seitz cell approximation for the calculations [12]

fig. 1-6 shows the applied three-zone Wigner-Seitz lattice cell approximation for the basic calculations. This cell was proposed for a common benchmark on plutonium recycling in PWRs organized by Electricite de France (EDF) and Forschungszentrum Karlsruhe (FZK) [12]. It is derived from a standard French 900MWe PWR-design and is representative for modern PWRs. Of course, a pin-cell calculation does not have the precision of a full core calculation. However, it requires very shorter computation times. This is very valuable for fuel cycle investigations, because this kind of investigations studies the influence of numerous parameters on numerous results. With the current computational power, pin-cell calculations are the only reasonable solution to do this. However, if the absolute precision of such a calculation is slightly lower than full core calculations, this does not degrade at all the quality of the results. Parametric investigations like in this work only aim to compare solutions with each other and to determine the influence of some parameters. This objective can be fully completed by pin cell calculations. Finer optimisation can then be done with more precise calculations, starting from the best solution identified by the pin cell calculations.

The calculation scheme of the KARBUS module used in this work is given in fig. 2-7. The names in the circles are data formats, the boxes contain the calculation steps. The nuclear data library JEFF3.1 was used for our calculations. The GRUBA library contains the multi-group cross-sections necessary for the neutron flux calculation. The user input including the geometry of the pin and the material composition is used by the MISCH module to prepare the nuclide densities to be used for the neutron flux calculation. Then, the neutron flux will be calculated in each zone (fuel, moderator and cladding) with 69 groups in 0 or 1 dimensions. These results will then be used to calculate the averaged flux and cross-sections necessary for the burnup calculation. The burnup calculation will then be made using this results and

the KORIGEN library. The arrow between the end “Burnup calculation” box and the “MISCH” module represents the data flow between two time steps. The burnup calculation module calculates the material densities after a defined irradiation time and returns it as an input for the calculations of the following time step.

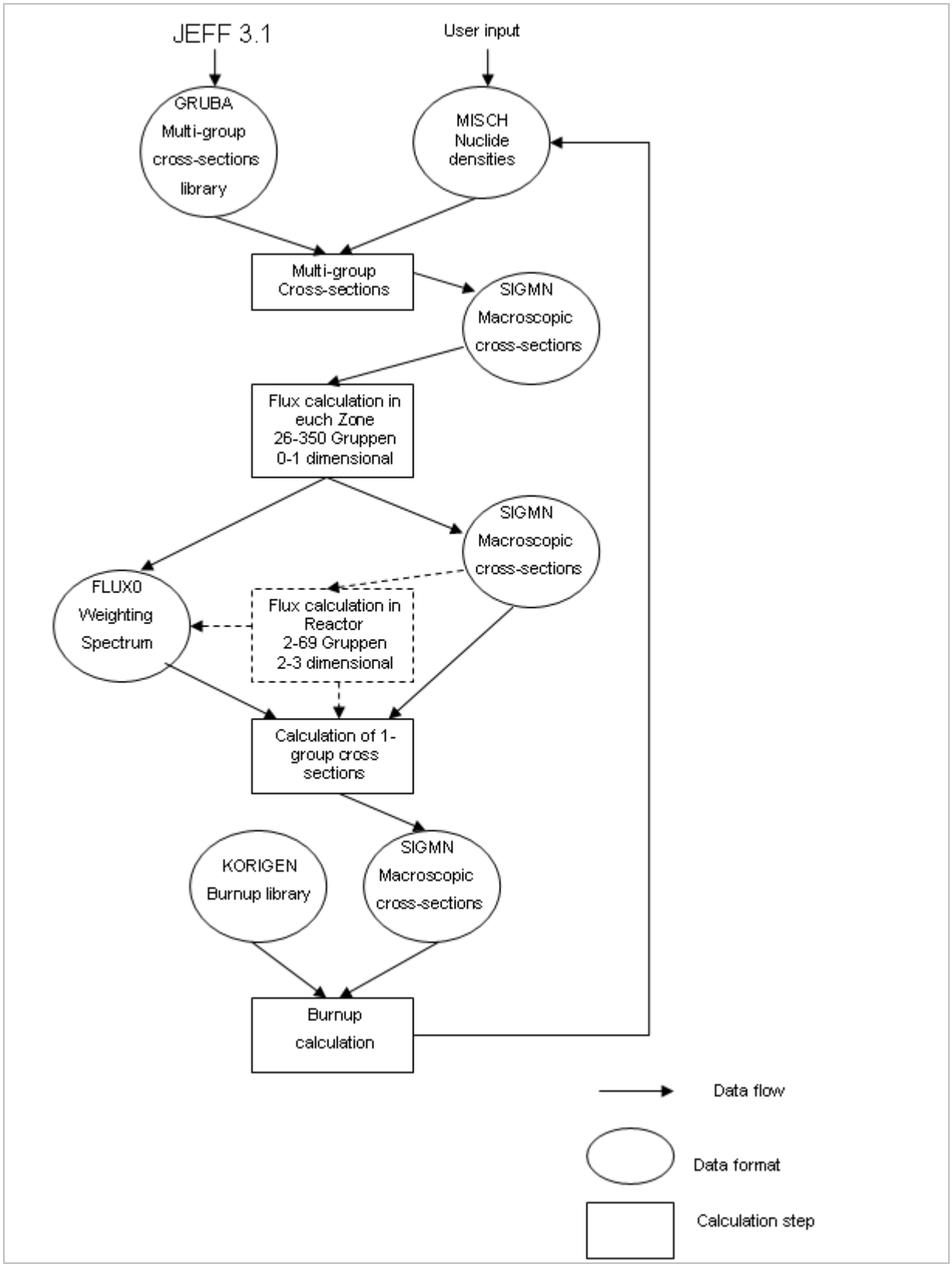

fig. 2-7 KARBUS calculation procedure [7]

The BURNUP module that realizes the burn-up calculation of KARBUS is based on KORIGEN [33], which is an adapted version of the program ORIGEN from the Oak Ridge National Laboratory [18]. This program calculates the build-up and depletion of nuclides during irradiation.

2.4 Validation of the KARBUS module for Thorium fuel investigations

In the framework of the validation of the KANEXT code system, as follow-up of KAPROS for modern computer architectures, also a detailed investigation for thorium fuel was performed. To validate the burn-up calculations with fuels with high thorium concentration, KARBUS calculations have been compared with Monte-Carlo calculations. A typical fuel composition and a typical moderation ratio (with a relatively wide lattice) has been chosen and used as input for all calculations made with different codes. The composition of this fuel is the following: 73% Th, 5.7% U-235 and 20.3% U-238. This is the fuel with the highest thorium concentration among the fuel that was studied within the fuel cycle investigations. This high thorium concentration was chosen because KAPROS was already validated in the past with classical enriched uranium fuel. The goal is here to maximize the potential thorium specific effects, that could affect the reliability of the calculations. The geometry of the fuel element described in fig. 1-6 was used in the MCNP input for all calculations. All calculations used the newest JEFF3.1 database. The composition of the fuel was adapted in our KARBUS calculations to reach a burn-up of 45 GWd/tHM.

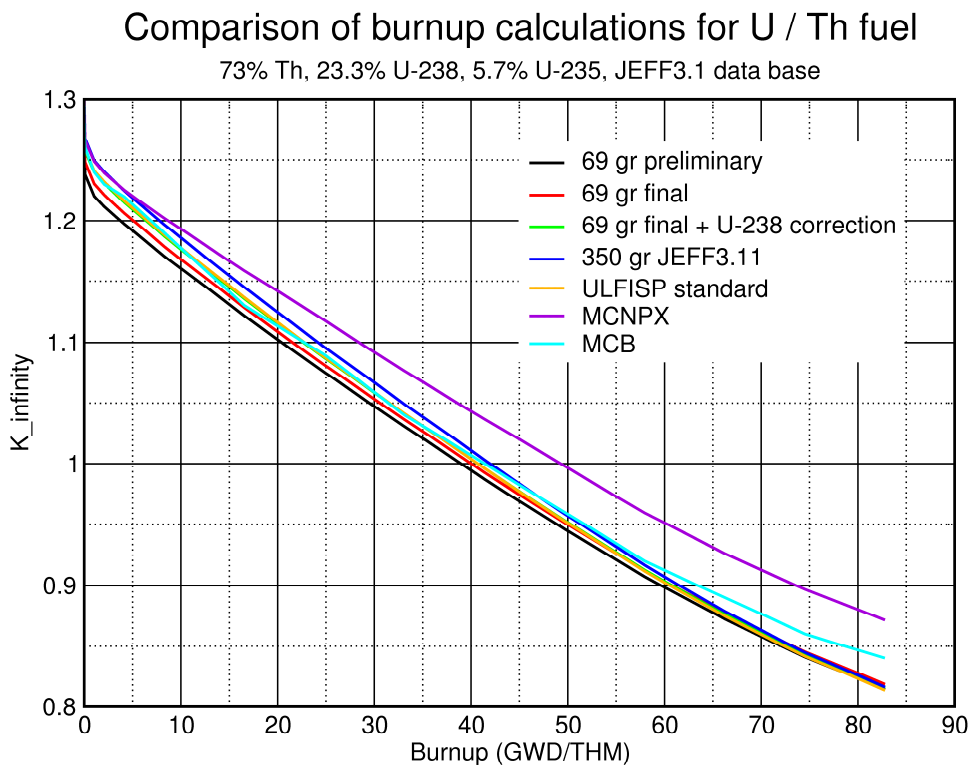

fig. 2-8 Comparison of 7 burnup calculations for one U/Th fuel

In fig. 2-8, 7 different calculations of the burnup behaviour of U/Th fuel are compared.

The “69 gr preliminary” is the KARBUS calculation used for the parametric investigations of the present work. A preliminary 69 groups library based on JEFF 3.1 data was applied.

The “69 gr final” corresponds to “69 gr preliminary” with minor modifications in the library, corresponding to the improvement work of the code during one year. For example, a better description of the delayed neutrons has been implemented during the year, which had not been implemented in the KAPROS version used for the fuel cycle investigations.

The “69 gr final + U-238 correction” includes a different calculation method for the self-shielding effect of the U-238 resonances. In the standard method, a group averaged total cross-section is used for the resonance to calculate the shielding for all other nuclides. Unlike for U-235 and Pu-239, resonances of U-238 are far apart from each other, which leads to an overestimation of the total cross-section, when the averaged total cross-section is used. This can be seen in Annex B. With U-238 correction, the potential cross-section is used, which gives better results [34].

“ULFISP standard” stands for ULtra FIne SPectrum and is the name of a special module of KARBUS. This simulation corresponds to the same conditions as the “69 gr final” calculation but with the new ULFISP module. ULFISP uses a finer technique to calculate the self-shielding of the resonances and gives improved results.

“MCNPX” is a monte-carlo burnup calculation made with MCNPX beta version 27a.

“MCB” is a monte-carlo burnup calculation made with MCNP version 4C [29].

In fig. 2-8 it can be observed that all KARBUS calculations and the MCB calculation show very similar results for the burn-up range relevant for fuel cycle investigations (0-50 GWd/tHM). The “69 gr preliminary” calculation shows the lowest k_infinity values for the full burnup range considered. However, it stays very close to the other calculations. The infinite multiplication factor may therefore have been slightly underestimated in our calculations. As a consequence, the results of the fuel cycle investigations should be better if the criticality was really underestimated.

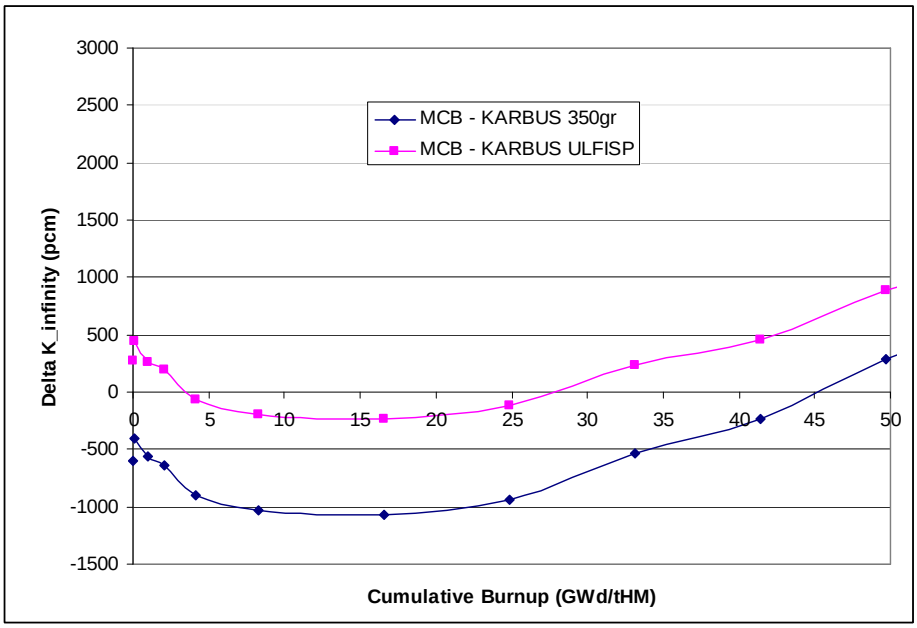

fig. 2-9 K_infinity change between MCB and two KARBUS models

fig. 2-9 shows the change in criticality between MCB and the two finest KARBUS calculations, “KARBUS 350 groups” and “KARBUS ULFISP”. The KARBUS 350 gr calculation remains in the range of (0-1000pcm) compared to the MCB, when the KARBUS ULFISP calculation remains in the range (-250pcm, +500 pcm) for the burnup-range (0-33GWd/tHM) used in the thorium fuel cycle investigations of this work.

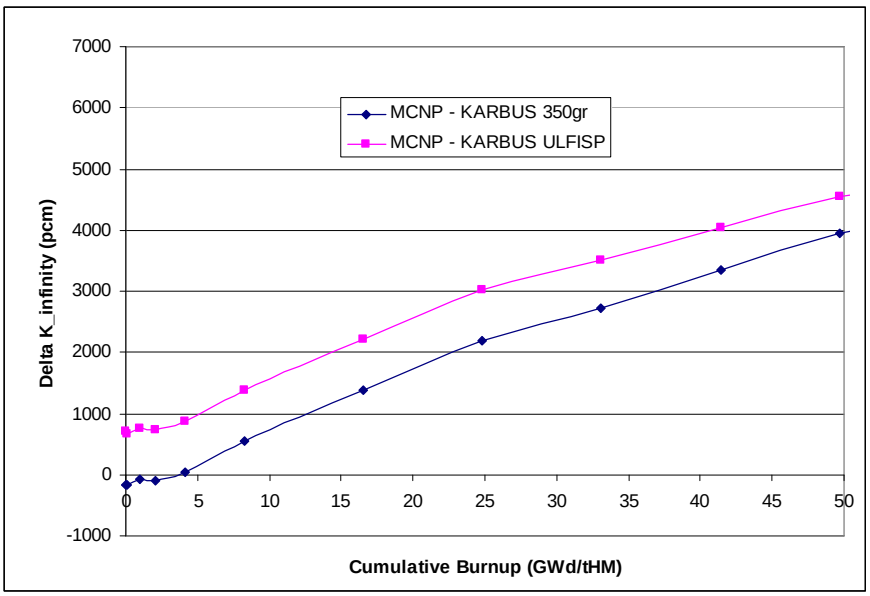

fig. 2-10 K_infinity change between MCB and two KARBUS models

fig. 2-10 is a comparison between the MCNPX burnup calculation and the two KARBUS calculations already compared with MCB in fig. 2-9. The comparison show extremely close results for MCNP and the 350 gr KARBUS calculation until a burnup of 5 GWd/tHM. After this point, the difference grows very rapidly. Despite a 750 pcm difference for the KARBUS UL-

FISP calculation, this comparison shows a very similar behaviour: the difference remains very stable until 5 GWd/tHM and then grows very rapidly. This behaviour could not be explained during this work.

2.5 Summary

The reactor physics basics and the reactor physics program system KAPROS were presented in this section. KAPROS was used for the fuel cycle simulations that will be presented in the next main sections. A validation work of KAPROS was conducted for specific thorium fuels. A representative fuel with high thorium content was simulated with KAPROS and with the monte-carlo codes MCNPX and MCB. The comparison of the results showed that the KARBUS calculations used for the fuel cycle investigations give very acceptable results for the relevant burnup range.

3 Uranium/Thorium Fuel cycles

In the Uranium/Thorium fuel cycle, the fuel will be a homogeneous mix of thorium and enriched uranium. The U-233 produced in each irradiation in reactor will be recycled and mixed to enriched uranium and thorium for the fabrication of the fuel for the cycle n+1.

To follow the scheme of the fig. 1-5, we will in this part consider all the options involving enriched uranium. The most important parameters of the scheme are the thorium content and the uranium enrichment, which can have different values for each cycle. The influence of these parameters will be assessed. It must be noted that every cycle (irradiation in reactor) simulated meet the requirement of a burnup of 45 MWd/tHM: the criticality k_∞=1.03 at BU=45 MWD/tHM, which corresponds to the usual burn-up in a PWR.

3.1 U-233 production

The U-233 production is one of the most important aspects of the thorium fuel cycle. The more U-233 is produced, the more natural uranium or enriched uranium will be saved. The most important parameters influencing this production of U-233 must be determined.

The production of U-233 has been simulated for a fuel composed exclusively of thorium and enriched uranium. The thorium concentration of the fuel has been varied from 0% to 72.5 w%.

Thorium Concentration BOC w%	Fissile material (U-235) Concentration BOC w%	U-235 enrichment of the Uranium vector w%
10	4,8	5,3
20	5,0	6,3
30	5,3	7,5
40	5,5	9,2
50	5,6	11,2
70	5,7	19

Tab. 3-1 Composition of the different fuels considered in the study

The fissile material content in the fuel must be increased when the amount of thorium in the fuel is increased because of the higher absorption cross-section for thorium than for uranium (see fig. 1-3). As we increase the thorium content in the fuel, the uranium content decreases, involving higher enrichment needs. The combination of these two effects leads to important

differences in the uranium enrichment needs for different thorium contents in the fuel. We can see in Tab. 3-1, that for a thorium content of 70 %, the uranium enrichment must be 19%. In comparison to this, the enrichment of uranium must only be 4,4 % (according to this type of calculations) when no thorium is present in the fuel. However, it is possible to use medium enriched uranium (uranium with an enrichment of less than 20%) for a wide range of thorium contents (until 72.5%).

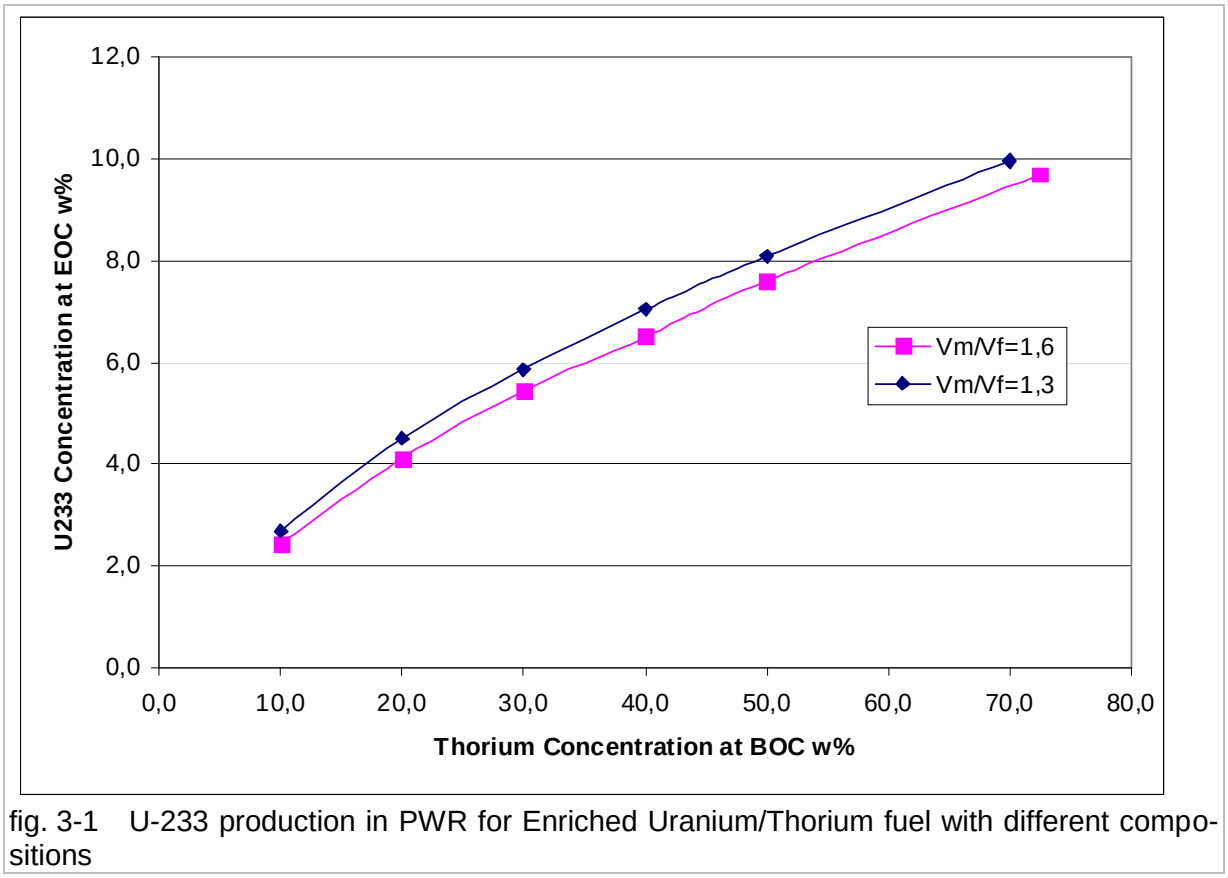

fig. 3-1 U-233 production in PWR for Enriched Uranium/Thorium fuel with different compositions

The fig. 3-1 represents the weight concentration of U-233 in the fuel at the End of Cycle (EOC), which means after 45GWD/tHM of irradiation, and this for two different moderation ratios. This fuel does not contain U-233 at the Begin of Cycle (BOC). The graph shows that the difference in U-233 production between a fuel with low thorium content and a fuel with high thorium content is significant, as it is multiplied by a factor 5 between the two limit cases. The use of a harder spectrum with a moderation ratio of 1.3 leads to a slightly increased production of U233, of around 10 % for 10% Th in the fuel and only 7% for 70% Th in the fuel.

3.2 Fissile element consumption

As mentioned before, a higher thorium concentration in the fuel leads to increased production of U-233, but requires higher amounts of fissile materials in the fuel. The overall con-

sumption of fissile material must be known for all these cases, in order to determine if a thorium fuel cycle can save natural uranium compared to a classical uranium fuel cycle.

Th w% BOC	ΔU5 kg/tHM	Δfiss kg/tHM	ΔUfiss kg/tHM
0,0	-29,0	-22,94	**-29,0**
10,0	-29,4	-21,15	**-27,0**
20,0	-29,7	-20,08	**-25,6**
30,0	-30,1	-19,50	**-24,7**
40,0	-30,7	-19,45	**-24,2**
50,0	-31,1	-19,27	**-23,5**
72,5	-32,0	-19,45	**-22,3**

Tab. 3-2 Fissile element consumption of fuels with different Thorium concentrations

The results from Tab. 3-2 correspond to the same calculations as described in 3.1. **ΔU5** is the balance in U-235 mass in kg in a reactor for one tonne of heavy metals. **Δfiss** is the balance in mass of fissile materials (U-235+U-233+Pu-239+Pu-241). **ΔUfiss** is the change in mass of fissile uranium isotopes (U-233+U-235). This distinction has to be made, because these consumptions in fissile material is significant only if we consider the recycling of the spent fuel, to reuse the non fissioned materials. In this case, the uranium and/or the plutonium will be chemically separated from the other elements. Depending on the strategies, the uranium, the plutonium, or both could be recycled.

In the reactor, the production of Pu-239 and Pu-241 from U-238, and the production of U-233 from Th-232 compensate the loss of fissile material that has already fissioned. We see in the table that the **ΔU5** increases with a growing thorium content in the fuel. U-235 is more consumed with thorium in the fuel as without thorium. On the other hand, if we consider that the produced plutonium and the uranium will be reprocessed, and reused, the balance (to which plutonium has been added) is more favourable. In this case, the consumption of fissile isotopes is 19,3 kg/tHM if there is 50% thorium in the fuel, compared to 23 kg/tHM without thorium in the fuel. If only the uranium is reprocessed, we have to consider the balance in fissile isotopes only for uranium. This is also favourable to thorium fuels: the consumption of fissile uranium isotopes **ΔUfiss** is 29 kg/tHM without thorium and 23,5 kg/tHM with 50% thorium in the fuel.

All fissile isotopes are different (cross-section, neutron yields…), which is not taken into account when we calculate “balances”, but these differences are significant. In addition to this, these balances should be different for irradiations with recycled fuel, which can have an influence on the results. We can conclude from this, that there is more precise work to do by considering recycling, which should lead to net savings of the uranium resource.

3.3 Uranium/Thorium closed fuel cycles

3.3.1 Introduction

A once-through cycle in a Light Water Reactor with a thorium containing fuel does not improve the performance of the reactor: the fabrication of the fuel requires more natural uranium. The analysis of the fissile element balance after reactor irradiation shows that the thorium fuel has a potential to reduce the uranium needs, but only if the spent fuel is recycled. The recycling allows the reuse of the fissile materials that have not been fissioned. These fissile materials are the U-233, U-235, Pu-239 and Pu-241. The recycling consists of a chemical separation of the uranium and/or plutonium from the spent fuel. The rest of the spent fuel is considered to be lost, and will be handled like a waste. It must be noted, that all uranium isotopes will be recycled, and will not be separated from each other. In addition to U-233 and U-235, U-238 and other isotopes like U-234 or U-236 will be present, which are neutron absorbers, and have a negative impact on the reactor. The concentration of these isotopes can grow if successive recycling are realised, which can lead to significant quantities. This can be a limitation for the number of recyclings.

As explained before, the conversion ratio of a thorium fuel in a LWR with conventional technology (with control rods) is always <1. When the uranium from the spent fuel is recycled, it must always be mixed with another fissile material in order to increase the content of fissile material in the fuel to the needed quantity.

The fig. 1-5 describes all the possibilities to mix thorium with other fissile material. In this part, the Uranium/Thorium fuel will be considered. Distinction is made between Medium Enriched Uranium (MEU) and High Enriched Uranium (HEU), the difference between the two being the limit of 20% enrichment. A material with an enrichment higher as 20% is considered as a proliferative material, because it is a lot easier to achieve very high enrichments using such a material, as for medium or low enriched uranium.

The choice of the concentration of thorium is a very important parameter for all fuel cycles. As can be seen in the Tab. 3-1, the concentration of thorium in the fuel determines the necessary enrichment of the uranium that is mixed within the fuel.

3.3.2 Recycling of spent fuel

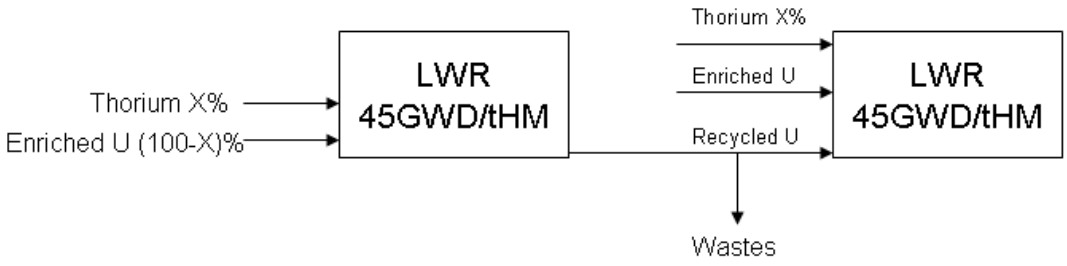

fig. 3-2 Uranium/Thorium fuel cycle

The fuel cycle recycling concept is represented in the fig. 3-2. Fuels with variable concentrations of thorium have been simulated: the X of the figure representing this concentration has been varied. Burn-up calculations have been realised for each one of these fuels. After 45 GWD/tHM of irradiation, we suppose that the fuel will be cooled for a period of around 7 years. We assume that this time will be sufficient, so that all the Pa-233 present in the spent fuel can decay into U-233. Indeed, the decay constant of Pa-233 is around 27 days.

Th concentration w%	U-235 enrichment w%
0,0	4,4
10,0	5,3
20,0	6,3
30,0	7,5
40,0	9,1
50,0	11,2
71,5	20,7

Tab. 3-3 Enrichment of the added uranium for each thorium concentration

After irradiation in the reactor, the uranium vector has changed. A significant part of the U-235 present at the beginning of the cycle has been lost. An amount of U-233 has been produced, but new fissile materials must be added to the fuel, if we want to reload it into a reactor. The most part of the initial U-238 remains in the spent fuel. There are many scenarios for the mixing of new enriched uranium to the spent fuel. As the spent fuel has a lower content of fissile material, it must be mixed with uranium with a higher enrichment. Depending on how high this enrichment is, the amount of this enriched uranium that must be mixed to the spent fuel can be bigger as needed to produce fuel for one reactor.

	BOC	EOC
TOTAL Th	200,0	194,06
U 232	0,0	0,01
U 233	0,0	3,74
U 234	0,0	0,35
U 235	50,0	20,29
U 236	0,0	5,20

U 238	750,1	733,97
TOTAL U	800,0	763,55

Tab. 3-4 Composition of the fuel at the BOC and the EOC in kg for one tonne of a fuel containing 20% thorium

With 20% of Thorium in this fuel, 50.0 kg of U-235 are needed at the BOC. At the end of the cycle, only 20.29 kg of U-235 and 3.74 kg of U-233 are left in the fuel. The total mass of uranium at the EOC is 764 kg. The mixing of new fuel must meet two requirements: after recycling and the mixing of enriched uranium, the new fuel must be 800 kg uranium, with a sufficient content of fissile material.

In this case, the concentration of U-235 in the fuel after the recycling must be 4.72 %. If the enrichment of the added uranium is 75 %, the mixing of 36 kg of this enriched uranium will give 800 kg of uranium, with enough fissile material for 45MWd.

The composition of the fuel after adding enriched uranium would be the following:

TOTAL TH	200,0
U 232	0,0
U 233	4,1
U 234	0,4
U 235	47,2
U 236	5,2
U 238	743,1
TOTAL U	800,0

Tab. 3-5 Composition of the fuel after recycling and mixing of enriched uranium in kg for one tonne of fuel

If the enrichment of the added uranium is lower than 75 %, more uranium must be added to the spent fuel. With an enrichment of 50%, 54kg must be added to the spent fuel, giving 764kg + 54kg =818kg of new fuel. This would give in this case 18 kg of fuel that could not be used. This rest of fuel could be valorised for the development of a pool of reactors using thorium fuel.

For each concentration of thorium in the fuel, the spent fuel has a different isotopic composition. To meet the two requirements (mass of the total fuel and content in fissile material), the added enriched uranium must have a precise enrichment.

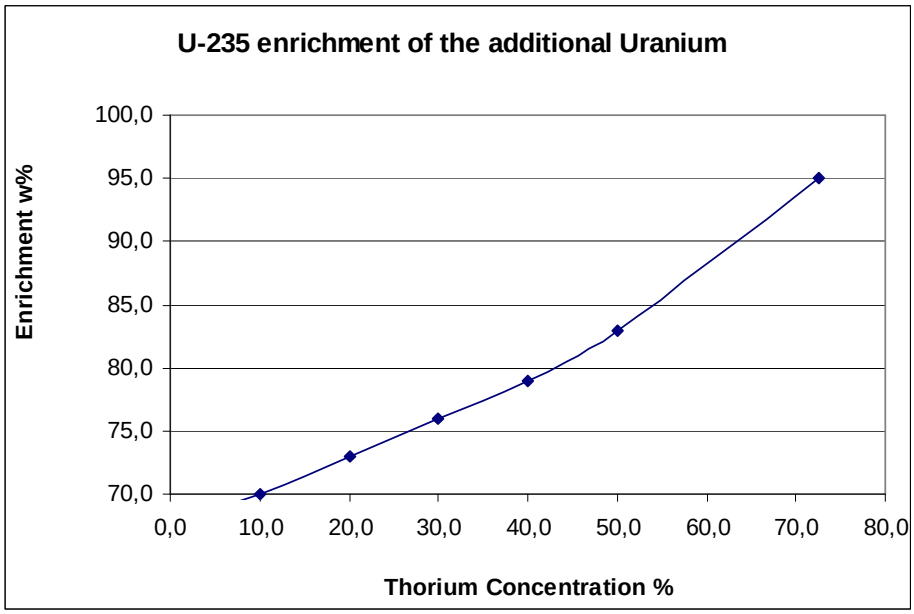

fig. 3-3 Enrichment of the added uranium for different fuels

In fig. 3-3 is represented the enrichment of the added uranium to produce the exact mass of fuel for cycle 2. This enrichment is given for fuels with different thorium concentration (varying from 10% to 73%). This added fuel remains in all cases a so called “High Enriched Uranium”. This enrichment is strongly dependant on the thorium concentration of the fuel. Indeed, for a concentration of 10% Th in the fuel, the enrichment of the additional uranium must be 70 %, but if there is 50%Th in the fuel, the enrichment of the additional uranium must be 83%.

Now if we assume a fixed thorium concentration in the fuel after each recycling, the enrichment of the uranium will vary for each cycle.

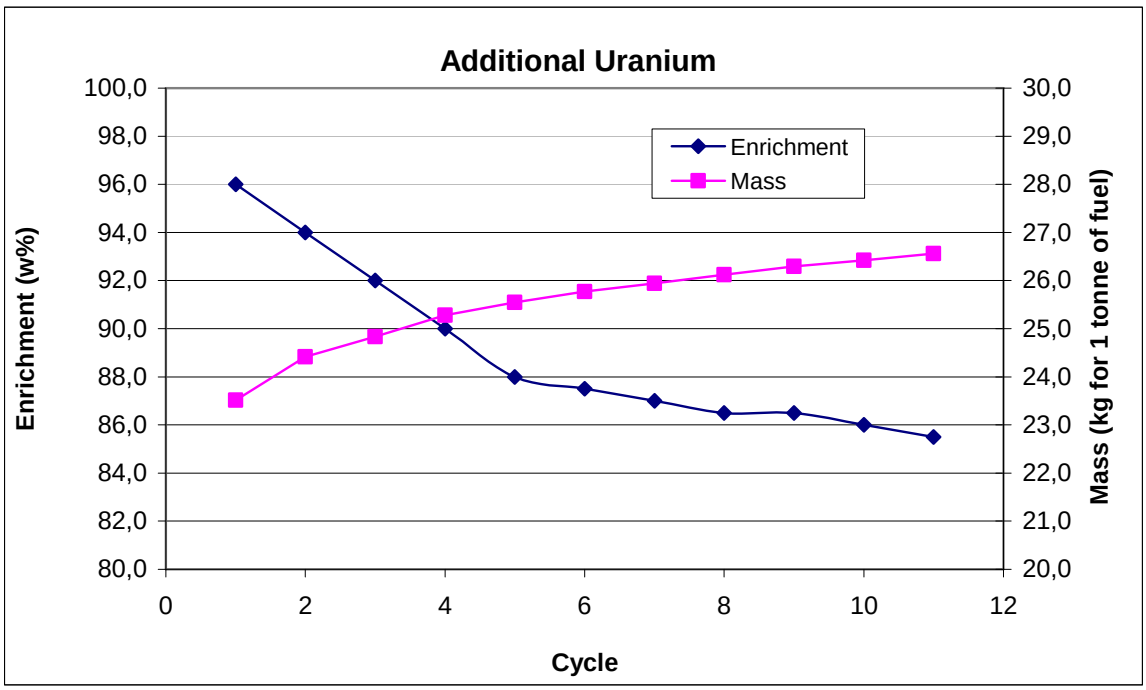

fig. 3-4 Enrichment of the additional uranium in w% for a fuel containing 73% Th

We can observe in fig. 3-4, that the ideal enrichment of the additional uranium goes down for each recycling. On the other hand, the mass of additional uranium that is needed at each recycling shows a slight increase.

Evolution of the isotopic composition of the fuel

The fuel undergoes a modification of its isotopic composition at each recycling. Only the thorium concentration remains the same, as new thorium is used at each refueling. This modification has consequences on the needs of fissile material, because of the growing concentration of U-234 and U-236.

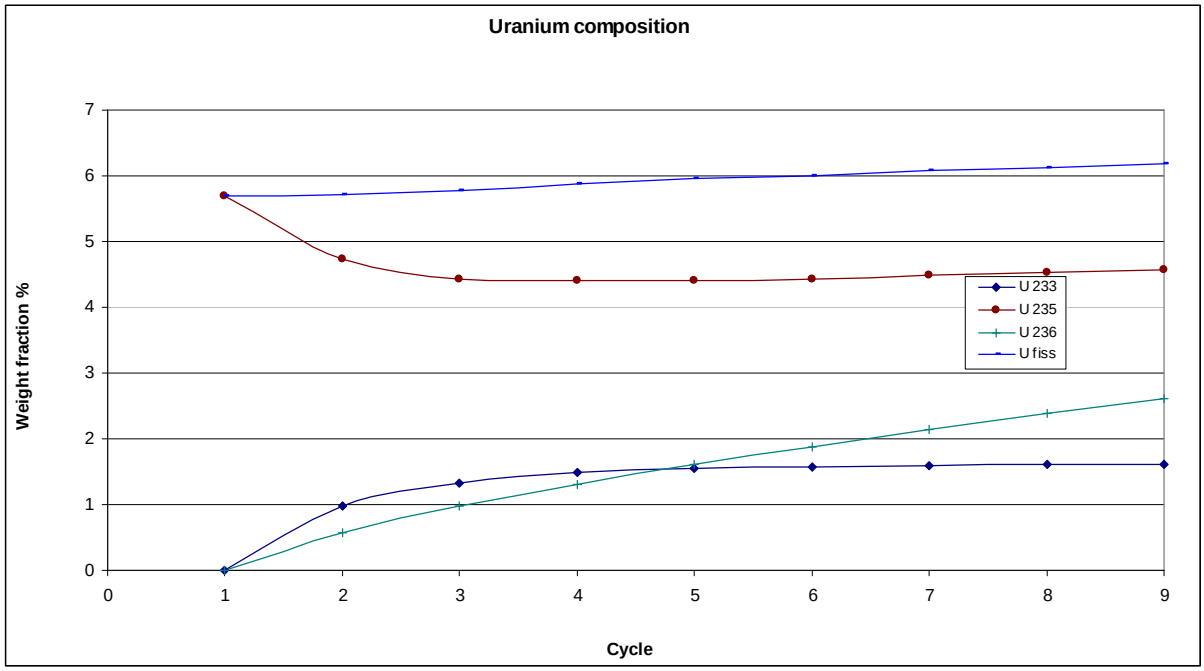

fig. 3-5 Uranium composition along the cycles for a 73% Th fuel at the BOC

It can be seen in fig. 3-5, that no U-233 is present at the beginning of the first cycle: U-233 is produced during the first cycle, and will be reused as fuel for the following cycles. We can observe a relatively quick stabilisation of the U-233 concentration at approximately 1,6 w% in the uranium vector after a few cycles. However, the U-236 concentration does not stabilize, because of its low absorption cross-section. Thus, the concentration of U-236 grows along the cycle, which has consequences on the neutron balance: the fissile material content of the fuel must be increased at each cycle. The blue line, which represents the concentration of fissile isotopes (U-233 and U-235 in the fuel) shows this growth. On the other hand, we can observe that the necessary U-235 concentration goes down for the two first cycles because of the fast growth of the U-233 content in the uranium vector. After stabilisation of it, the U-235 concentration must increase because of the growing concentration of U-234 and U-236.

3.3.3 Natural uranium consumption

It has been shown in the last paragraphs, that the presence of thorium in the fuel produces U-233, which could theoretically lower the consumption of fissile material of the reactor. A recycling of the fuel, with addition of high enriched uranium at each cycle must be realised in order to make real these possible uranium savings. The main aspects of this recycling has been highlighted in the previous paragraph, but it has now to be precisely assessed how much is a thorium closed cycle more favourable than a uranium closed fuel cycle. The criterion of the consumption of natural uranium will be used for the comparisons.

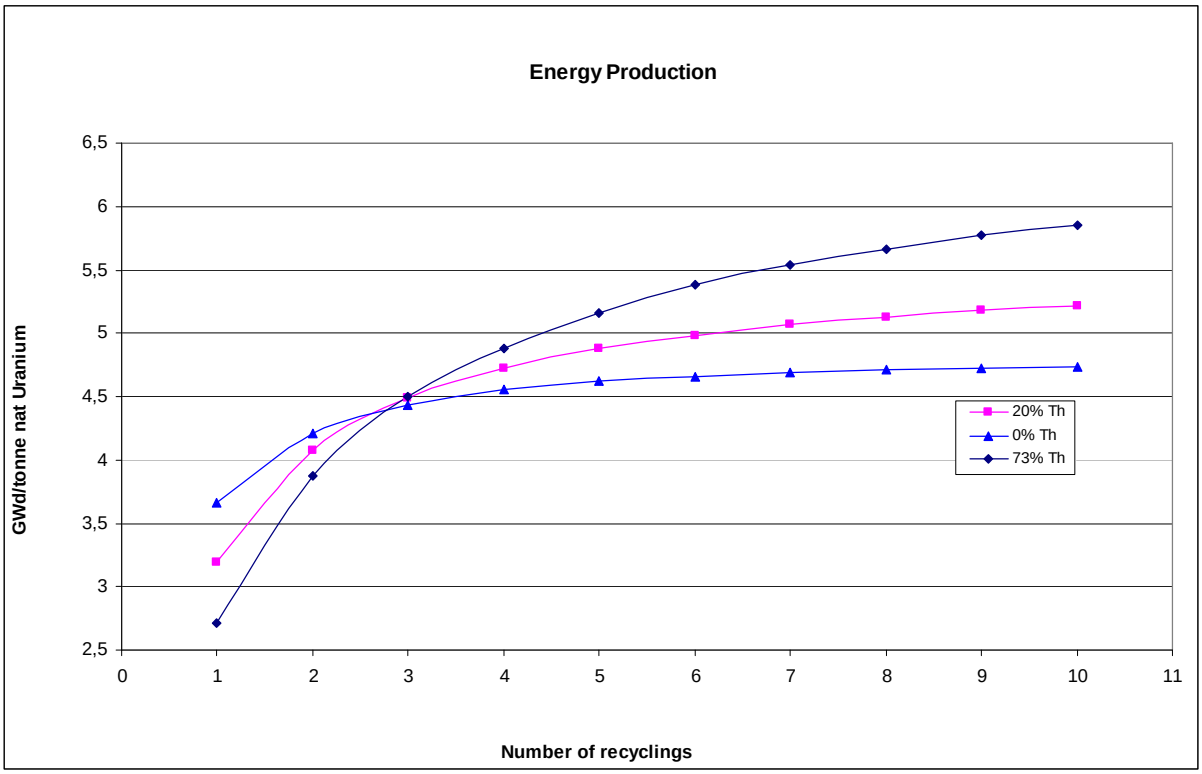

fig. 3-6 Energy production per unit of natural uranium consumed after each recycling for different thorium concentrations in the fuel

In fig. 3-6, three different fuel cycles have been simulated: classical uranium fuel (named 0% Th in the figure), Uranium/Thorium with 20% thorium, and Uranium/Thorium with 73% thorium. The quantity of natural uranium needed to produce enriched uranium have been calculated assuming a concentration of 0,30% of U-235 in the waste uranium of the enrichment process.

This graph shows that the energy production per tonne of natural uranium of a thorium cycle exceeds the one of a pure uranium cycle, and this only after a few cycles (three in these cases). This threshold depends on the concentration of thorium in the fuel. Indeed, the absorption cross-section of Th-232 is higher as the U-238 one. For this reason, the fissile material content (and the natural uranium needed) in the fuel must be higher for thorium-containing fuels, which has a negative contribution to the energy production when it is calculated per unit of mass of natural uranium. After a few cycles, the savings of natural uranium due to the U-233 production tend to be dominating.

As can be seen in fig. 3-6, a thorium containing closed fuel cycle can achieve great savings of uranium in comparison to a closed cycle with only enriched uranium as fuel. After 10 cycles, the fuel containing 73% of Thorium produces 23% more energy per tonne of uranium than the fuel containing only enriched uranium, and 60% more energy compared to a classical once through cycle with uranium fuel.

As observed in section 3.3.3, the fuel cycle providing the best performances in terms of uranium consumption is the fuel cycle containing the more thorium possible. However, there is an upper limitation for the thorium concentration in the fuel. This limitation comes from the needs of enrichment for the high enriched uranium that is added to the spent fuel at each recycling. If the thorium content of the fuel is higher than 73%, it is not possible to produce a new fuel from the spent fuel by adding high enriched uranium, even if it were pure U-235. It would be necessary in this case to dilute the spent fuel into more additional uranium than needed to produce one new fuel, which would give rests of fuel, giving poor energetic performances.

3.3.4 Influence of the moderation ratio

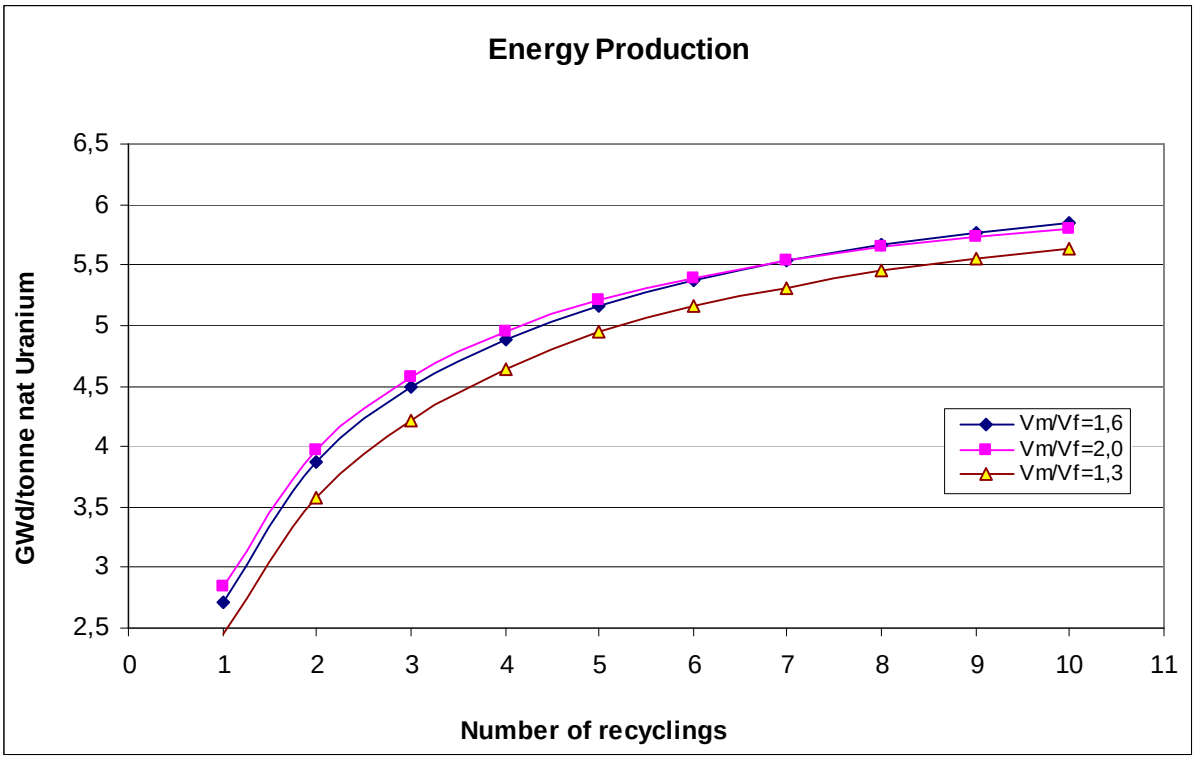

fig. 3-7 Energy production per unit of natural uranium consumed after each recycling for different moderation ratios of a fuel with 73% thorium

The fig. 3-7 presents the energy production per tonne of natural uranium for three different moderation ratios (ratio of the moderator volume and the fuelrod volume). It appears clearly that a change of the moderation ratio (MR) does not make a significant difference in terms of efficiency of the cycle. The cycles with the moderation ratio of 1,6 and 2,0 are very similar, the cycle with Vm/Vf=1,3 offers lower performances. The cycle with the moderation ratio of 2,0 requires a slightly lower enrichment in fissile material than the cycle with 1,6, but has a slightly lower production of U-233, which explains the differences between the two cycles. Indeed, the MR of 2,0 has a higher production of energy at the beginning of the cycle, where

the initial inventory in fissile material plays an important role, but the energy production becomes lower as the MR 1,6 after 8 cycles because of the highest production of U-233, which becomes significant.

3.3.5 Safety aspects

Doppler reactivity coefficients

The doppler coefficient describes the effect on the reactivity of a temperature change in the fuel. The main interest of a doppler effect coefficient calculation is to see the effect of a rise in the temperature, corresponding to a sudden augmentation of the power. The temperature effect on reactivity coefficients has been analysed in numerous works and can be described in this way [7]:

$$a = \frac{dk}{dT} = \frac{A}{T^B} \tag{3.1}$$

$$k = \frac{A \cdot T^{1-B}}{1-B} + C \tag{3.2}$$

a : Doppler coefficient (usually in pcm/°K)
k : Reactivity coefficient
A, B, C : Fuel specific coefficients to be determined for each fuel

The normal temperature of the fuel was set to 800°K in the KARBUS calculations. The criticality has been calculated for a total of 4 temperatures: 600°K, 800°K, 1000°K and 1200°K. The Doppler coefficient a can be calculated for each Burnup step. This has been done for the BOC and the EOC. Equation 2.2 has been fitted to the curve obtained from the 4 points to calculate the coefficients A, B and C.

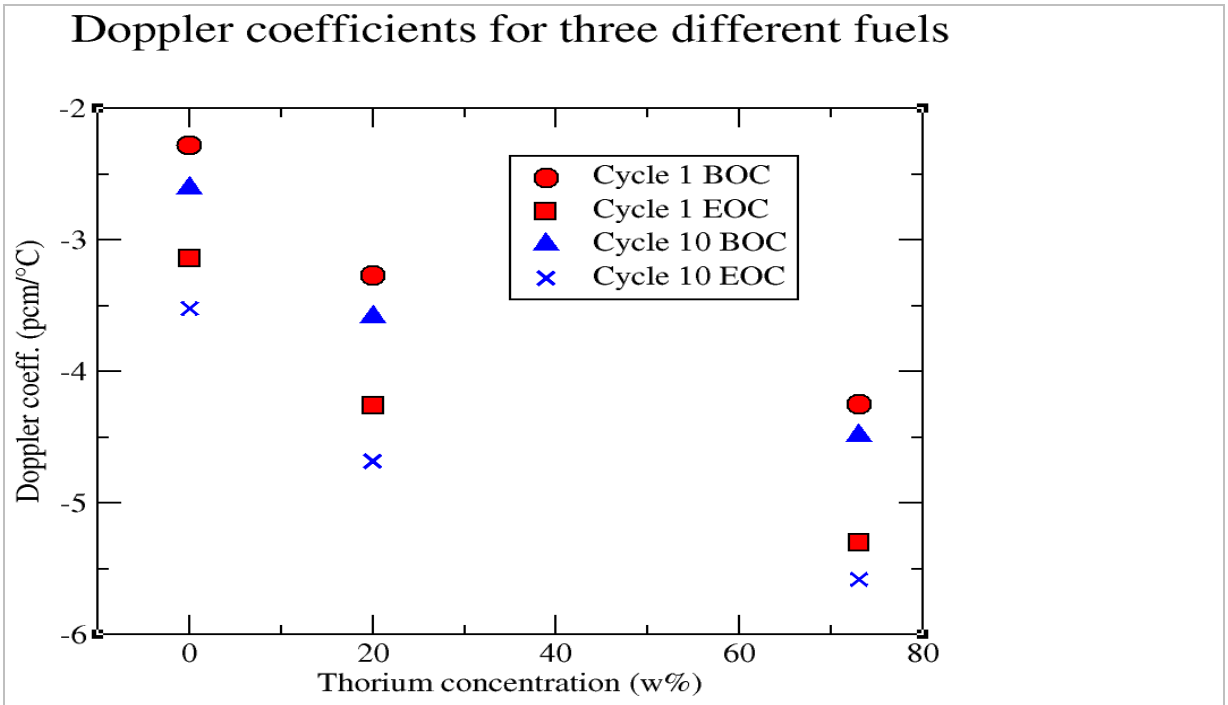

fig. 3-8 Doppler coefficient a for three different fuels in pcm/°K

The results of the Doppler coefficient calculations are presented in fig. 3-8. The calculations have been made for the three cases presented in section 3.3.3, which are three closed fuel cycles, with different thorium concentration for each fuel: 0% (only enriched uranium), 20% and 73%. Each closed fuel cycle is composed of numerous irradiation cycles. The doppler coefficients are presented for the cycle 1 and 10. We can see that the doppler coefficient increases after each recycling in all the cases, which means that the safety of the reactor increases, even after a lot of recycling. It must be noted that the safety of the reactor increases too when the thorium content of the reactor increases. We can observe that a high thorium content in the fuel increases strongly the reactivity feedback to a temperature increase. For the cycle 1, the doppler coefficient of the classical uranium fuel is -2,3 pcm/°C, but it reaches -4,25 pcm/°C for the fuel containing 73% of thorium.

Void reactivity coefficient

The void reactivity change describes the change in reactivity that occurs when there is a total loss of coolant in the reactor. This accidental situation must lead to a negative change of the reactivity in the reactor.

Thorium concentration in the fuel	Cycle 1	Cycle 10
0%	-0,54	-0,47
20%	-0,49	-0,41
73%	-0,51	-0,4

Tab. 3-6 Void reactivity change (Δk) for 3 different fuels for the first and the tenth recycling

This change in reactivity has been simulated for the three fuels considered before, for the first and the tenth cycle. The values of Δk are very acceptable and assure a very good safety of the reactor from this point of view.

3.4 Potential for the reduction of spent fuel radiotoxicity

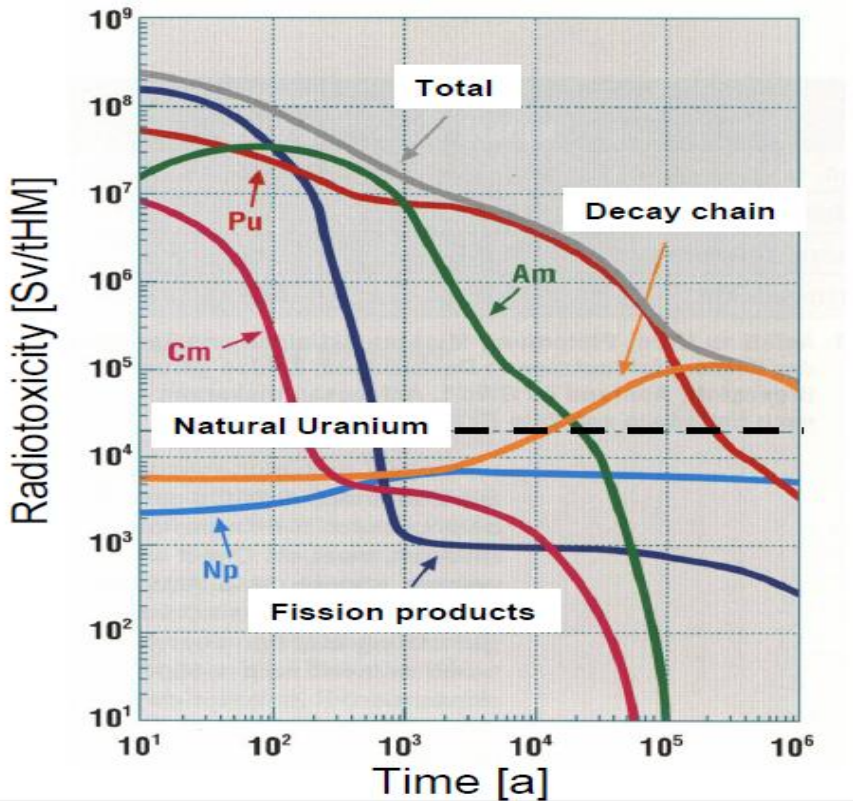

fig. 3-9 Spent fuel radiotoxicity of a typical LWR [2]

The radiotoxicity of the spent fuel is a major issue for commercial fuel cycles. In particular the time after which the spent fuel radiotoxicity becomes equal to the radiotoxicity of the natural uranium. The fig. 3-9 shows the whole radiotoxicity and the individual contributions to this radiotoxicity from the major contributors in the spent fuel of a typical LWR fuelled with enriched uranium. We can see that the minor actinides (which account for around 0,1 % of the total spent fuel mass) and the plutonium are the most important contributors to this radiotoxicity, with the fission products for the first 1000 years. It appears clearly in this scheme that there is a great potential to reduce the whole radiotoxicity of the waste if the production of plutonium and minor actinides (in particular americium and curium) could be decreased. The study of radiotoxicity requires taking into account all the nuclides present in the spent fuel. The contribution of the minor actinides and the plutonium with the potential for their reduction has been studied for thorium based fuel. A more complete study should consider in particular the effect of the presence of U-233 in the spent fuel.

3.4.1 Minor actinides reduction

The minor actinides considered are neptunium, americium and curium. Their typical production routes and rates in a conventional LWR are the following [8] [7]:

-Np-237 from U-235 (2 neutron captures, 650g/tHM for 45GWD/tHM)
-Am-241 from U-238 (3 neutron captures, 130g/tHM for 45GWD/tHM)
-Cm-244 from U-238 (6 neutron captures, 47g/tHM for 45GWD/tHM)

It is obvious that the production rate of an isotope of minor actinide decreases when the needed number of neutron capture to produce it increases. The production of minor actinides from thorium would thus be lower: the mass number of thorium is 232, which compared to uranium-238, increases the needed number of neutron captures by 6 for the production of a particular minor actinide isotope. This number is increased by 2 as well for U-233 (produced from Th-232) compared to U-235. The use of thorium as part of the fuel should therefore lead to a lower production of minor actinides compared to a classical uranium fuel.

	0% Th fuel		73% Th fuel	
Cycle	Particular	Cumulative	Particular	Cumulative
1	0,61	0,61	0,46	0,46
2	1,04	1,65	0,84	1,3
3	1,37	3,02	1,07	2,37
4	1,64	4,66	1,25	3,62
5	1,87	6,53	1,39	5,01
6	2,06	8,59	1,52	6,53
7	2,23	10,82	1,63	8,16
8	2,38	13,2	1,74	9,9
9	2,51	15,71	1,83	11,73
10	2,63	18,34	1,91	13,64

Tab. 3-7 Minor actinides production in kg for 45GWd for two different fuels

In the Tab. 3-7, the production of minor actinides of two different fuels is presented: a classical uranium fuel, and a fuel with high thorium content of 73%. The column “particular” gives the production of MA for each cycle. The column “cumulative” is cumulated production of MA, which takes into account all the previous cycles. The minor actinides considered are: Np-273, Np-239, Cm-242, Cm-243, Cm-244, Am-241, Am-242 and Am-243. For the two fuels, we observe an increase of the production of minor actinides at each step: +330% for the 0%Th fuel and +315% for the 73% Th fuel between the 1^{st} and the 10^{th} cycle. This increase is due to the increase of the production of Np-237: the production of americium and curium decreases slightly for the following reasons:

-The two main sources of Np-237 have a growing concentration in the uranium vector:

-The concentration of U-236 grows in the uranium vector recycling after recycling as can be seen in fig. 3-5

-The concentration of U-235 is increased in each new fuel after the recycling

- The U-238 (precursor of Am and Cm) content in the fuel decreases at each recycling

If we compare the two fuels, we can see that the fuel containing 73%Th has produced 25% less minor actinides after 10 cycles than the pure uranium fuel. The production of americium and curium is 300 % lower for the thorium containing fuel as for the uranium fuel, and the production of neptunium is 25% lower for the thorium fuel. As the neptunium production is around 100 times the production of Am and Cm, we can say that the performance of the thorium fuel in terms of reduction of minor actinides production is not directly due to the lower production of Am and Cm from the Th-232 as from the U-238, but is far more due to the replacement of U-235 by U-233 as fissile material.

We can conclude from these results that the thorium fuel potential for reducing the production of minor actinides is confirmed. The production of americium and curium has been divided by three, and the production of Np-237 decreased by 25%.

3.4.2 Plutonium reduction

In the context of the reduction of the radiotoxicity of the spent fuel, the plutonium present in the spent fuel must be considered too. If the plutonium is not recycled and burned, it plays a significant role in the long term radiotoxicity of the wastes, as highlighted in fig. 3-9. Plutonium is mainly produced by a neutron capture of U-238. Therefore, the use of thorium in a fuel, which is a replacement of a part of the U-238 by Th-232, should lead to decreased plutonium production.

	0% Th fuel		73% Th fuel	
Cycle	Particular	Cumulative	Particular	Cumulative
1	8,96	8,96	4,11	4,11
2	9,53	18,49	4,57	8,68
3	9,97	28,46	4,83	13,51
4	10,31	38,77	5,01	18,52
5	10,6	49,37	5,16	23,68
6	10,84	60,21	5,29	28,97
7	11,05	71,26	5,4	34,37
8	11,23	82,49	5,5	39,87
9	11,4	93,89	5,59	45,46
10	11,54	105,43	5,66	51,12

Tab. 3-8 Pu+ Minor actinides production in kg for 45GWd for two different fuels

The Tab. 3-8 shows the cumulated production of minor actinides and plutonium. If the plutonium produced in the spent fuel is not recycled, it is more relevant to compare a thorium based fuel with a uranium fuel in this way. The cumulative Pu+MA production of 10 cycles show a 52% reduction for the fuel containing 73% Th in comparison to the classical uranium fuel, which compared to the 25% reduction of only MA production, is another argument for the thorium fuel if the plutonium is not recycled.

3.5 The U-232 issue

One very important aspect of a thorium cycle is the production of U-232 because of its radiation hazard. There are two main ways to form U-232:

(n,2n) reaction with U-233 or (n,2n) reaction with Pa-233+Beta decay of the formed Pa-232.

Cycle	1	2	3	4	5	6	7	8	9
U-232 (ppm)	40	118	166	196	215	224	230	236	237

Tab. 3-9 U-232 concentration in the uranium vector in the 73%Th fuel

The concentration of U-232 in the uranium vector has been calculated for the case of a fuel containing 73% Th. The concentration grows quickly in the first cycles and tends to stabilise at a concentration of 237 ppm, U-232 having a non-negligible neutron capture cross-section.

U-232 is an alpha-emitter with half life of 73.6 years, its decay product is the Pb-208. Two strong gamma emitters appear at the end of the decay chain: Tl-208 (Eg=2.6 MeV) and Bi-212 (Eg=1.8 MeV). The consequences are that the used fuel is more complicated to handle (recycling, transport, disposal…) and remote handling can be necessary. However, this fact can be claimed as a proliferation resistance factor [6]. Indeed, the U-233 can be used in a weapon as well as Pu-239 and U-235. But in the thorium cycle, U-233 breeding will automatically be associated with U-232 production. Indeed, U-233 has a critical mass of 8,4 kg, which is comparable to Pu-239, which has a critical mass of 7,5 kg [6]. The radiation coming from the U-232 decay chain would cause significant radiation doses to workers involved in nuclear weapon fabrication. According to the US authorities, the maximal allowed radiation dosis for a worker is limited to 5rem/yr [7]. As can be seen in the table 2.10, a worker would be allowed to work only 1,7 hours at 0,5 m from a sphere of 5kg of the spent fuel considered in this study even after the first cycle, only 10 hours of work would be allowed.

Material	Hours
Weapon-grade Plutonium	3800
Reactor grade Plutonium	610
U-233 containing 40 ppm U-232	10
U-233 containing 237 ppm U-232	1,7

Tab. 3-10 Unshielded working hours required to accumulate a 5 rem dose [6]

3.6 Summary

A mixed Thorium/Enriched uranium fuel cycle has been investigated. The basic calculations and assumptions made allow us to make conclusions on the potential of the thorium fuel, even if no detailed reactor calculations have been performed. The investigations show that the mixing of thorium to a classical uranium fuel, which can be seen as a replacement of a part of the U-238 present in the fuel by Th-232, can provide large savings of uranium: the

natural uranium consumption of a thorium based closed fuel cycle is 23% lower than for a uranium-based closed fuel cycle and 60% lower than for a uranium based once through cycle. However, this kind of fuel requires a closed fuel cycle to use the breeding properties of thorium. The thorium fuel has as well a potential to reduce the minor actinide production by 25% and the plutonium production by 50% compared to a uranium closed fuel cycle. The thorium fuel cycle has even good proliferation resistance properties, because of the production of U-232 and its radiation hazard, and because of the constant presence of U-238 in the fuel, which denaturates the U-233. For this reason, this closed fuel cycle requires remote handling and shielding. The main safety parameters have been investigated and show good safety characteristics of the reactor for all fuels considered.

4 Plutonium/Thorium fuel cycles

The thorium fuel mixed with enriched uranium has been studied in the last chapter. Here we want to study the possibility to mix plutonium to thorium. The plutonium would provide the fissile material needed because of the absence of fissile isotope in thorium. The burning of plutonium in thorium produces a very high concentrated U-233, which is a very valuable fissile material, but can cause proliferation concerns. There are two approaches for the Plutonium/Thorium fuel cycles: the first one is to try to use the fact that Th-232 – unlike U-238 – does not breed any plutonium, and can provide a very efficient burning of reactor grade Plutonium or excess military grade plutonium from existing stock piles. The second approach is to use the U-233 produced by the burning of plutonium in thorium to reduce the needs of uranium in a closed Uranium/Thorium fuel cycle in the same way as described in chapter 2.

4.1 Plutonium incineration

The incineration of existing military plutonium stockpile is a very important issue as the USA, Russia and other countries took the decision to further reduce their inventory of military plutonium in the future. The actual solution for this purpose is to dilute the military plutonium into uranium for the fabrication of MOX (Mixed Oxyde Fuel). After the irradiation of the MOX in a reactor, about the half of the plutonium has been consumed, and the change in its isotopic composition has denatured it because of the presence of isotopes with even mass numbers, which are less fissiles. However, the most part of the MOX fuel is composed of U-238, which produces Pu-239 by neutron capture. Thus, a part of the Pu-239 that has fissioned is replaced by new Pu-239 bred from U-238, affecting the efficiency of the plutonium incineration. Th-232 produces only U-233 in reactor; plutonium incineration should therefore be more efficient in this case than in MOX fuel.

In Th	**Military grade Pu**		**Reactor grade Pu**	
Vm/Vf	Pu cons. (kg/TWh)	Ma prod. (kg/TWh)	Pu cons. (kg/TWh)	Ma prod. (kg/TWh)
1,3	36,6	0,56	37,7	3,76
1,6	37	0,52	41,7	3,4
In U	**Military grade Pu**		**Reactor grade Pu**	
Vm/Vf	Pu cons. (kg/TWh)	Ma prod. (kg/TWh)	Pu cons. (kg/TWh)	Ma prod. (kg/TWh)
1,3	16,1	0,81	21,8	3,75
1,6	18,4	0,76	22,9	3,24

Tab. 4-1 Plutonium consumption for Pu/Th and Pu/U fuels

The Tab. 4-1 compares the efficiency of the plutonium incineration in the two cases described before: plutonium mixed in uranium and plutonium mixed in thorium. The consumption of plutonium concerns the entire plutonium vector of the fuel. The production of minor actinides (Np, Cm, Am) is presented as well. The so-called military grade plutonium has a very high content of Pu-239. Its detailed composition is given in the Tab. 4-2. Reactor grade

plutonium is the plutonium recycled from the spent fuel of a LWR using enriched uranium as fuel. Reactor grade Plutonium has a lower content in fissile Pu-239 and Pu-241, and a higher content in non fissile even isotopes (Pu-240 and Pu-242). Concerning the military grade plutonium, we can see that for the two moderation ratios considered, the incineration of plutonium is about twice more efficient when plutonium is mixed to thorium, as when it is mixed with uranium. The consumption of plutonium in the case of reactor grade shows similar performance when mixed in thorium as the military grade plutonium. However, the consumption of reactor grade plutonium is higher by 25% than the consumption of military grade plutonium when it is mixed in uranium.

The change of the isotopic composition of the military plutonium during its incineration is not affected by the use of thorium as carrier (see Table 3.2). On one hand, the Pu-239 content after irradiation is slightly lower for the Pu/Th fuel than the Pu/U fuel; on the other hand, the Pu-241 is slightly higher. The building of even isotopes in the plutonium vector is very similar for the two fuels, with around 30% of Pu-240 and 3% Pu-242. The assumption that the military plutonium has been denatured by the irradiation remains acceptable for the thorium/plutonium fuel.

The minor actinide production from the incineration of military grade plutonium is reduced by 30% when this plutonium is mixed to thorium in comparison to incineration of MOX fuel. The reactor grade plutonium is less favourable concerning the production of minor actinides, with much higher production rates, and this for the two cases (Pu/U and Pu/Th). Indeed, reactor grade plutonium has poorer neutron physics properties than military plutonium because of the presence of even isotopes. The total amount of plutonium must therefore be higher in the fuel to reach the same burnup, which leads to a higher production rate of minor actinides. The fact that thorium does not produce minor actinides is compensated by the higher needs in plutonium because of the absorption cross-section of Th-232. This partially explains why the minor actinide production is not lower for the Pu/Th fuel for reactor grade plutonium.

However, the case of incineration of reactor grade plutonium is of other interest: because of its large availability worldwide and of its large production, it is suitable as topping fuel to start a thorium closed cycle, as described in the last paragraph. Therefore, the breeding of U-233 from this start cycle will be more of interest than the plutonium consumption. Moreover, reactor grade plutonium is a much less sensitive material than military grade plutonium.

	Military Grade Pu			Reactor Grade Pu		
Isotope	**Initial composition**	End of Pu/U cycle	End of Pu/Th cycle	**Initial composition**	End of Pu/U cycle	End of Pu/Th cycle
Pu 238	**0,0**	0,2	0,2	**3,2**	3,3	3,8
Pu 239	**93,6**	54,7	49,2	**56,3**	41,1	34,8
Pu 240	**5,9**	28,1	31,0	**26,6**	31,8	35,3
Pu 241	**0,4**	14,0	16,4	**8,0**	15,5	16,7
Pu 242	**0,1**	3,0	3,2	**5,9**	8,4	9,4

Tab. 4-2 Evolution of the isotopic composition of the Pu during incineration

4.2 Production of U-233 with Reactor grade Plutonium

We consider in this part the possibility for plutonium to be burned in a thorium matrix. Plutonium shall provide the fissile material necessary to run the reactor and provide the neutrons for the fertile captures in thorium. The produced U-233 will be recycled and used as fissile resource for Th/Uranium cycles, as described in chapter 2. For this solution, reactor grade plutonium will be considered. Indeed, all this must be done at a large scale, which could not be done with military plutonium because of its difficult availability for the majority of the countries having a nuclear industry. In addition to this, military plutonium will not be available a long time, if it were used at a large scale.

4.2.1 Plutonium needs

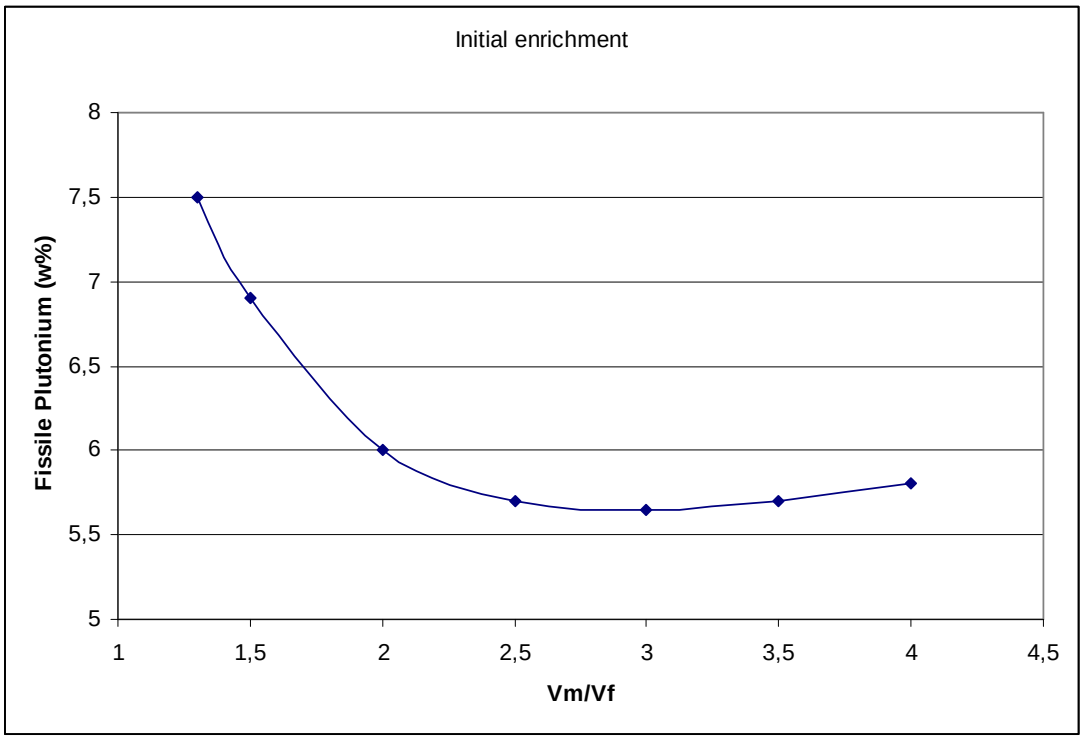

fig. 4-1 Plutonium content in the fuel at the BOC for different moderation ratios. EOC Burnup= 45MWd/tHM

Cycles with different moderation ratios have been simulated, with a common burnup at the end of cycle of 45 MWd/tHM. This means that the k_∞ is equal to 1,03 at 45 MWd/tHM. Each different moderation ratio implies a particular loss of reactivity along the cycle. Therefore, the quantity of plutonium that is present in the fuel at the beginning of the cycle determines the attainable burnup. If we fix a value for the burnup, each moderation ratio will determine a different plutonium content in the fuel at the BOC. fig. 4-1 shows the moderation ratio dependence of the needed quantity of Plutonium at the beginning of the cycle. The graphe shows an optimum of moderation for Vm/Vf=3: in this case only 5,5% of fissile plutonium is

needed. The less favourable case simulated (Vm/Vf=1.3) needs 7,5% of fissile plutonium isootopes at the beginning of the cycle. This is an important difference, but the needs of plutonium is not the most important parameter, and the real moderation ratios could be those from classical LWRs (1,3-2,0).

4.2.2 U-233 breeding

Vm/Vf	1,3	1,6	2,0
U233 breeding (kg)	10,6	10,1	9,1

Tab. 4-3 U233 production in kg for one ton of fuel for 3 different moderation ratios

Tab. 4-3 gives the production of U-233 in one tonne of fuel containing thorium and plutonium after 45 GWd/tHM, which corresponds to a conventional burnup in a light water reactor. The results are presented for three moderation ratios representing no major modifications in conventional reactors. There is 15% difference between the most favourable and the less favourable case. If possible, the hardest spectrum possible should be used, but this does not change the feasibility of the cycle. The best moderation ratio in terms of U-233 breeding does not correspond to the best moderation ratio in terms of plutonium needs.

U Composition	EOC
U 232	0,2
U 233	**93,8**
U 234	5,7
U 235	0,7
U 236	0,1
U 238	0,0

Tab. 4-4 Isotopic composition of the uranium produced during a Pu/Th cycle

Tab. 4-4 shows the composition of the uranium vector from the spent fuel after a Pu/Th cycle of 45 MWd/tHM, Vm/Vf=1.6. The composition is given for a uranium vector separated around 7 years after the end of the irradiation, which means that all the Pa-233 present in the spent fuel has decayed into U-233. The concentration of U-233 is very high, which makes this uranium a very sensitive material. The IAEA considers as high enriched a uranium containing more than 20% of U-235. There are two solutions to this problem:

-Denaturate the uranium by adding U-238

-Determine if the U-232 content of the uranium is sufficient to consider it like proliferation resistant

The first solution will be considered in 4.4, as part of a closed fuel cycle with multi-recycling like in chapter 2. The U-232 concentration in the Uranium vector after irradiation of a reactor

grade Plutonium/Thorium fuel is about 100 ppm, which is very comparable to the values of the enriched Uranium/Thorium fuel cyle described in chapter 2.

4.3 Thorium/U-233 fuel

The uranium produced from a plutonium/thorium cycle as described above has a very high content of U-233 (about 94%). This uranium is a very useful resource of fissile material because of the very good properties of U-233 as fuel in a reactor. This part intends to study the possibility to use U-233 mixed with thorium as fuel.

If we want to mix directly the uranium reprocessed from the Pu/Th cycle to thorium to produce a fuel, the needs would be around 50 kg of this uranium for 1 tonne of heavy metal, for a classical burnup of 45 GWd/tHM. The breeding of U-233 has been studied in 4.2.2. The maximal U-233 production would be 10.6 kg/tHM for a moderation ratio of 1,3. If we take the case of Vm/Vf=1,6, 10.1 kg/tHM of U-233 can be produced, which means that 5 reactors would be necessary to produce enough U-233 to feed a reactor with a U-233/thorium fuel. This possibility will be envisaged, but another strategy would be to consider that the U-233 produced by one reactor Pu/Th will be used in one reactor U-233/thorium. In this case, an addition of other fissile materials will be necessary. This possibility will be analysed and compared to the uranium/thorium closed fuel cycle described in 3.3.

We want here to discuss the feasibility of a fuel containing thorium mixed with the uranium recycled from the Pu/Th fuel described in 4.2. The composition of this uranium with high U-233 content is given in Tab. 4-2. The fuel must contain enough fissile material to reach a burnup of 45GWd/tHM.

To determine the best moderation ratio for this fuel, values of the criticality at the end of cycle (after 45GWd/tHM) have simply been put together in fig. 4-2. They show that the best moderation ratio for this fuel is around Vm/Vf=1,6. If we choose this moderation ratio, the enrichment in U-233 of the fuel must be 4,6w%. The fuel would be in this case composed of 95% of thorium and 5% of the uranium recycled from a previous plutonium/thorium cycle.

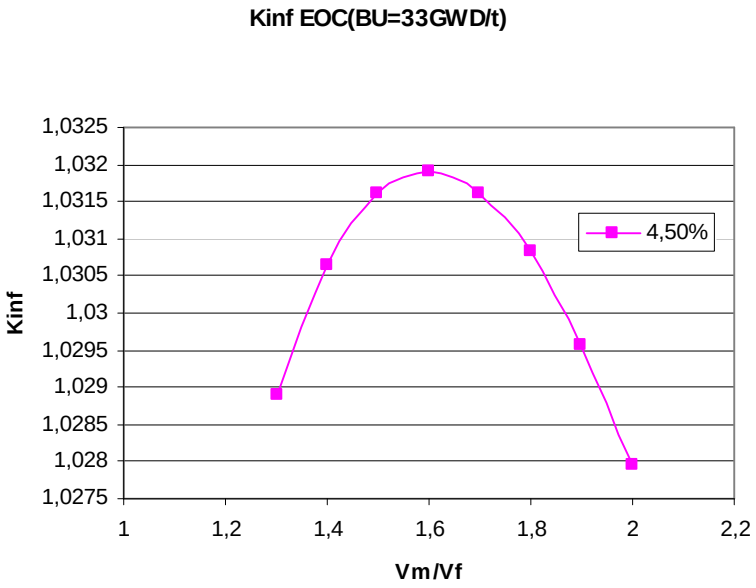

fig. 4-2 K_infinity at the EOC for different moderation ratios

The fig. 4-3 represents the values of the criticality for a cycle with a fuel containing 4,6w% of fissile U-233. The criticality at the beginning of cycle reaches k_∞ = 1.48 which is a very high value. A reactor must always have an effective reactivity coefficient of k_{eff}=1. For this purpose, burnable neutronic poisons like soluble boron (dissolved in the cooling water) or gadolinium (in the fuel) or control rods containing silber are inserted into the core, so that sufficient neutrons are absorbed, lowering the overall criticality to 1, in combinations with a neutron leakage contribution. For a typical PWR, the BOC k_∞ is around 1.3. Values of 1.5 could be too high. One solution could be to mix the U-233 with U-238, which can help to reduce the BOC k_∞, but this requires higher enrichment.

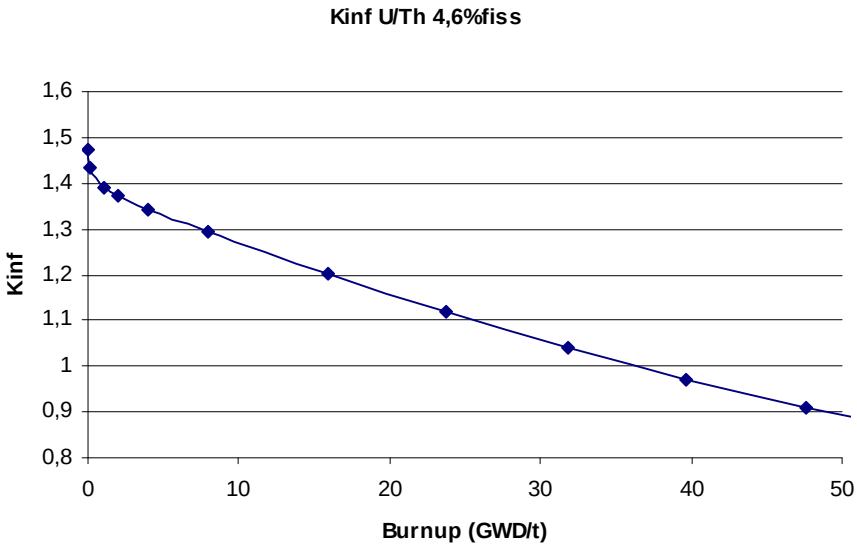

fig. 4-3 k_∞ for a thorium/uranium fuel containing 4,6% of U-233

To evaluate this effect, U-238 has been introduced in the fuel at different concentrations, in direct replacement of Th-232. All the fuels use only uranium reprocessed from Pu/Th as source of fissile material. The U-238 that is added to the fuel is supposed to be free of U-235. The results presented in fig. 4-4 show that it is possible to reduce the BOC criticality (and to reach the same burnup) by adding U-238. The BOC criticality can be reduced from 1,5 to 1,4 by adding around 25% of U-238 in the fuel.

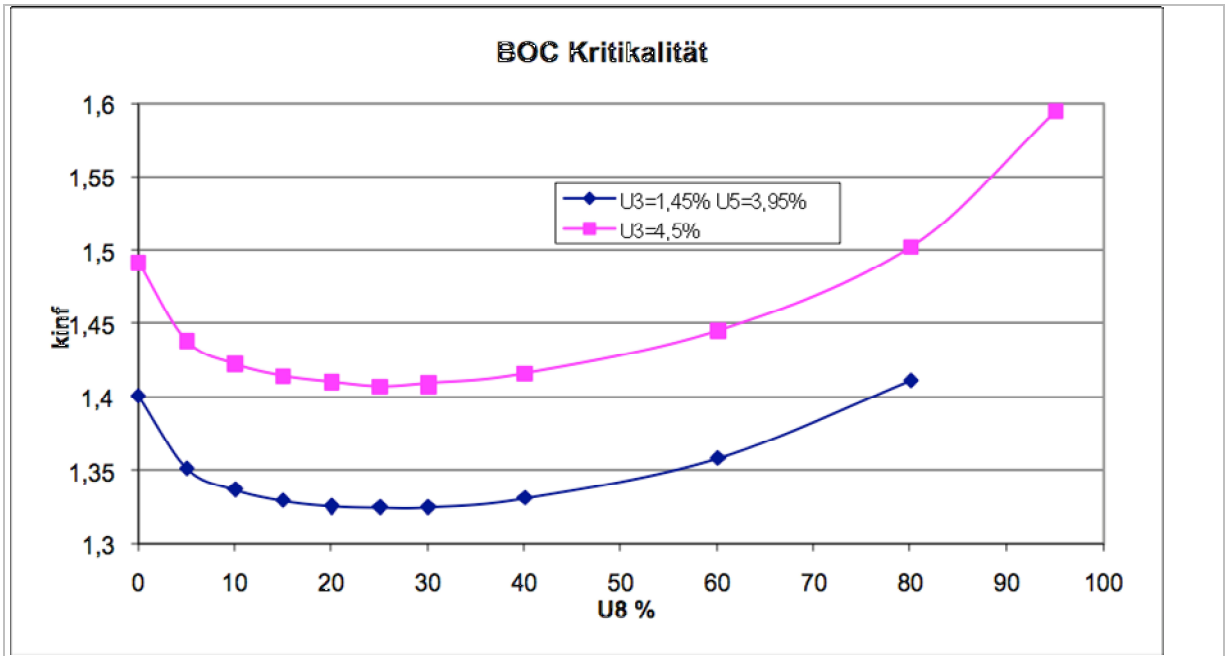

fig. 4-4 k_∞ for a thorium/uranium fuel containing 4,6% of U-233

Another solution to lower the BOC criticality is to replace a part of the U-233 by U-235, combined to the replacement of Th-232 by U-238. This is equivalent to the adding of enriched uranium to the U-233/thorium fuel. This can be seen in figure 3-4 with the blue line: the use of 1,45% of U-235 without adding of U-238 would lower the BOC criticality from 1,5 to 1,4. This criticality can go down to 1,33 with the replacement of 20% of the Th-232 by U-238.

Moderator density reactivity coefficients

When the temperature of the coolant -water in our case- changes, the pressure being constant in a Pressurized Water Reactor, the density of water decreases. Water is the coolant and the moderator. A decrease of the water density has two effects: the macroscopic absorption cross section of water decreases, and the neutrons are less moderated. The first effect has a positive contribution to the fission rate, unlike the second. Depending on the fuel (Burnup, enrichment…), one effect can dominate and the resulting Moderator density coefficient can become positive.

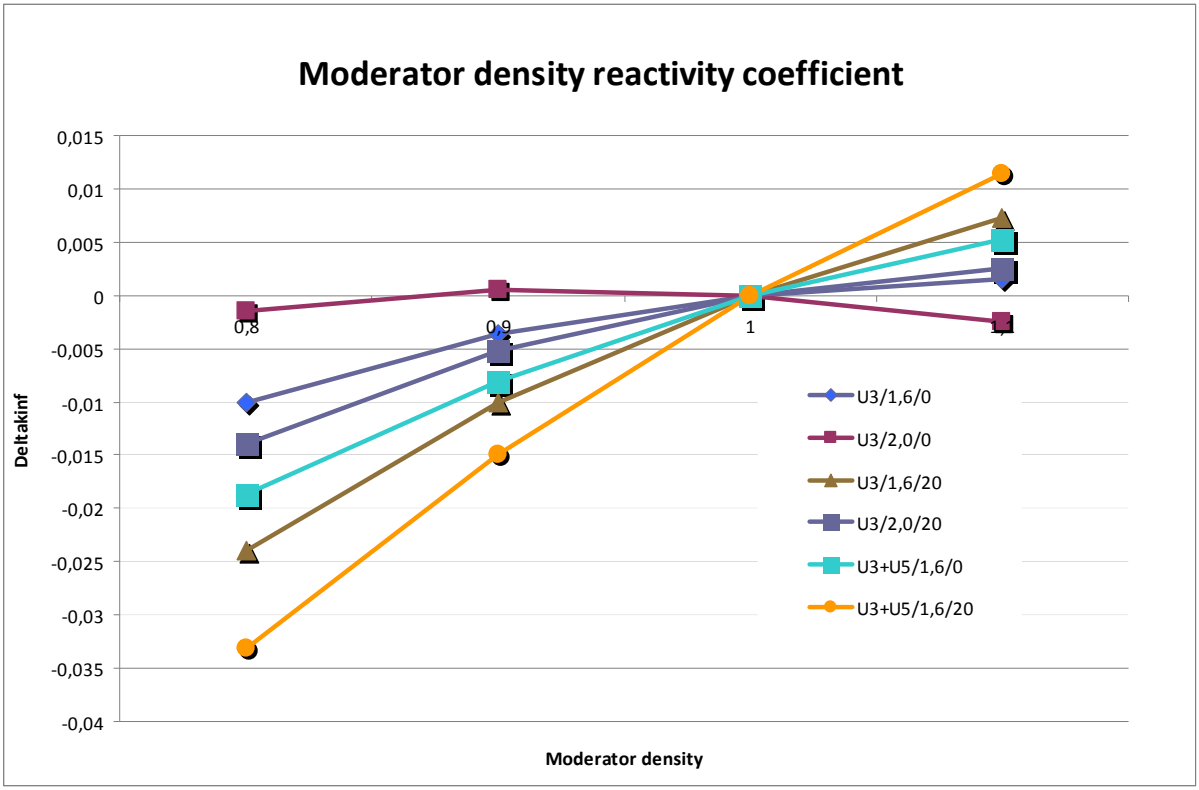

fig. 4-5 Moderator density reactivity coefficient for different fuels

The fig. 4-5 shows the moderator density coefficient for selected cases, with a legend “Fissile material/Vm_Vf/U-238 concentration. One case presents a positive MDC (moderator density coefficient): the fuel containing thorium and uranium recycled from the Pu/Th with very high U-233 content and a moderation ratio of 2,0. In all cases, the replacement of 20% of the thorium by uranium or the replacement of a part of the U-233 by U-235 implies a better MDC. Even if calculations in whole core should give better MDC, these MDC do not show a clear negative reaction to a change in the density of the reactor. However, the use of pure thorium and almost pure U-233 in the same fuel appears clearly to be the worst choice.

For these reasons and because of the problem of the too high criticality at the beginning of the cycle for U-233/Th fuel, the second approach mentioned in the introduction of this chapter should be preferred. Indeed, in this approach, the uranium (93% U-233) produced in the first Pu/Th cycle will be used as fissile resource in a fuel containing thorium, enriched uranium, and U-233. This fissile resource can reduce the needs of natural uranium of a whole closed fuel cycle in the same way as in described in the second chapter.

4.4 Thorium/U-233/enriched Uranium closed fuel cycle

For this part, we will study a typical case, according to the results of the previous analyses, and compare it to the closed uranium/thorium fuel cycles described in 3.3. This fuel will be composed of the uranium recycled from a Pu/Th fuel cycle with Vm/Vf=1,6, containing 10,1 kg of U-233, mixed to thorium and enriched uranium. The concentration of thorium will be

73% to help the comparison with the fuel cycle studied in the second chapter. The concentration of U-238 will be 21% for the reasons mentioned in 4.3. The concentration of U-235 that must be present in the fuel will be determined by the need to achieve a burnup of 45 GWd/tHM.

TH232	73,1
TOTAL TH	73,1
U 232	0,00
U 233	1,01
U 234	0,07
U 235	4,49
U 236	0,01
U 238	21,35
TOTAL U	26,9

Tab. 4-5 Composition of the fuel considered in w%

The Tab. 4-5 shows the final composition of the fuel after mixing of the three components: uranium (93% U-233) recycled from Pu/Th cycle, thorium and enriched uranium (17,4w% U-235).

This fuel will be irradiated in a reactor for 45 GWd/tHM, the uranium will be separated from the other elements of the spent fuel after around 7 years of cooling, and remixed with thorium and enriched uranium for the fabrication of a new fuel.

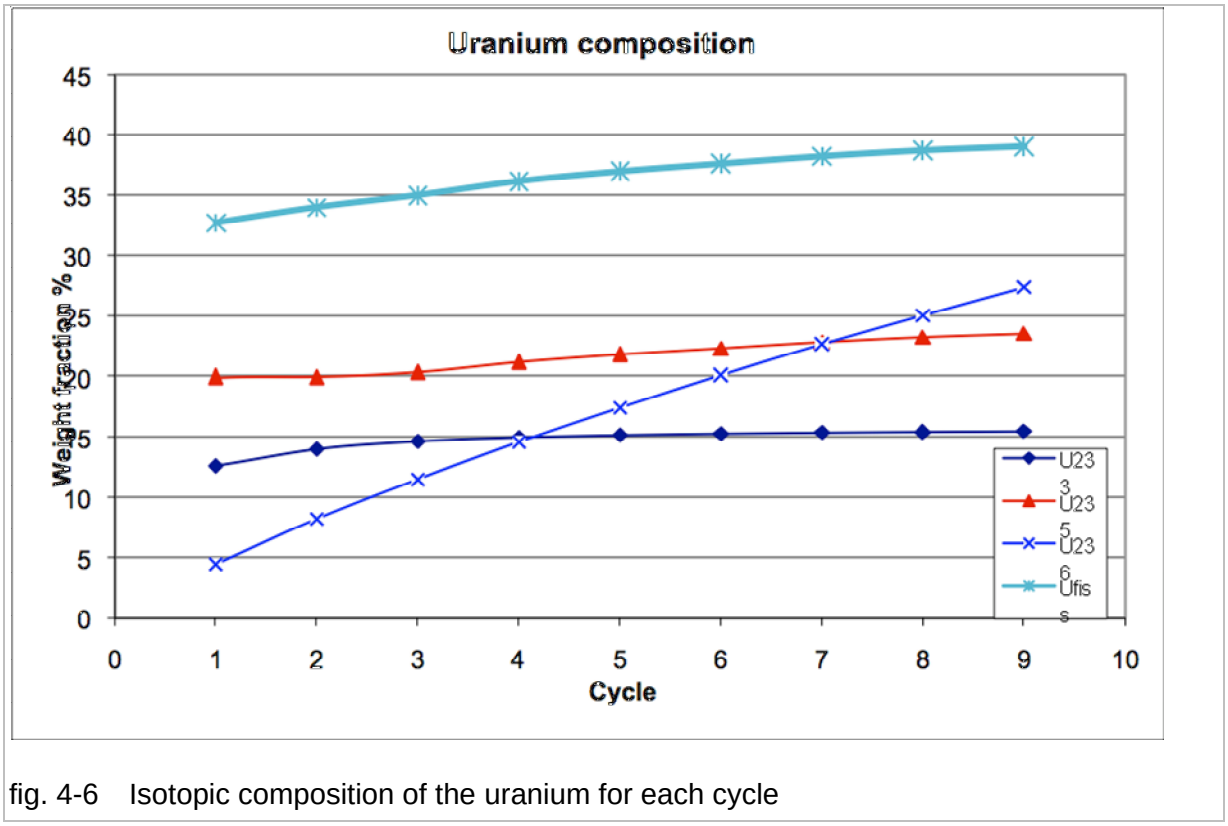

fig. 4-6 Isotopic composition of the uranium for each cycle

When we compare this fig. 4-6 to fig. 3-5, we see a very similar behavior of the fuel. The U-233 concentration does not grow so much because a high quantity of U-233 is present at the first cycle. Indeed, in the enriched uranium/thorium cycle, U-233 was not present at the beginning of the cycle. The concentration of saturation of U-233 is almost exactly the same for the two fuels, which should indicate that the performances of these two closed fuel cycles should be very similar in terms of natural uranium consumption.

In the fig. 4-7, the energy production per unit of natural uranium consumed is presented for the thorium/enriched uranium cycles presented in the chapter 2, and for the U-233/Th cycle currently considered, which is represented by the legend “73% Th+ U3 from Pu/Th”. As the cycle analysed in this part concerns a fuel with 73% thorium, we can really compare it to the fuel containing 73% thorium studied in the second part, represented by the dark blue line. The fuel “73% Th+ U3 from Pu/Th” clearly shows the best performances in terms of natural uranium consumption over all fuel studied. This is mainly due to the fact that the 10 kg of U-233 produced in the first Pu/Th do not have a “cost” in natural uranium like in the classical Th/enriched uranium closed fuel cycle, where no U-233 is present in the fuel at the beginning. However, after 9 cycles, the energy production is only 4% higher for the “73% Th+ U3 from Pu/Th” fuel in comparison to the fuel containing thorium and enriched uranium.

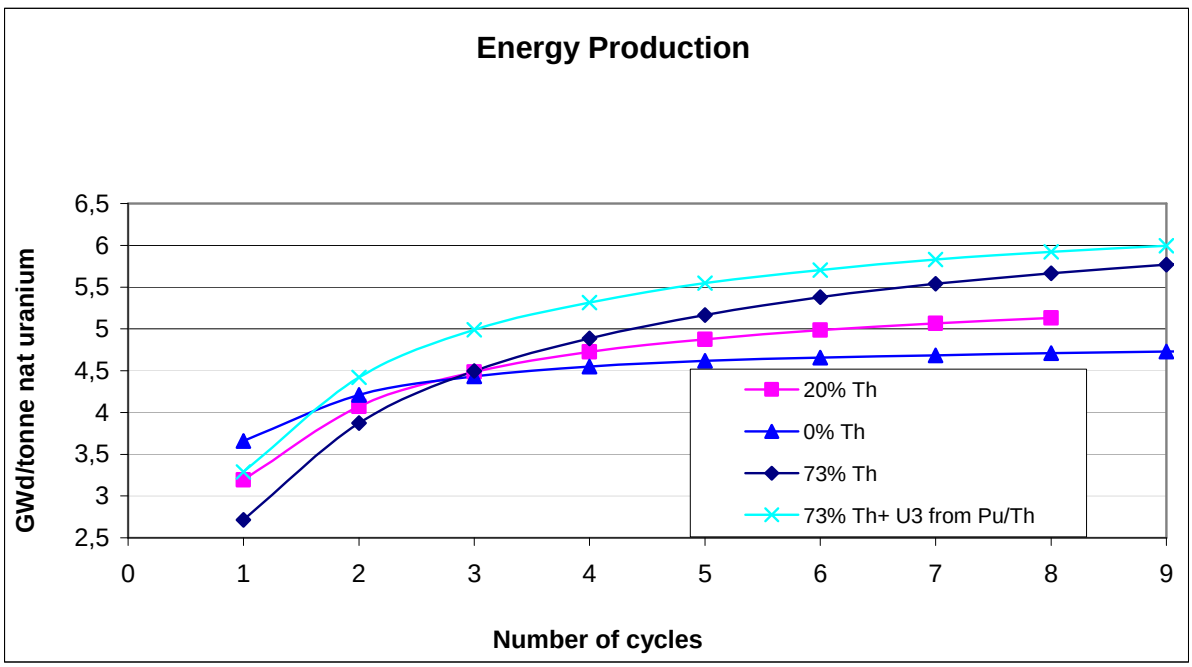

fig. 4-7 Energy production per unit of Natural Uranium consumed after each recycling for different fuels

Concerning the safety aspects, this kind of fuel cycle has exactly the same behaviour as described in the chapter 2. Indeed, the composition of the fuel is very similar, if not identical. This means very acceptable doppler coefficient, moderator density coefficient and void reactivity.

4.5 Summary

Our calculations showed that the incineration of military plutonium in a thorium matrix is twice more efficient than with an uranium matrix as far as the consumption of plutonium is concerned. The production of minor actinides is reduced by 30% at the same time. This process produces uranium containing around 93% of U-233. The U-233 can be recycled and used as a fissile resource in combination with enriched uranium in a closed fuel cycle with multi-recycling. Such a solution is very similar to the closed fuel cycles described in the second chapter (with only thorium and enriched uranium), but can present a slightly higher efficiency in terms of natural uranium consumption.

5 Modification of the scattering kernel of Th-232

5.1 Introduction

A thorium fuel cycle is based on the production of U-233 from Th-232. The production of U-233 will help to replace enrichment efforts of U-235 and save natural uranium. The efficiency of a closed thorium fuel-cycle is therefore strongly dependant on the production of U-233. This generation of U-233 is itself dependent on the neutron capture rate of Th-232, since this capture will produce Pa-233, which will decay into U-233. Therefore, this process must be very good described in the neutron physics codes used to simulate thorium based fuel cycles. The physical model describing this process in the KARBUS calculations used in this work is the common model used in most of neutron physics codes in the world (deterministic and monte carlo codes). This model has undergone improvements since many years, which can be found in references [22] and [27]. These theoretical improvements have been very recently implemented in MCNP and KAPROS as optional modules. It is of high interest to verify these improvements experimentally and to analyze the consequences on the results of the thorium fuel cycle investigations. For this purpose, a dedicated experiment was done during this work. Results of this experiment and an estimation of the changes in U-233 production due to the new scattering kernel and its consequences on the fuel cycle investigations made in this work has been performed and the results are given in this section.

5.2 Description of the scattering kernels

This section gives an overview of the two models used for the scattering kernel: the old model that was used in the thorium fuel cycles investigations and the new one. The Boltzmann equation (2.2) described in section 2.1 contains a transfer term (5.1) that describes the probability for a neutron to be scattered at a different energy:

$$\int dE' \int d^2\Omega' \Sigma_s(\vec{r}, \vec{\Omega}' \rightarrow \vec{\Omega}, E' \rightarrow E, t) f(\vec{r}, \vec{\Omega}, E, t) \qquad \text{(5.1)}$$

Transfer

The energy of the neutron after a collision is a very important parameter. It strongly influences the capture probability of this neutron, as will be explained in the following. This transfer probability is determined by the term $\Sigma_s(\vec{r}, \vec{\Omega}' \rightarrow \vec{\Omega}, E' \rightarrow E, t)$, which is called the double differential scattering kernel. The scattering cross-section is energy and temperature dependent. However, the scattering kernel which is

$$\frac{d s_s(\vec{r}, \vec{\Omega}, E, t)}{d\Omega dE}$$

is calculated in the usual codes starting from a 0K and energy independent scattering cross section $s_s(\vec{r}, \vec{\Omega}' \rightarrow \vec{\Omega}, E' \rightarrow E, t)$.

The so-called asymptotic kernel describes the secondary energy of a neutron after collision at T=0K. In this case, the classical kinetic laws are used to describe the collision. The loss of energy of a neutron scattering on a nuclide with the mass number A, with the scattering angle y is:

$$\Delta E = E\frac{2A}{(A+1)^2}(1-\cos y)$$

The maximum loss of energy occurs when y =180°, the minimum when y =0°. The corresponding energy of the neutron after the collision varies between the initial energy E and the energy a E, with $a = \left(\frac{A-1}{A+1}\right)^2$ depending only on the mass number of the collisioned nuclide.

The secondary energy of the neutron after the collision has the same probability for any energy between a E and E, if E is the initial energy of the neutron.

$$\Sigma_n(E \to E') = \begin{cases} \frac{\Sigma_n}{E} \text{ for } E' < E \\ 0 \text{ for } E' > E \end{cases} \qquad (5.2)$$

This model is used in the calculations when the target is considered to be at rest, which is the case when T=0 or when the energy of the neutron is sufficiently high so that the thermal motion of the nuclide can be neglected. For example, MCNP considers that “If the energy of the neutron is greater than 400kT and the target is not Hydrogen the velocity of the target is set to Zero” [21]. KAPROS uses the asymptotic kernel in any cases in the classical burn-up calculations.

If we now consider that the target is at a temperature T, the scattering cross section can be expressed in this way: $s(v,T) = \frac{1}{v}\iint v_r s(v_r, T=0) P(\mathbf{r}(\vec{v}) d\vec{v}$

The scattering cross section at T≠0 can be calculated based on the T=0K scattering cross section. At T≠0K, the target has a thermal motion, which must be taken into account when calculating the scattering cross section. The T=0K cross section that only depends on the relative velocity between the neutron and the target must simply be integrated over all relative velocities, taking the Boltzmann distribution into account for the velocity of the target. The Boltzmann distribution is given by:

$$P(\mathbf{r}(\vec{v}) = \left(\frac{Am}{2p\,kT}\right)^{\frac{3}{2}} \exp^{-\frac{Am\mathbf{r}_2}{2kT}}$$

To obtain the distribution of the energy of the neutron after the collision, the double differential scattering kernel must be calculated. For this calculation, the T=0 scattering cross-section is usually considered as energy independent for the calculation of the scattering kernel in most of the main reactor calculation codes. This is for example the model used by MCNPX for E<400kT. This approximation gives very good results for the secondary energy of the scattered neutrons except in the vicinity of resonances. To include resonance effects, a new model has been developed by Dagan and Rothenstein [22] and a method has been devel-

oped to implement it in MCNPX [23]. In this new model, the resonance dependant scattering cross section is used to calculate the double differential scattering kernel.

The fig. 5-1 shows the results of the simulation of the scattering kernel for the three models:

1. the “asymptotic kernel” model, which considers T=0 (blue curve)

2. the “constant cross section” model (or Wigner-Wilkins model), but temperature dependant (black curve)

3. the “energy dependant cross section” model, which is the newest model developed by Dagan and Rothenstein (red curve)

This simulation has been made for one neutron at the energy 6.52 eV, which corresponds to the lower interference dip of the 6.67eV resonance of U238.

The three models obviously give very different results. For the “constant cross section” model, the shape of the scattering kernel differs significantly from the shape of the asymptotic kernel: the energy domain of the scattered neutrons is broader, and significant amounts of neutrons are upscattered, when no neutrons are upscattered for the asymptotic kernel. The new “energy dependant cross section” model shows even more upscattered neutrons:

- compared to the resonance dependant kernel where about 30.1% of the neutrons are upscattered, 81% of the neutrons are upscattered with the new model

- compared to the asymptotic kernel where 100% of the neutrons are downscattered, less than 20% of the neutrons are downscattered

As said before, the double differential scattering kernel can have a strong influence on the absorption of neutrons. Indeed, a resonance of the scattering cross-section is always associated with a resonance in the capture cross-section. We can see in fig. 5-1 that for the case of a neutron with the energy 6.52 eV, the neutron will be significantly more scattered to the energy of the capture cross-section than in the standard case, which enhances the probability of capture for this neutron. However, this scattering kernel depends on the initial energy of the neutron, and the fig. 5-1 only gives the scattering kernel for one neutron energy. To estimate the change in the absorption, the whole neutron spectrum must be taken into account in the calculation. This change of the absorption rate due to a change in the scattering kernel will only be significant if the scattering resonance and the resonance capture are both of relatively high magnitude. Indeed, if a new scattering kernel introduces an important change in the neutron scattering because of a high scattering resonance, which will provide a lot more neutrons in the vicinity of the resonance; this will have no effect on the net neutron capture if the absorption resonance is not high as well.

To assess the consequences on criticality and fuel composition in reactor, the neutron spectrum must be calculated to estimate the overall change in neutron capture for each nuclide. In particular, a modified neutron absorption by Th-232 will have an effect on the production of U-233 and on the criticality, as well as for U-238.

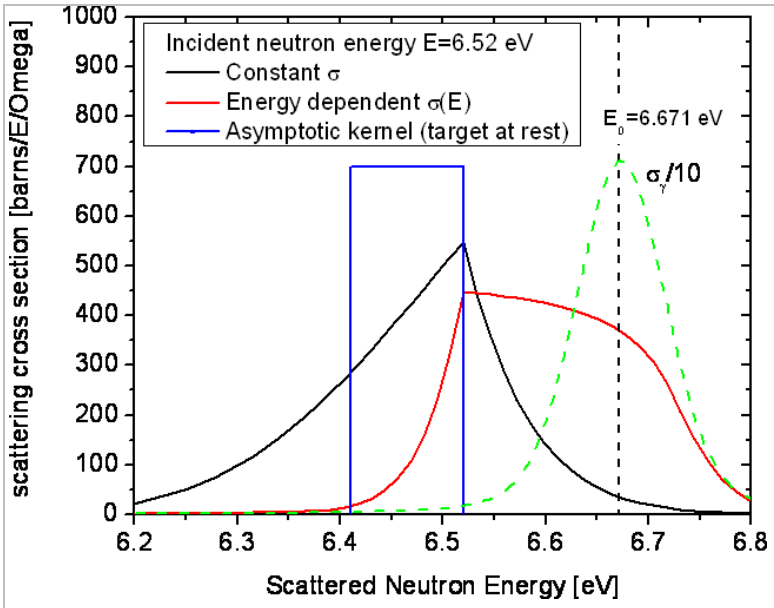

fig. 5-1 Energy dependence of the U-238 scattering kernel

The results presented in fig. 5-1 only account for the scattering kernel integrated over all the angles. The new double differential kernel developed by Dagan and Rothenstein [22] is though able to handle the angle dependence of the scattered neutron as well as the energy dependence described before.

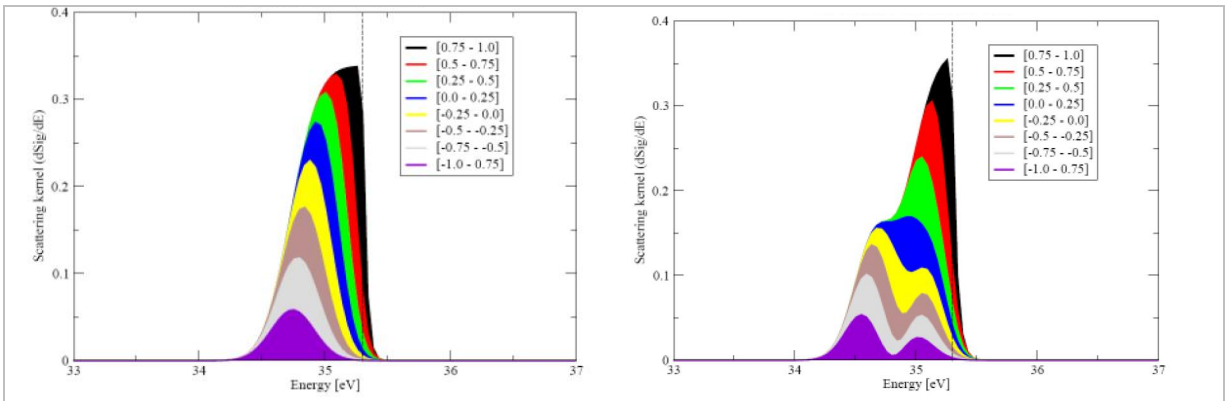

fig. 5-2 Comparison between the Standard MCNP scat. Kernel (left) and the Stochastic Double Differential scat. Kernel (right) [19]

The fig. 5-2 shows the standard MCNP scattering kernel and the resonance dependant scattering kernel of U-238 for a neutron with the energy 36,3 eV for 8 angle. Each coloured surface represents the intensity of the neutrons scattered in a defined angle interval. Results show significant differences between the standard and the new model.

If there is not much difference to see in this case for the scattering kernel if integrated over all the angles in terms of upscattering, the angle dependency and the shape of the scattering kernel for downscattering show many differences. In particular for backscattering, for example the violet surface (cosine between -1 and -0,75), where the scattering kernel of the new

model shows to distinct peaks, whereas the old model only shows one peak. Again, this scattering kernel varies with the initial energy of the scattered neutron and with the temperature of the target. The results presented here are only given for one precise neutron energy. More complete comparisons at different temperatures and neutron energies can be found in reference [19].

5.3 Benchmark experiments

5.3.1 Absorption experiment in CERN

To confirm the energy dependence of the modified scattering kernel, an experiment was conducted in CERN by F. Gunsing and E. Berthoumieux [35]. The results of this experiment can be seen in fig. 5-3. The neutron capture yield of a Th-232 sample measured by neutron time of flight ("n_TOF data" curve) is compared to a simulation of the experiment with a standard method ("MCNPX standard") and with a method including the resonance dependent scattering kernel ("MCNPX DBRC" and a modified version of "GEANT4"). The measurements show a good agreement between the two calculations using the new scattering kernel and the experiment, with a higher capture yield than for the standard MCNPX calculation for the left part of the 69,32 eV resonance. For capture yields in the right part of the resonance, the results show a poorer agreement between the new models and the experiment, but the results from the new models remain better than for the standard MCNPX calculation. This experiment partially validates the model, but the deviations for the right part of the resonance need to be explained.

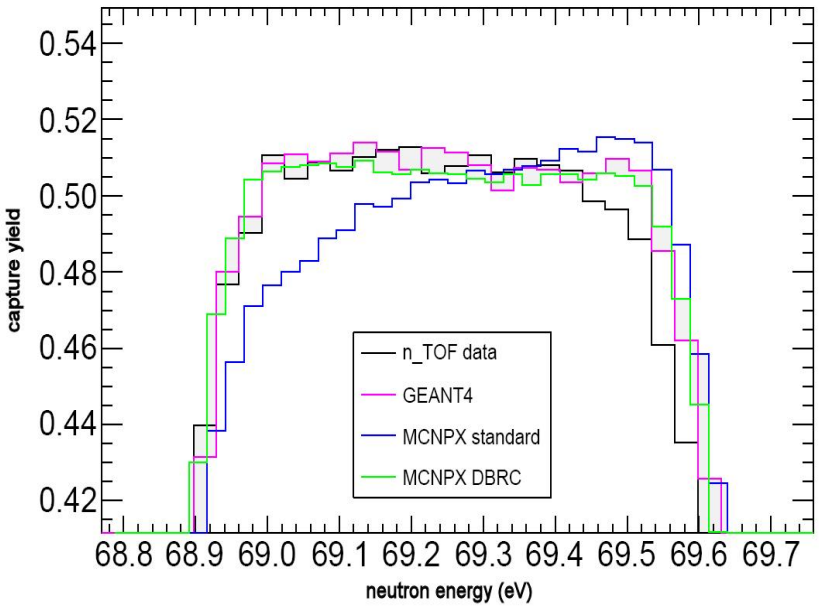

fig. 5-3 Th-232 Neutron capture experiment realised by F. Gunsing and E. Berthoumieux in CERN

The experiment presented fig. 5-3 confirmed that the new model offers better results for the simulation of the neutron capture, but the observation of neutron captures cannot fully confirm the model because the angle dependence of the scattering kernel is suppressed in such an experiment. Therefore, one needs to set up an experiment capable to measure the angle dependence of the scattering kernel to validate the new model.

5.3.2 Scattering experiments in RPI

To try to confirm the validity of the angle dependence of the new scattering kernel, a scattering experiment has been realised. The absorption experiment in CERN confirmed that the new scattering kernel allows a better description of the absorption within MCNP. However, this experiment could only measure a consequence of this new scattering kernel. This consequence is a modified absorption. In addition, it does not provide any information on the angle dependence of the kernel. In RPI (Rensselaer Polytechnic Institute,Troy, USA), an experiment was set up that allows the detection of neutrons scattered in one precise angle.

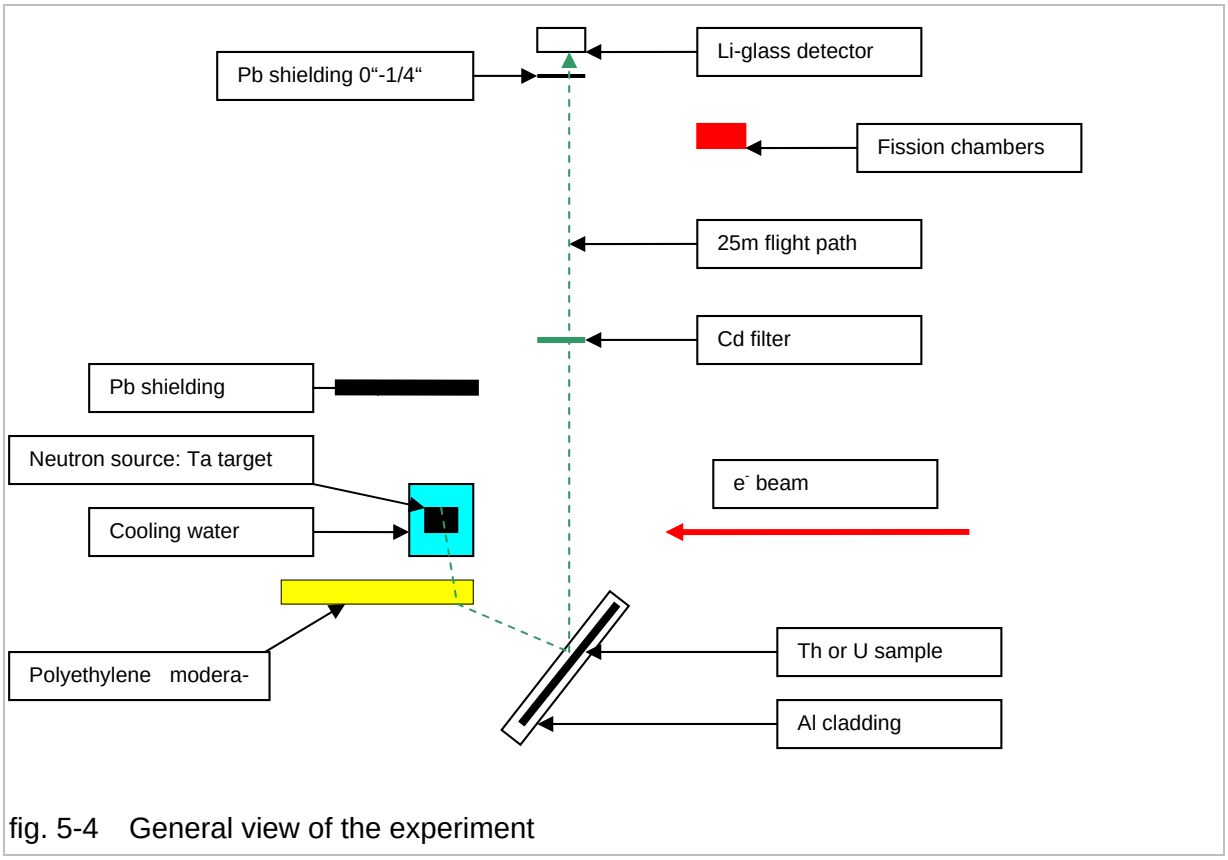

fig. 5-4 General view of the experiment

The general organization of the experiment can be seen in fig. 5-4. Neutrons are produced by the interaction between the electron beam from the accelerator and the water cooled Tantalum target. The represented polyethylene moderator can be removed and is not always present during experimentation. The thorium or uranium sample is held by a frame in a aluminium cladding. A view of this frame can be seen in in fig. 5-10. The energy of the scattered neutrons will be measured by time of flight with a Li-Glass detector after 25m of flight. The

angle of the scattered neutrons can be varied by moving the sample frame, as can be seen in fig. 5-5. The distance of 17 cm of the figure between the sample and the target can be reduced or increased, modifying the scattering angle. Fission chambers allow the measurement of the neutron flux, for monitoring purposes. The cadmium filter has two functions: it is a low energy neutron filters and it allows the time to energy conversion. Low energy neutrons must be filtered, because the pulse width being very short, they could be counted with another pulse and lead to imprecise measurements.

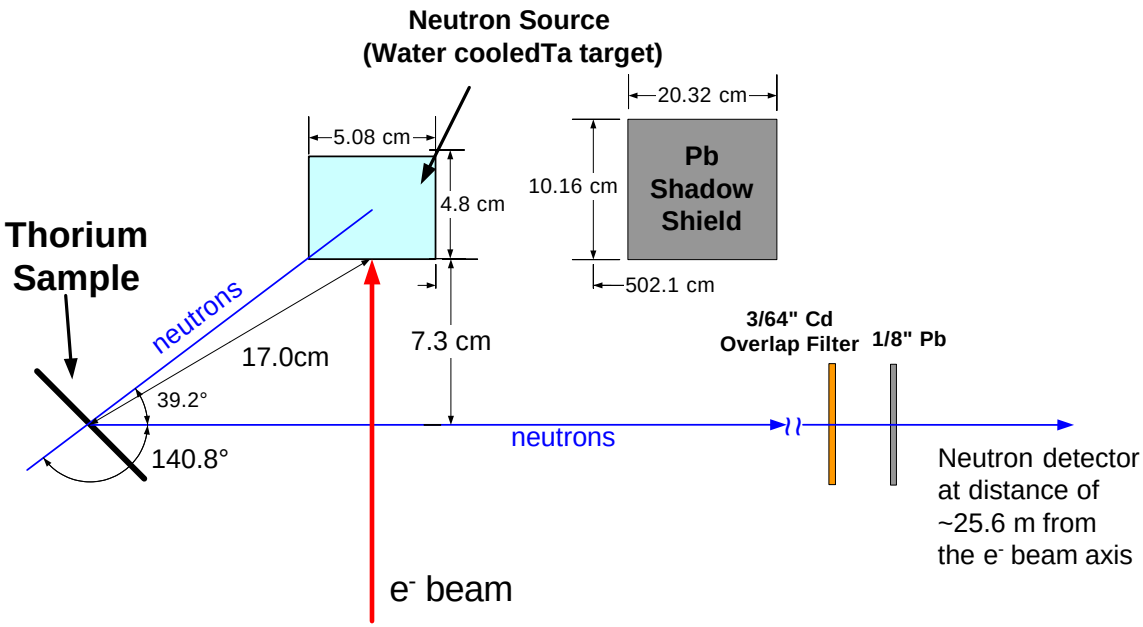

fig. 5-5 Geometry of the backscattering experiment [20]

fig. 5-5 shows the geometry of the experiment as it was set up for the neutron resonance scattering kernel measurement on Th-232 and U-238. A summary of the results will be given in section 5.3.2.2.

5.3.2.1 Neutron production

As said above, the neutrons necessary for the experiment will be produced by interaction between a high energetic electron beam and a Tantalum target. The electron beam will be provided by the linear electron accelerator the “Gaerttner LINAC. General information on this accelerator can be found in Annex C. Electrons will be accelerated in nine acceleration sections to about 60 MeV.

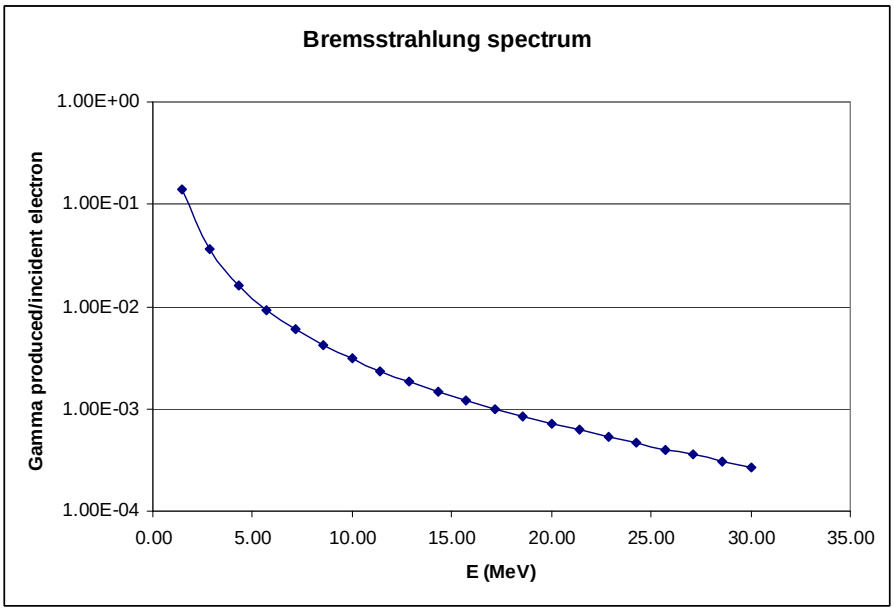

fig. 5-6 Bremsstrahlung spectrum of the Tantalum simulated with MCNPX

The high energetic electrons produced by the accelerator are slowed down in this target and produce gammas by “bremsstrahlung” effect. fig. 5-6 shows the energy distribution of the gammas produced in the Tantalum target. This bremsstrahlung spectrum was simulated with MCNPX, with the same geometry as the experimental set-up. We can observe a rise of the number of gammas emitted for energies under 5 MeV. These gammas will then produce photoneutrons. However, the (γ,n) cross-section for Tantalum has a threshold energy of 7,58 MeV as can be seen in fig. 5-7.

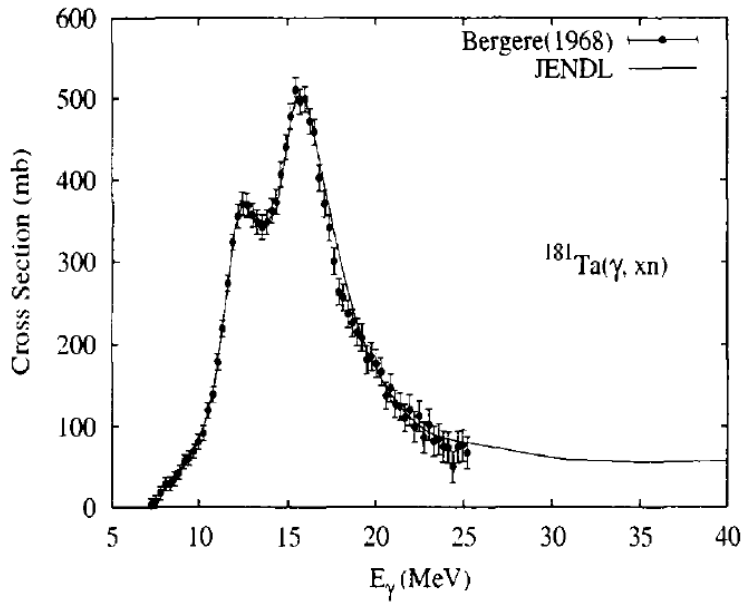

fig. 5-7 Inclusive photoneutron yield cross section for Tantalum [32]

Besides Tantalum, other elements like Tungsten or Uranium can be used for neutron production with a linear electron accelerator. These other elements generally have slightly higher

neutrons yields than tantalum. Tantalum was chosen because of its ease of use, although it has not the best neutron yield. Unlike tungsten, tantalum does not corrode in water, which allows an easy water cooling of the target. Uranium is subject to induced fission caused by the produced neutrons, which produces high radioactive fission products. For this reason, Uranium needs to be maintained in a cladding. A cladding failure would a dangerous issue, and imposes stricter safety measures.

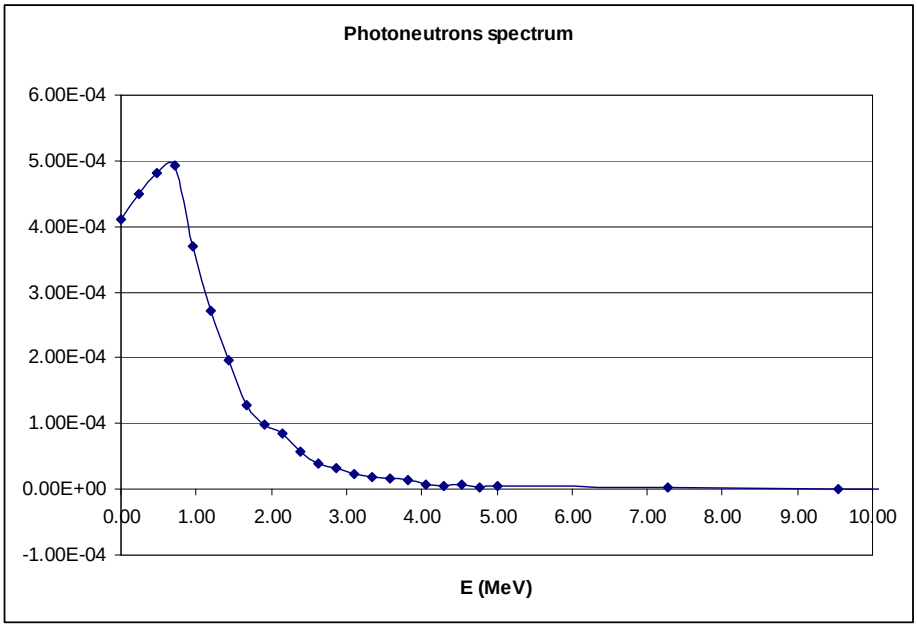

fig. 5-8 Spectrum of the photoneutrons produced in the Tantalum simulated with MCNPX

The neutron production spectrum of fig. 5-8 shows a peak of the neutron production at around 1 MeV, with a decrease in neutron production for lower energies. The resonances studied in this experiment are situated in an energy range from 1 eV to 200 eV. Therefore, a moderator will be eventually placed between the target and the sample to enhance the number of neutrons at lower energies and in particular at the relevant resonances. When a higher production rate of neutrons is needed for these energies, the neutrons coming from the target are moderated by a 2.54 cm thick piece of polyethylene, which gives a source of neutrons with a spectrum 1/E^(1.2).

5.3.2.2 U-238 experiment

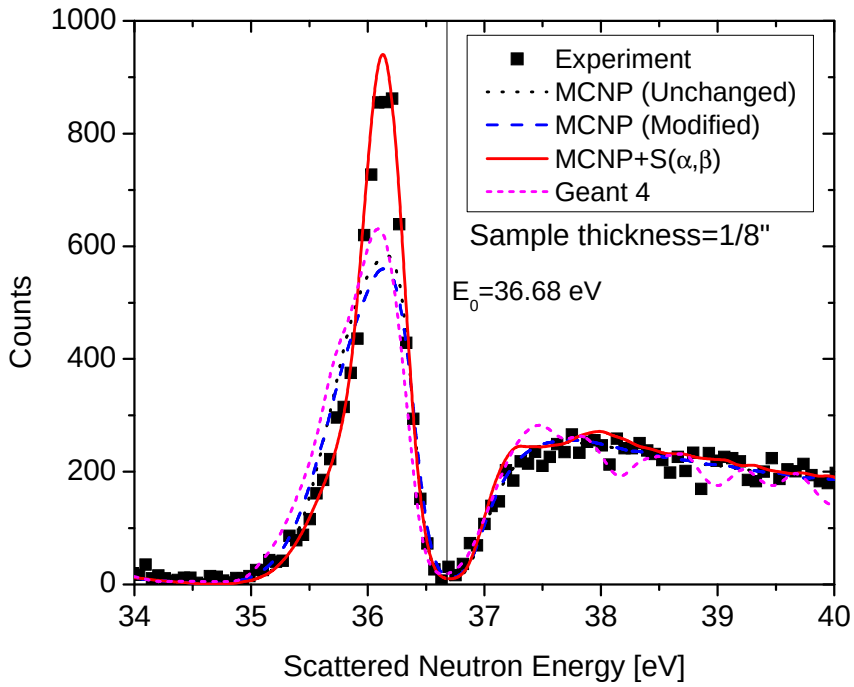

fig. 5-9 Experimental and calculated results for neutron back scattering from a depleted Uranium sample [20]

The experiment described above has been realized with a depleted Uranium sample. The scattered neutrons have been counted for back scattering (144,8°) and forward scattering (36,2°). In parallel, the experiment has been simulated with different codes, with and without the new resonance dependant scattering model. The results are plotted in fig. 5-9, for the example of the 36,68 eV resonance of U-238. For the forward angle, no significant differences have been found between experiment and the two MCNP models (old and new). The backscattering measurement shows large differences, as can be seen in fig. 5-9. The “MCNP (modified)” calculation corresponds to the removal of the restriction to E<400kT for the use of the constant cross-section model, which is called the Wigner model [25]. The results show a very good correlation between the experiment and the new model. The two old MCNP models and the GEANT 4 codes underestimate the intensity of the back scattering by a factor of 2. Despite the good results of the new models, this experiment is not sufficient to conclude on the new model. Indeed, only two angles were measured, where the full angle range should be covered. Furthermore, only U-238 has been experimented. For this purpose, a measurement of the scattering kernel of Th-232 has been conducted.

5.3.2.3 Th-232 experiment

In the frame of this work, the same experiment as described in section 5.3.2.2 with Uranium was realised with thorium. The set-up of this experiment is the same as described in section 5.3.2. Only backscattering measurements were done. The angle of the scattered neutrons

was 156°.In this particular case, the current varies between 30 µA and 40 µA depending on the pulse width. The repetition time used in the experiment was 4 ms, with a pulse width that can be varied between 150 ns and 300 ns in our case. The channel length of the neutron time of flight can be set to 128 ns or 256 ns.

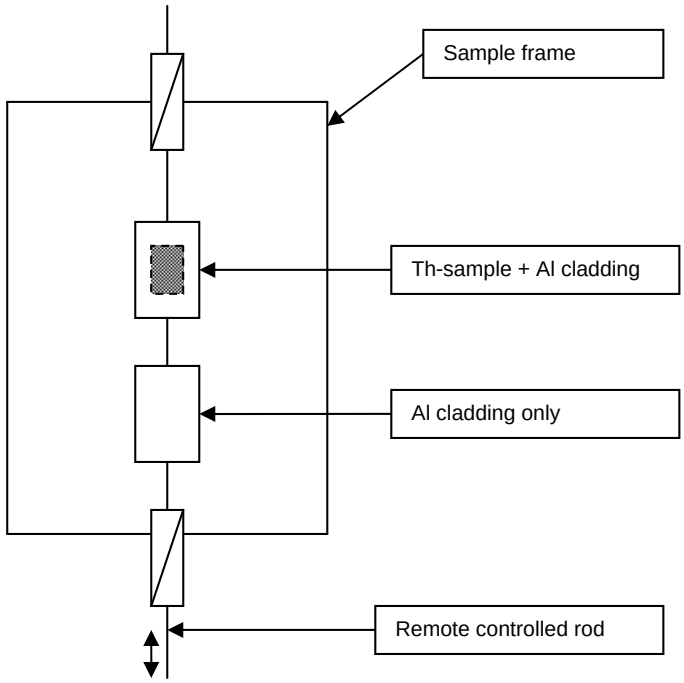

fig. 5-10 View of the sample frame

The thorium sample is held in a thin Al cladding to prevent the spread of radioactive Thorium dust as can be seen in fig. 5-10. The frame contains one Thorium sample with Aluminium cladding and one sample with the same Aluminium cladding but without Thorium. The rod holding the two samples can be controlled from outside of the accelerator room. Each sample can be alternatively replaced in front of the neutron beam. The Al sample allows the measurement of the background. Measurements were done with two different Thorium samples: one “thin sample” of 0.1524 cm thickness and one “thick sample” of 0.3048 cm thickness.

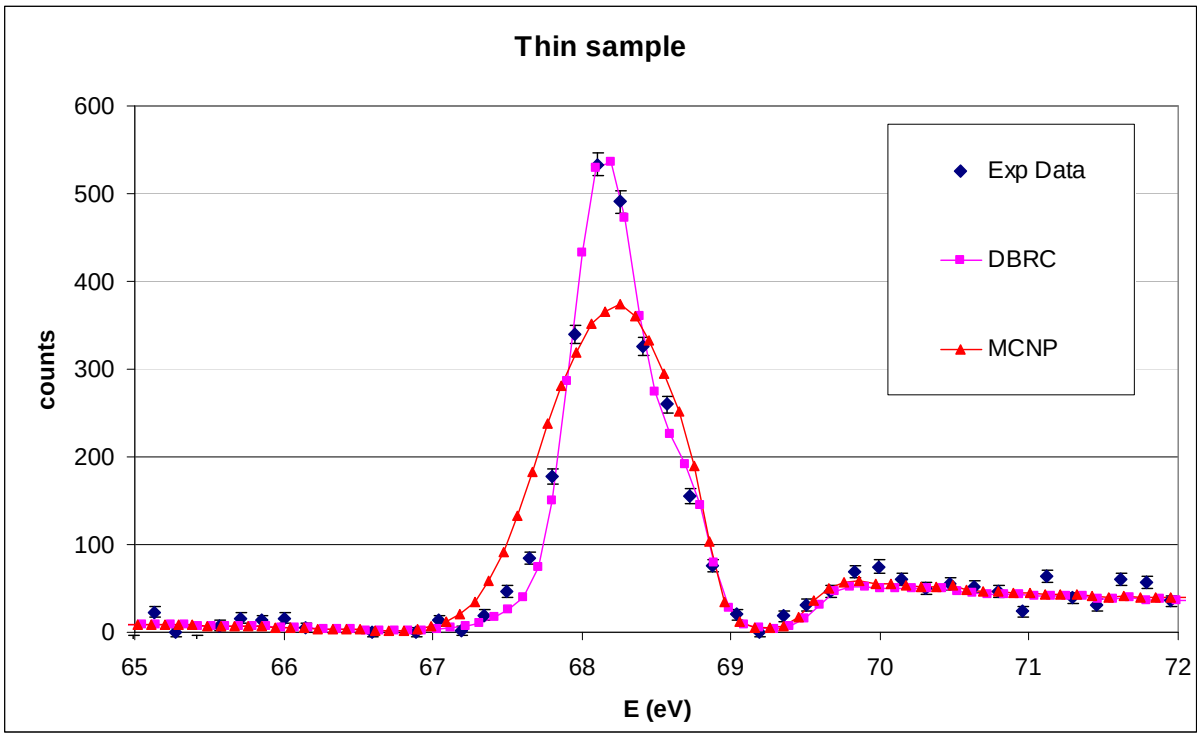

fig. 5-11 Thin sample, no moderator, 300 ns pulse width, 156° backscattering, 256 ns channel

The results for the thin sample show a clear improvement for the DBRC model compared to the classical MCNP model. However, these values are very sensible to the normalisation of the data. The classical MCNP and the modified MCNP give the same results for the right shoulder of the resonance. Therefore, the normalisation is done on this shoulder, but the statistical error in this region is quite high for the thin sample. The shapes of the resonances from the experiment and DBRC agree in general a lot better with each other than the shape of the unmodified MCNP.

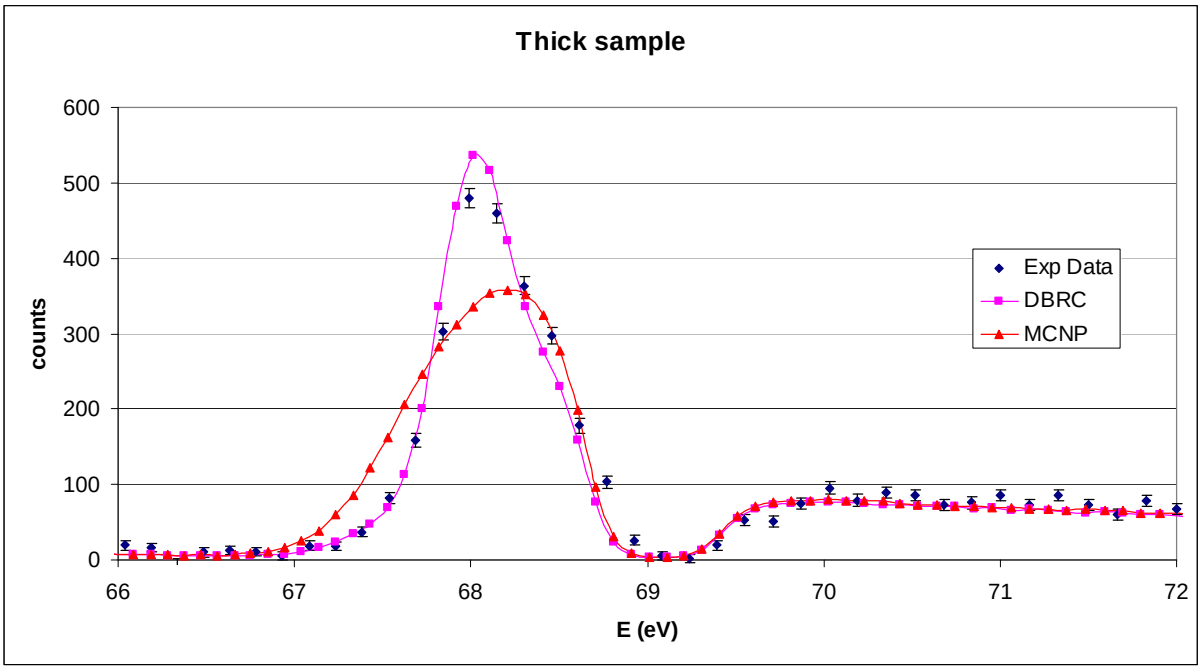

fig. 5-12 Thick sample, no moderator, 300 ns pulse width, 156° backscattering, 256 ns channel

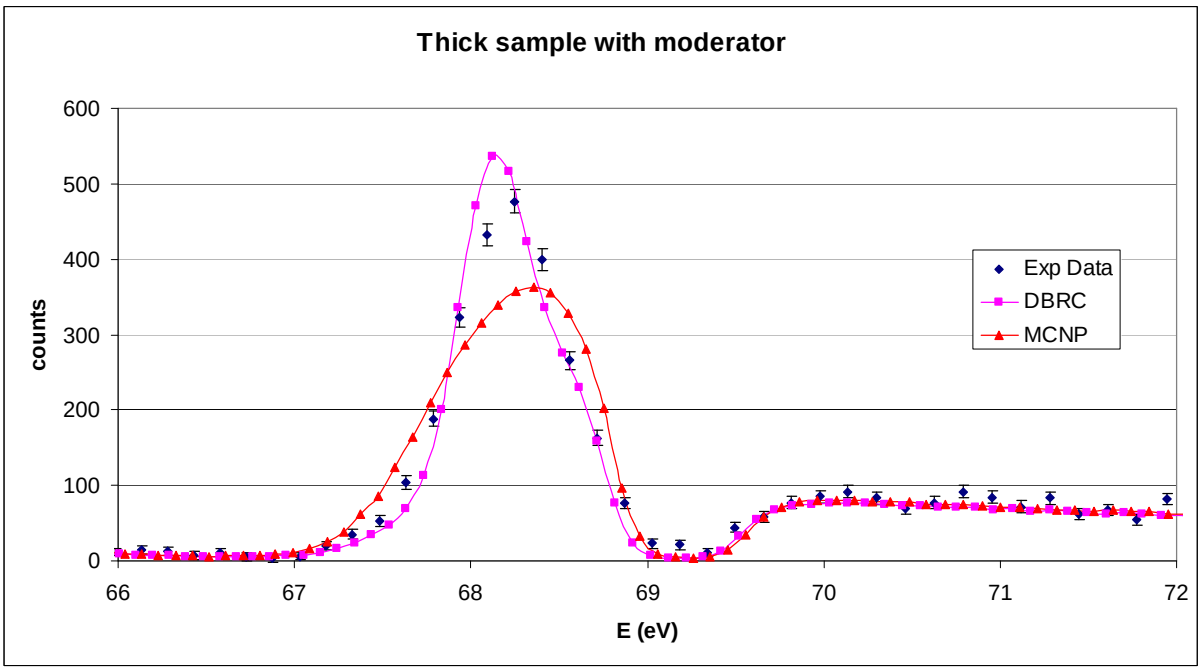

fig. 5-13 Thick sample, moderator, 300 ns pulse width, 156° backscattering, 256 ns channel

fig. 5-12 and fig. 5-13 show the results of the thick sample with and without moderator. The use of the moderator increases the number of neutrons by factor 3 in the epithermal regions and improve the statistics for the same number of runs. The use of a thick sample enhances the number of counts in the right shoulder of the resonance. These two effects make the normalisation more reliable than in the case of the thin sample.

Again, the shape of the DBRC simulation agrees clearly better with the experiment than the unmodified MCNP. At the peak, the DBRC simulation is 17% higher than the experimental data, when the unmodified MCNP simulation is 32% lower. However, it is obvious that the peak of the resonance is not well resolved by the experiment because of the large pulse width chosen as a compromise with the constraint of the radiation time. Consequently the comparison with MCNP is more sensitive to the fitting of the shape. However, to account for this issue, the channel width was indeed divided by two for another set of runs, reaching 128 ns. In parallel, the pulse width was set to 150 ns.

Further it should be mentioned that the MCNP calculation was done with a point source, which gives results for one precise backscattering angle. In the experiment, the moderator causes a spread of the angle of the incident neutrons. As explained before, the moderator improves the neutron flux. This means that non negligible amounts of neutrons arriving at the target come from slightly deviated angles due to the moderator, which is 15 cm long. Therefore, the results of the experiment do not correspond to one precise angle. However this will only call for second order effect errors.

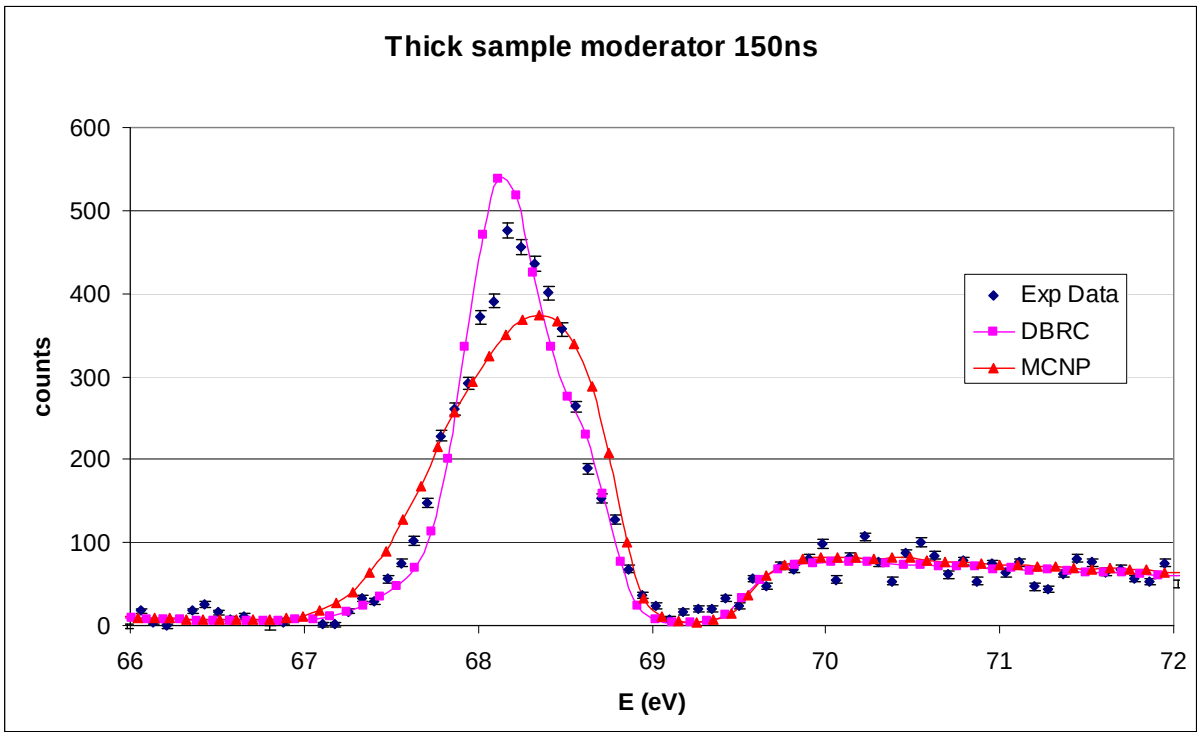

fig. 5-14 Thick sample, moderator, 150 ns pulse width, 156° backscattering, 128 ns channel

The results with a higher resolution of fig. 5-14 show once more a better agreement of the DBRC to the experimental data concerning the shape. At the peak, the DBRC is 16% higher than the experimental data. The unmodified MCNP is 26 % lower. Despite the higher resolution, the normalisation at the shoulder is difficult, because of poorer statistics compared to the lower resolution. Indeed, the division by two of the channel length and the shorter pulse width mathematically lower the counts. We can observe a deviation of the shape for the peak of the resonance. This could be observed with the lower resolution and is here confirmed. This fact remains to be explained. It can result from an approximate description of the experiment for the input of the MCNP simulation, or from a physical effect. The moderator introduces a spread of the angle of the neutrons coming to the target. This can have an influence on the results. Considering the model used for the description of the scattering kernel, which is based on the free gas model, we can suppose that it could be a solid-state physics effect too. This deviation was not observed during the U-238 experiment. A sensitivity analysis should be conducted in the next future to account for the effect of the moderator. However, one needs to have more accurate experimental results (longer irradiation time) or results with another nuclide to really conclude on this effect.

5.4 Influence of the new scattering kernel on thorium fuel cycle investigations

After the validation of the new scattering kernel for thorium, the influence of this new kernel on the results of the thorium fuel cycle investigations has been estimated. The same representative fuel as for the validation of KAPROS described in section 2.4 has been chosen. This fuel has a high thorium content of 73%, the rest being U-235 enriched uranium. This allows the estimation of the thorium specific differences implied by the new model. The new scattering kernel has a known influence on fuels containing U-238. Therefore, the highest amount possible of thorium in the fuel has been chosen in order to lower the influence of U-238 in the results with the modified scattering kernel. Indeed, the thorium fuel cycle investigations relies on a comparison between “uranium only” closed fuel cycles and “thorium/uranium” closed fuel cycles. Even if the modified model gives very different results with small amounts of thorium, this would have very little importance since it has to be compared with uranium-only closed fuel cycles with the new model.

Burnup calculations with this representative fuel have been made with the MCNPX code without and with modified scattering kernel. A comparison was also made with deterministic codes. Indeed, the resonance dependant scattering kernel was also implemented in KAN-EXT, which is the followup version of KAPROS. Results of this comparison can be found in section 5.4.2.

5.4.1 Estimation with monte carlo calculations

5.4.1.1 Impact on criticality

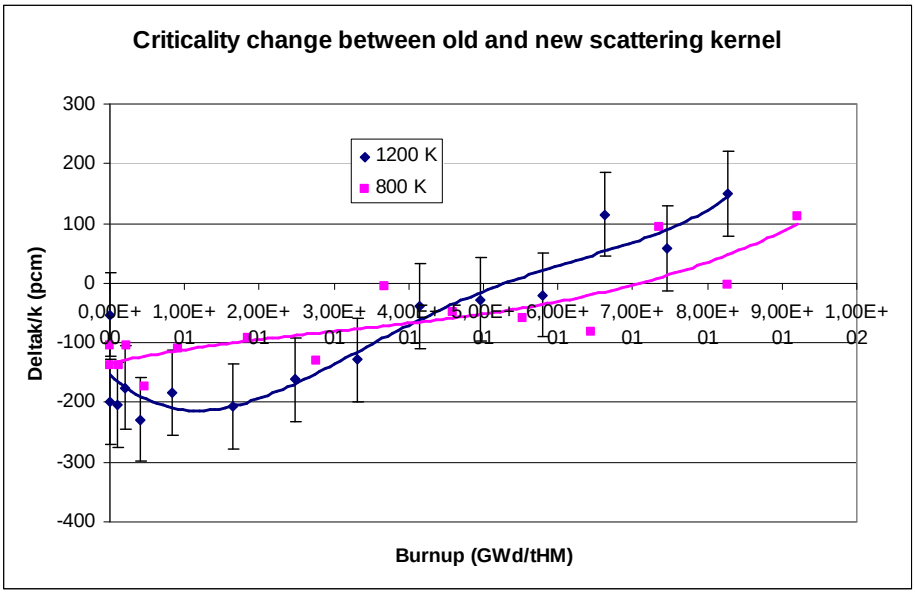

fig. 5-15 Criticality change for a 73% Th containing fuel between the old and the new scattering kernel

The results are presented in fig. 5-15 for two different fuel temperatures. For the simulation of a LWR with a fuel pin, the temperature of 800K is probably the most realistic. However, it is interesting to see the sensibility of the criticality change to the temperature. For 800K, the criticality change remains quite stable around -100 pcm for the burn-up range of 0-33 GWd/tHM corresponding to the typical irradiation time in a LWR. For the 1200K temperature, the criticality change remains between -100 pcm and -200 pcm for the same burn-up range.

5.4.1.2 Isotopic composition change

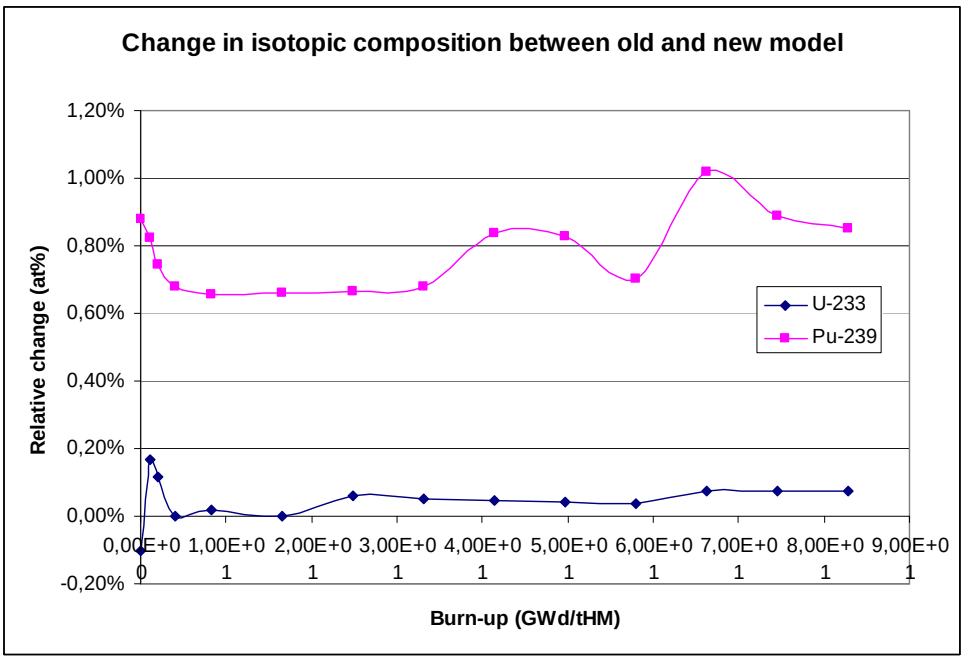

fig. 5-16 Change in the composition in U-233 and Pu-239 of the fuel for a 73% Th containing fuel between the old and the new scattering kernel

The change of concentration between the two calculations is significant for Pu-239, ranging from 0,65 to 1%. This is a consequence of the enhanced neutron absorption by U-238 with the improved scattering kernel. However, the change of U-233 concentration is negligible, around 0,1%, which means that the neutron absorption by Th-232 remains practically unchanged with the new model.

5.4.2 Estimation with deterministic calculations

The impact of the improvement of the treatment of neutron scattering in the vicinity of cross section resonances was recently investigated systematically for U-Pu fuel in a PhD work [34]. The KAPROS module ULFISP for the calculation of problem dependant fine fluxes for the group constant weighting, using the classical slowing down kernel was extended to handle the improved scattering kernel. The influence on the burnup characteristics was investigated in detail. The overall results show differences in the criticality of a few hundreds of pcm. A typical result for the influence on the collision density in the vicinity of the dominant resonance of U-238 at 36.68 eV is shown in fig. 5-17. fig. 5-18 shows this effect for the Th-232 resonance at 69.23 eV. These results are taken from the test examples in the KAPROS successor code system KANEXT.

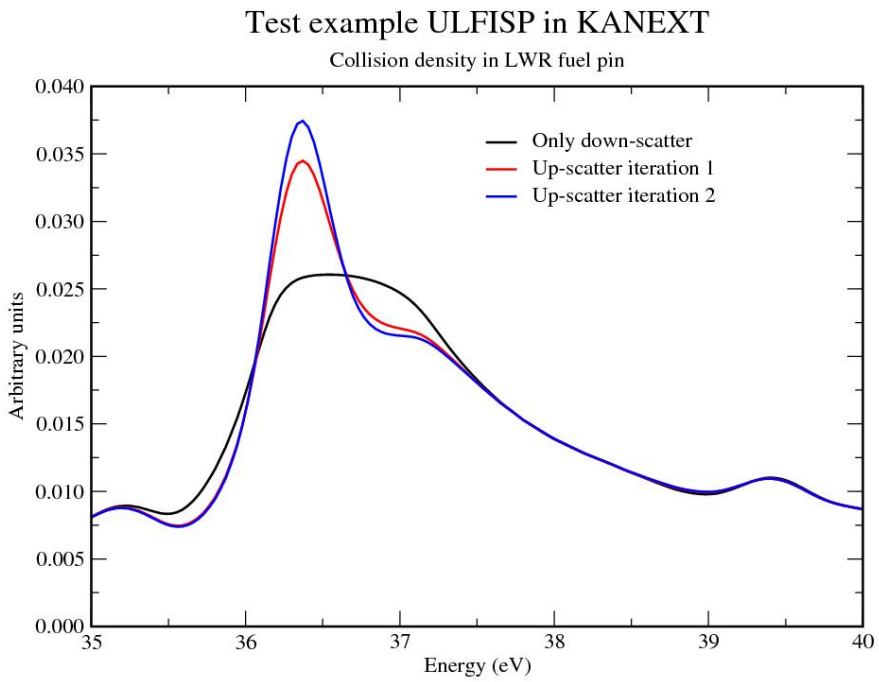

fig. 5-17 Collision densities in a LWR fuel pin containing U-238

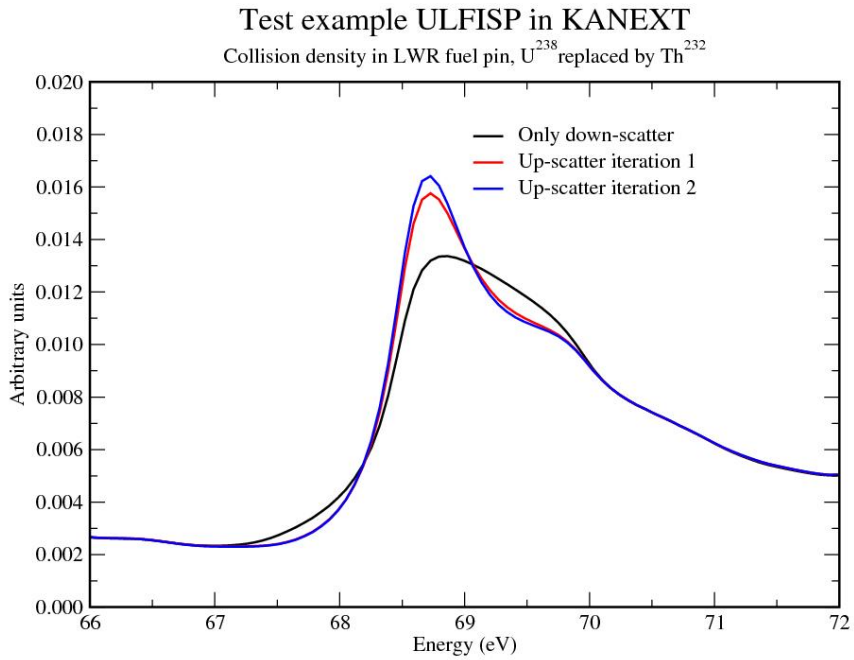

fig. 5-18 Collision densities in a LWR fuel pin containing Th-232

To assess the impact of the new scattering kernels on burnup in thorium based fuel, first preliminary burnup calculations with the procedures developed in [34] were performed for the fuel lattice with 73% thorium, as discussed in section 5.4.1. Typical influences could be observed in the weighting fine fluxes, but the impact on the integral reactivity values is small. These results are in agreement with the Monte Carlo results discussed before.

6 Conclusions

The implementation of thorium based nuclear fuels in light water reactors has been studied. It was found during this work, that an open cycle with thorium fuel has poorer performances than a classical uranium based open cycle. For best performances, thorium should be used mixed with enriched uranium in a closed fuel cycle. Numerous thorium containing fuels were studied, and the most important parameters of their optimisation have been identified. It was showed that a closed fuel cycle containing thorium mixed with high enriched uranium is feasible and is more favourable than a closed fuel cycle with only uranium, regarding many aspects. Investigations showed that it is possible to reduce the consumption of natural uranium of a reactor by 23 % with a thorium-based closed fuel cycle compared to a uranium-based closed fuel cycle, and 60% compared to a uranium-based once through cycle. The main safety coefficients of thorium fuels were estimated. Doppler coefficient, moderator density coefficient and void coefficient remain at very acceptable values, often even better than for uranium. The minor actinide production was reduced by 30% compared to a uranium closed cycle. The plutonium, which represents a large part of the radiotoxicity of the spent fuel, is 50% less produced with thorium fuel than with a uranium fuel. Calculations showed that U-232 is produced in significant amounts in a thorium fuel. Experts claim that this uranium isotope gives very good proliferation resistance properties to the spent fuel.

The possibility to use a mixed plutonium/thorium fuel has been investigated. Thorium can help to incinerate military grade plutonium with an efficiency increased by 100% compared to MOX fuel, reducing at the same time the minor actinide production by 30%. It has been shown that the incineration of reactor grade plutonium at a large scale can provide U-233, which can be reused in a closed fuel cycle very similar to the thorium/HEU fuel cycle, assuring as well 23% reduction of the natural uranium consumption.

In addition to the work on the thorium fuel cycle, a validation work of a new model for resonance neutron scattering developed at FZK has been conducted. This new physical model improves the description of the secondary energy of the neutrons after scattering for resonance energies. Applied to heavy metals like uranium or thorium, it can have important consequences on a reactor: changes in the criticality and isotopic composition of the fuel. This new model was partially experimentally confirmed in the past for U-238. As part of this work, a neutron scattering experiment was done with Th-232. Time-of-flight measurements showed that the new model agrees a lot better with the experiment. As the thorium fuel cycle investigations done in this work used the old model, investigations were done to investigate the possible consequences of the new model on the results. It was shown that the influence of this new model on thorium fuel cycles is minor for high thorium contents.

The future of the thorium fuel for light water reactors will strongly depend on the willingness to save the uranium resource. Indeed, this technology involves higher costs and is technically more complicated because of the needs to recycle the fuel. The thorium fuel will only have chances to be implemented if the decision is taken by some countries to develop large reprocessing facilities and to use closed fuel cycles at large scale. In this case, it is more competitive than uranium.

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Annex A Initial task definition

Investigations on enhanced nuclear fuel utilization in light water reactors by mixing of uranium and thorium based heavy metals.

Energy production in nuclear reactors is based on neutron-induced fission of heavy metals. Only the elements thorium and uranium are available in significant amounts. The required neutrons either may be provided by external neutron sources or by fission neutrons from isotope fission. In the early years of nuclear energy production only the fissile isotope U-235 was available as 0.7% fraction in natural uranium. Natural thorium does not contain a fissile isotope. It was found that in a heavy water moderated system with natural uranium a self-sustaining fission chain reaction is possible. In light water moderated systems uranium with U-235 enrichment (3-5%) is required. Large scale technical installations were established to perform the required U-235 enrichment. In the early stage of nuclear energy production also thorium based systems with U-235 fissile component were proposed and tested. However, up till now only an industrial nuclear fuel cycle based on uranium is practically developed. Nevertheless, broad international interest to apply thorium in nuclear energy production may be observed in the past decades. See e.g. IAEA-TECDOC-1319 “Thorium fuel utilization: Options and trends”, November 2002 and the work of Prof. Rubbia and his group for the “Energy Amplifier” around 1995.

At present, large amounts of fissile isotopes have been accumulated from nuclear weapon programs and in, partly reprocessed, discharged fuel inventories from irradiation in nuclear power reactors. Several international programs are in progress to investigate long term utilization of nuclear fission reactors, e.g. the EC projects for fast reactors and the international Generation IV (Gen-IV) activities. Most of these projects consider mainly the uranium fuel cycle.

The idea for the proposed work is based on the following assumptions and considerations:

1. Energy production in nuclear power reactors shall play an important role in the world for a long period (several 100 years).
2. For such nuclear energy production scenario a closed fuel cycle with nuclear fuel reprocessing is mandatory.
3. Thermal spectrum reactors have improved safety characteristics compared to fast spectrum systems.
4. Light water reactors (LWR) are well developed systems and undergo steady improvements.
5. Large scale nuclear fuel reprocessing is state of the art for uranium based fuel.
6. Small scale experiments for thorium based fuel were performed and the results indicate that large scale application will be feasible.
7. In FZK5784 it was shown that a closed fuel cycle with uranium is possible in LWR if (PuU)O2 mixed oxide (MOX) is used. To fulfill safety requirements (moderator density coefficient) use of up to 5% U-235 enriched uranium is required. Minor actinides have

to be handled separately in the back-end of the nuclear fuel cycle, e.g. by incineration in source driven sub-critical systems (ADS).

8. Neutron irradiation of the single thorium isotope Th-232 generates non-fissile protactinium Pa-233, decaying in about 28 days to the fissile isotope U-233.
9. The build-up of heavy metals by neutron irradiation, especially minor actinides (neptunium, plutonium, americium and curium), leads to less long term high level wastes in a thorium based cycle, compared to uranium.
10. U-233 has very good fissile properties. For proliferation considerations U-233 enrichment in uranium must not exceed 10-20%. This means that too high U-233 breeding in pure thorium must be avoided.
11. On the other hand, U-233 breeding in a mix of uranium and thorium can replace U-235 enrichment efforts in a closed combined fuel cycle.
12. The proposed work should perform a systematic evaluation of the main aspects of a long term closed fuel cycle of LWR, utilizing mixtures of uranium and thorium resources. In addition, incineration of existing plutonium stock piles from nuclear weapon programs and from existing discharged nuclear reactor fuel, together with uranium and thorium, could be considered.

The following tasks may be identified:

1. Literature study on previous related activities. IAEA-TECDOC-1319 is a good starting reference.
2. Becoming familiar with the calculation tools, especially the application of the burnup options of the KAPROS/KARBUS code in FZK/IRS
3. Analysis of world-wide thorium and uranium resources.
4. Analysis of characteristics of fission products from uranium and thorium fuel cycle. For this topic specific “physics faculty” aspects could be evaluated.
5. Parametric investigations, starting from non-published available intermediate results in IRS. Optimization with respect to aspects to be defined (proliferation, safety coefficients, discharged fuel characteristics,..)
6. Preparation of the diploma thesis and, if possible, of a common journal publication.

Tasks 1 to 3 can be performed in parallel, probably for a period of about 3 months. Task 4 must be very carefully assessed and may take some time if successful. Task 5 and 6 can be done in parallel, probably for about 3-4 months.

Dr. C.H.M. Broeders 10.9.2008

Annex B Total cross section for U-238, U-235 and Pu-239

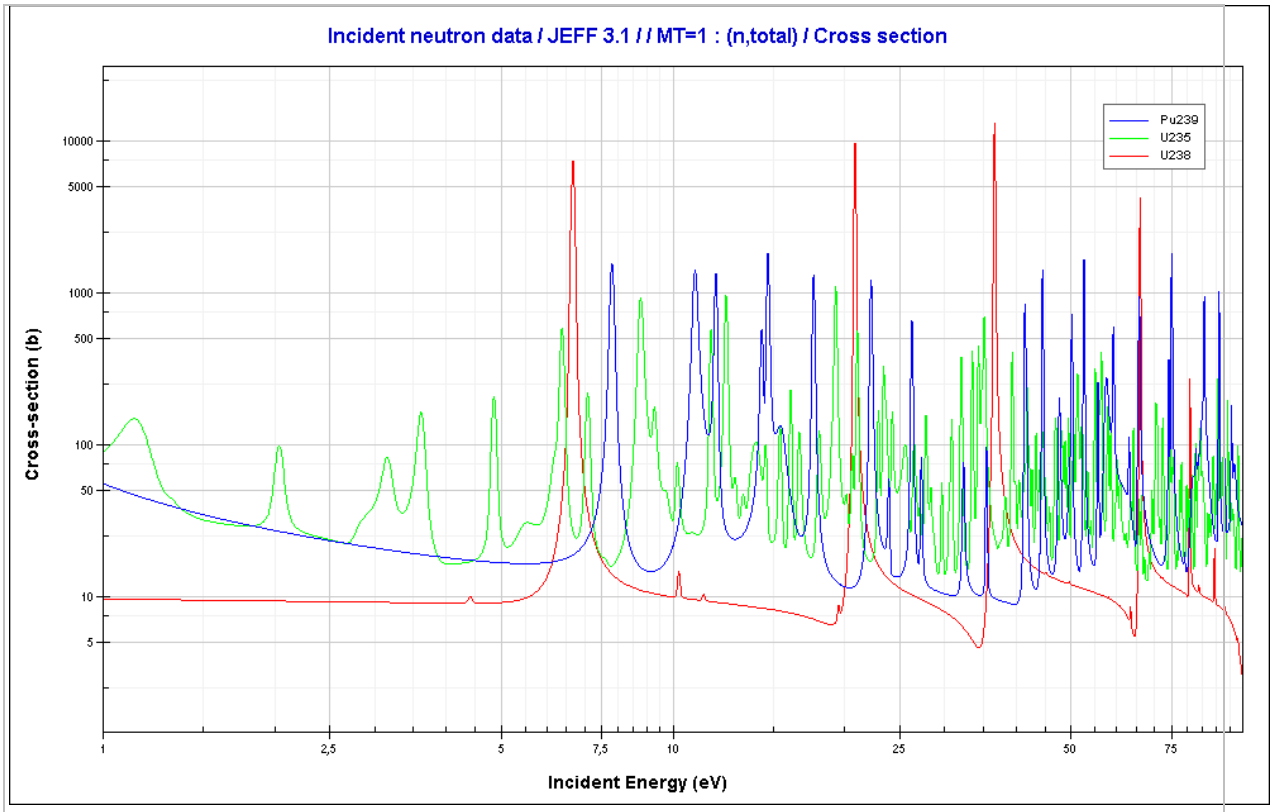

fig. B-1 Total cross section for U-238, U-235 and Pu-239

Annex C Gaerttner LINAC main characteristics

ELECTRON LINAC

Name of Linac : *Gaerttner*
Function : *Electron LINAC for Various Research Experiments (1)*
Institution and address : *Rensselaer Polytechnic Institute, Tibbits Ave., Troy, NY 12180, USA*
Person in charge : *Dr Robert C. Block, Director*
Name of person supplying these data : *Peter J. Brand*
e-mail : *brandp@rpi.edu*
tel. : *+1 518 276 6406* fax : *+1 518 276 4007*

HISTORY AND STATUS

Const. started : *1958* ; first beam : *1960*
Present status : *Operating*
Cost of facility :
Present linac staff : *4 man-years*
Present yearly operation time : *700* h

LINAC PARAMETERS

Electron Sources
Types : *Triode* ; energy : *75* keV
Beam intensity (peak) : *0 - 40* A
Normalized emittance (1σ) : π mm-mrad

Injector
Longitudinal matching :
Output : MeV; intensity : A
Pulse width, spacing :
Normalized emittance (1σ) : π mm-mrad

Acceleration System
Total linac length : *20.1* m
No. sections : *8* ; lengths : *1* m
Field mode : *π/4* ; frequency : *1.3* GHz
Wave type : *TW* ; filling time : *1.25* µs
v_g/c range : ; Q :
Shunt impedance : *12.0* MΩ/m
Iris : aperture : diameter : mm
thickness : mm
Attenuation/section : *0.36* Np
Power units, Number : *8* type : *Klystron*
RF power peak : *10* MW; mean : *15* kW

Focusing System
Type, No. of elements, and spacing :
Axial magnetic field incorporated in accelerator section

Beam Pulse Structure (If applicable)
No. of bunches/pulse :
No. of particles/bunch :
Bunch separation :

LINAC PERFORMANCE

	Normal Operation	Max, or Design	
Final energy	*(2)*	*0.090*	GeV
Accel gradient	*7.5*	*11*	MeV/m
ΔE/E (FWHM)	*10*	*10*	%
Rep. rate	*(3)*	*720*	Hz
Pulse length	*0.015 - 5*	*5*	µs
Beam intensity	*≤3*		A
Norm. emit. (1σ) :			π mm-mrad

OTHER RELEVANT INFORMATION

(1) Neutron cross-section, isotope generation, electronic testing, gemstone coloration and other research needs, as required.
(2) Two extraction ports are available. One provides energy from 5 to 25 MeV, the second from 25 to > 60 MeV.
(3) Single to 720

fig. C-1 Main characteristics of the Gaerttner LINAC [24]