Advanced fuels for thermal spectrum reactors

JITKA ZAKOVA

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Abstract

The advanced fuels investigated in this thesis comprise fuels non-conventional in their design/form (TRISO), their composition (high content of plutonium and minor actinides) or their use in a reactor type, in which they have not been used before (e.g. nitride fuel in BWR). These fuels come with a promise of improved characteristics such as safe, high temperature operation, spent fuel transmutation or fuel cycle extension, for which reasons their potential is worth assessment and investigation. Their possible use also brings about various challenges, out of which some were addressed in this thesis.

TRISO particle fuels with their superior retention abilities enable safe, high-temperature operation. Their combination with molten salt in the Advanced High Temperature Reactor (AHTR) concept moreover promises high operating temperature at low pressure, but it requires a careful selection of the cooling salt and the TRISO dimensions to achieve adequate safety characteristic, incl. a negative feedback to voiding. We show that an AHTR cooled with FLiBe may safely operate with both Pu oxide and enriched U oxide fuels.

Pu and Minor Actinides (MA) bearing fuels may be used in BWR for transmutation through multirecycling; however, the allowable amounts of Pu and MA are limited due to the degraded feedback to voiding or low reactivity. We showed that the main positive contribution to the void effect in the fuels with Pu and MA content of around 11 to 15% consist of the decreased thermal capture probability in Pu240, Pu239 and Am241 and increased fast and resonance fission probability of U238, Pu239 and Pu240. The total void worth moreover increases during multirecycling, limiting the allowable amount of MA to 2.45% in uranium-based fuels. An alternative, thorium-based fuel allows for 3.45% MA without entering the positive voiding regime at any point of the multirecycling. The increased alpha-heating associated with the use of transmutation fuels, is at level 24–31 W/kgFUEL in the uranium-based fuels and 32–37 W/kgFUEL in the thorium-based configurations. The maximum value of the neutron emission, reached in the last cycle, is 1.7·10^6 n/s/g and 2·10^6 n/s/g for uranium and for thorium-based fuels, respectively.

Replacing the standard UO₂ fuel with higher-uranium density UN or UN₃ZrO₄ fuels in BWR shows potential for an increase of the in-core fuel residence time by about 1.4 year. This implies 1.4% higher availability of the plant. With the nitride fuels, the total void worth increases and the efficiency of the control rods and burnable poison deteriorates, but no major neutronics issue has been identified. The use of nitride fuels in the BWR environment is conditioned by their stability in hot steam. Possible methods for stabilizing nitride fuels in water and steam at 300°C were suggested in a recent patent application.
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- Johan, for being my sunshine.
- My family, who have always supported me.
  Děkuji své rodič za podporu, kterou mi vždy prokázovala.
List of publications

The following articles are included in the thesis. My contribution to the work consisted of setting up the computational models, carrying out the calculations, analyzing the results and writing the texts of the articles.

Included papers


Papers not included in the thesis

The below listed publications are not included in the thesis, they were however produced during my PhD at KTH. In these articles I prepared the MCNP [1] models and carried out the MCNP calculations. MONK calculations, which were part of the article Criticality assessment for prismatic high temperature reactors by fuel stochastic Monte Carlo modeling were performed by Dr. Alberto Talamo.


II: Jitka Zakova, Alberto Talamo. Effect of neutron source on the incineration of light water reactor waste by prismatic high temperature reactor. 8th International Topical Meeting on Nuclear Applications and Utilization of Accelerators, ACCAPP’07, Pocatello, USA 2007.

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<tr>
<td>AHR</td>
<td>Aqueous Homogeneous Reactors</td>
</tr>
<tr>
<td>AHTR</td>
<td>Advanced High Temperature Reactor</td>
</tr>
<tr>
<td>AVR</td>
<td>Arbeitsgemeinschaft Versuchsreaktor</td>
</tr>
<tr>
<td>BIDO</td>
<td>Bistructural ISOtropic</td>
</tr>
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<td>BWR</td>
<td>Boiling Water Reactor</td>
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<td>CANDU</td>
<td>CANada Deuterium Uranium</td>
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<td>CERMET</td>
<td>CERamics fuels dispersed in METallic matrix</td>
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<td>DR</td>
<td>Dominance Ratio</td>
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<td>FR</td>
<td>Fast Reactor</td>
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<td>FBR</td>
<td>Fast Breeder Reactor</td>
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<td>FPD</td>
<td>Full Power Day</td>
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<td>GCR</td>
<td>Gas Cooled Reactor</td>
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<tr>
<td>HM</td>
<td>Heavy Metal</td>
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<tr>
<td>HTR</td>
<td>High Temperature Reactor</td>
</tr>
<tr>
<td>LOCA</td>
<td>LOSS of Coolant Accident</td>
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<tr>
<td>LWGR</td>
<td>Light-Water-cooled, Graphite-moderated Reactor</td>
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<td>LWR</td>
<td>Light Water Reactor</td>
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<tr>
<td>MA</td>
<td>Minor Actinides</td>
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<td>Magnox</td>
<td>Magnesium non-oxidising</td>
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<tr>
<td>MCB</td>
<td>Monte Carlo Burnup simulation code</td>
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<td>MCNP</td>
<td>Monte Carlo N-Particle Transport Code</td>
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<td>MCNPX</td>
<td>Monte Carlo N-Particle eXtended</td>
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<td>METMET</td>
<td>METallic fuels dispersed in METallic matrix</td>
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<td>MOX</td>
<td>Mixed-OXide fuels</td>
</tr>
<tr>
<td>ORNL</td>
<td>Oak Ridge National Laboratory</td>
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<tr>
<td>PHWR</td>
<td>Pressurized Heavy Water moderated and cooled Reactor</td>
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<td>PRTR</td>
<td>Plutonium Recycle Test Reactor</td>
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<tr>
<td>PWR</td>
<td>Pressurized Water Reactor</td>
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<tr>
<td>RBMK</td>
<td>Reaktor Bolshoy Moschnosti Kanalniy, eq. LWGR</td>
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<tr>
<td>SCWR</td>
<td>Supercritical Water Reactor</td>
</tr>
<tr>
<td>THTRT</td>
<td>Thorium High Temperature Reactor</td>
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<tr>
<td>TRISO</td>
<td>Tristructural-isotropic</td>
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<td>TRU</td>
<td>TRans-Uranium elements</td>
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<tr>
<td>Abbreviation</td>
<td>Description</td>
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<td>-------------</td>
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<tr>
<td>VCR</td>
<td>Void Coefficient of Reactivity</td>
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<td>VHTR</td>
<td>Very High Temperature Reactor</td>
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<tr>
<td>VVER</td>
<td>Vodo-Vodyanoi Energetichesky Reactor, eq. WWER</td>
</tr>
<tr>
<td>WWER</td>
<td>Water-Water Power Reactor, eq. VVER</td>
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Introduction

The reasons for studying feasibility and application of non-conventional fuels are multiple. These include:

- Increasing the length of the in-core fuel cycle
- Enhancing the safety
- Improving the fuel utilization
- Allowing incineration of the spent fuel

In research reported in the articles that form part of this thesis we studied several advanced fuel concepts, focusing predominantly on assessments of the feasibility of nuclear waste transmutation and fuel cycle extension. Dealing with the spent fuel without leaving an unnecessary legacy for future generations is considered important for the public acceptance of nuclear power. In this context, we investigated the feasibility of Pu and MA incineration in a Boiling Water Reactor BWR and Pu burning in an Advanced High Temperature Reactor (AHTR). The innovative AHTR concept may however offer more than just burning of Pu in thermal spectrum. The combination of the TRISO fuel and molten salt coolant enables safe, low pressure, high temperature operation, which is suitable for efficient hydrogen or process-heat production combined with electricity generation. Another non-conventional concept investigated in this work is Uranium Nitride (UN) fuel in a BWR. The use of UN leads to an increased in-core fuel residence time.

The first chapter of the thesis gives an overview over the use of advanced fuels in current as well as past designs and concepts of thermal reactors and it indicates some candidates for future fuels. The second chapter gives a theoretical background to the assessments of coefficients of reactivity, which form a central part of Papers I, III and IV. Chapter three introduces the MCNP code [1], [2], [3], provides a description of the nuclear data libraries and gives details of the methodology we used to evaluate the void and the Doppler feedbacks. Complementary to studies presented in the attached technical papers, chapter four covers additional topics presented, which are not part these publications. Namely it deals with the problem of source convergence in Monte Carlo calculations, it gives an estimation of the
Chapter five is a summary of the included papers.
Chapter 1

Overview of advanced fuels in thermal reactors

Since the first nuclear reactor reached criticality in 1942 in Chicago, many reactor designs have been suggested, built and operated and many fuels have been tested and utilized. This chapter summarizes some of the fuels that were used in various types of thermal reactors at some point in time and also fuels that are still used in current thermal reactors. It also indicates possible candidates aimed at deployment in either the existing reactors or in advanced reactors of next generations. While some of these advanced fuels were tested in past, some others are quite innovative and they have been investigated only at a theoretical level. An example of such highly innovative fuels are transmutation fuels with plutonium and Minor Actinides (MA), which were studied in Papers II and IV.

1.1 Fuels in current thermal reactors

1.1.1 Power reactors

According to IAEA [4], nearly 100% of the currently operating nuclear power reactors are thermal reactors. Figure 1.1 shows the shares of power reactor types on nuclear energy production. The largest share (61%) is represented by Pressurized light Water moderated and cooled Reactors (PWR), followed by Boiling light Water cooled and moderated Reactors (BWR) (21%) and Pressurized Heavy Water moderated and cooled Reactors (PHWR) (10%). Light Water cooled, Graphite moderated Reactors (LWGR) and Gas Cooled, graphite moderated Reactors (GCR) present together 7% of the reactor park [5]. Fast Reactors (FR) make up less than 1% of the installed capacity. Most of the thermal power reactors use enriched or natural uranium in the form of uranium dioxide pellets in a once-through cycle. For the purpose of this thesis we may call the UO\(_2\) fuel for conventional fuel and all other fuels for non-conventional. The non-conventional fuels that are used in
current power plants include fuels, that are non-conventional by their form - like metallic or composite fuels, as well as fuels non-conventional by content of unusual fissile of fertile isotopes - like Pu and Th.

![Diagram of power reactor types in 2011](image)

**Figure 1.1.** Power reactor types in 2011 [4]. Fast Reactors (FR) present less than 1%.

Natural uranium in metallic form encased in a magnesium alloy cladding [6] as shown in figure 1.2 is used in some of the GCR operated in Great Britain and North Korea [7],[8]. These reactors are called Magnox, after the magnesium non-oxidizing alloy used for the cladding. Magnox consists mostly of Mg (99%) and Al [9] which feature low neutron capture cross-sections. The promise of a good neutron economy was originally the reason for deployment of Magnox, because it eliminates the need for enrichment. A certain disadvantage of this fuel is the temperature limit imposed by the deformation of the uranium crystal lattice which occurs at 660 °C. Moreover, the maximum tolerable temperature of the Magnox cladding is 420 °C [10]. These limitations leads to low operational temperature, pressure and power density, which translate into a less economical operation.

Metal alloy fuels are not used in any other thermal power reactors. However, because they have some attractive properties, for instance high thermal conductivity, there has been some work done to address the low melting temperatures and swelling under irradiation in order to make metallic fuels further deployable. Scientists in the sixties of the last century started to look into “dilution” of these fuels by some convenient inert matrix material [11], [12], [13], [14]. Currently, active research on use of these composite metallic fuels in thermal Light Water Reactors (LWRs) is ongoing in Russia and the feasibility of the METMET concept- dispersed METallic fuel in a METallic matrix, has been demonstrated [15].
Another type of composite fuels for LWRs employ the metal alloy only as the matrix material. So-called CERMET fuels contain dispersed CERamics fuel embedded in METallic matrix. The purpose of this design is to increase the thermal conductivity, which is good in metals but less good in oxides. This concept is currently being successfully tested in Water-Water Power Reactors (WWERs) [15].

![Fuel elements for Magnox reactors with cooling fins to maximize the heat transfer. Reproduced with permission of [16].](image)

Figure 1.2. Fuel elements for Magnox reactors with cooling fins to maximize the heat transfer. Reproduced with permission of [16].

About 2% [17] of the oxide fuels in power reactors represent another type of non-conventional fuel. This fuel, referred to as Mixed-OXide (MOX), is non-conventional by isotopic composition as it contains both uranium and plutonium oxide. The purpose of the MOX fuel is to recover some of the energy left in the spent conventional fuel or to incinerate plutonium from nuclear weapons. Facilities for MOX fabrication from spent fuel are located in Belgium, France, United Kingdom, Russia, India and Japan [18]. While there are some countries using MOX without fabricating it (Switzerland), UK, on the other hand does not use their MOX in their domestic reactors. Some countries decided not to reprocess their spent fuel because of economic (Sweden), proliferation (USA) and environmental concerns. This is why the MOX fuels are less common than pure uranium fuels. Incineration of surplus weapons-grade Pu is planned, but not yet performed, in USA.

Another non-conventional fuel type that is being used in the current thermal power reactors is thorium. Thorium, which is about 3 times more abundant than uranium [19] may be used for breeding of U-233, an alternative fissile material to U-235. Utilization of thorium would to a large extent remove any concerns about the future of nuclear fuel supplies. Thorium fuel cycle is moreover expected to
produce less long-lived TRans-Uranium elements (TRU) as compared to uranium fuel cycle [20]. From a physics point of view, thorium dioxide exhibits higher thermal conductivity, higher melting temperature and better stability than uranium dioxide [8]. Thorium is currently employed in PHWRs in India. Even though historically the breeding has been repeatedly demonstrated, the Indian PWRs use Th mostly to flatten of their power profile and they keep the option of thorium fuel cycle open for future. Canadian pressurized heavy water reactors are called CANDU, an example of a CANDU fuel element is shown in figure 1.3.

![Fuel for CANDU reactors, reproduced with permission of AECL [21].](image)

1.1.2 Other reactors

Some other the non-conventional nuclear fuels are being used in thermal reactors for research purposes. For example, water-cooled research reactors often utilize enriched uranium-aluminum dispersion fuels in form of plates [15]. TRIGA reactors employ homogeneous mixture of uranium and zirconium hydride (UZrH), which provides a large, prompt negative reactivity feedback [22]. These fuels are not convenient for power reactors, because they cannot withstand high operational temperatures and provide a sufficient margin to failure.

Small-scale, high-temperature, gas cooled and graphite moderated reactors in China and Japan are using coated-particle fuels, which are finding their way back to the reactor fleet after their time-limited deployment in Germany. Fuel particles, in this case called TRISO, consist of enriched-uranium kernels encapsulated in several layers of graphite. TRISO particles, which usually measure less than 1 mm in diameter, are mixed with graphite forming fuel spheres (Chinese HTR-10) or pins loaded into prismatic graphite blocks (Japanese HTTR). In HTR-10 the spheres are simply poured into the core, forming a “pebble bed”, which gives the name to this type of reactors. In HTTR the core is built of prismatic blocks stacked on each other. Both the HTR-10 and HTTR projects are aimed at industrial deployment of Very High Temperature Reactors (VHTR) in the future. The great potential of VHTRs consist in high temperatures they can deliver to industrial applications or to hydrogen production. High temperatures also increase the efficiency of electricity generation [23].
A special category of nuclear reactors are marine propulsion reactors. Propulsion reactors have output up to 500 MWth, which makes them small compared to most power reactors. The special properties of propulsion reactors are their small size, high-power densities and susceptibility to motion and radiation damage. Most of these reactors are PWRs fueled with CERMET dispersed fuels, figure 1.4, [24], [25]. Some icebreakers and cargo ships utilize metallic fuels, which do however show significant swelling (up to 40%) under irradiation [15].

Figure 1.4. Cermet (Pu,Am)O$_2$–Mo pellet fabricated for test irradiation in Phenix, reproduced with permission of ITU.

1.2 Non-conventional thermal reactor fuels in history

The history of the nuclear reactors started with the first controlled, self-sustaining nuclear chain reaction ignited in the Chicago Pile-1 reactor in December 1942. Pile-1 utilized natural uranium fuel in the form of oxide and metal, graphite as a moderator and surrounding air as coolant. The core consisted of fuel lumps spaced on a cubic lattice within layers of moderating graphite.

Soon after this experiment the first PWR design, primarily aimed at deployment in nuclear submarines, followed. It was indeed the nuclear submarine Nautilus, which carried the first reactor of this kind. The fuel used in Nautilus was in the form uranium plates (4% U-235) in zirconium alloy with zirconium cladding [26]. The nuclear propulsion program was the basis of present day commercial LWR technology (both PWR and BWR) [27]. The land reactors, that evolved from their propulsion counterparts, had however different requirements to satisfy, which led to the use of different fuels. The power demands on land are not as variable as those imposed on the propulsion reactors. The required power is usually higher, while
the power density may be lower. Stability of the fuel at high burnup is strictly required. This led in the 1960s to the adoption of ceramic fuels [28] across the world. The ceramic fuels used thereafter carried uranium, plutonium and thorium.

Oxide fuels, non-conventional by use of plutonium, were used already in the early thermal reactor experiments. The development of these fuels was already back then driven by the desire to recover the fissile Pu remaining in the spent fuel and thereby to increase the fuel utilization. An example of an early reactor with MOX fuel is the heavy water cooled and moderated Plutonium Recycle Test Reactor (PRTR), which was built at the end of fifties [29], [30], [31].

With similar incentives for increased sustainability of nuclear power, scientists in sixties started to look into the possibility of using naturally abundant thorium as fuel. While pure thorium is not able to sustain a fission reaction, it can be used in presence of “driving” fissile material to produce fissile U-233 by neutron capture. In the historical thorium experiments, the driving fuel was often very highly enriched uranium (over 90%). Since a thorium-based breeding fuel cycle can be established in a thermal spectrum, there were several experiments involving thorium and thermal reactors through history. Thorium has been tested in PHWRs, HTRs, BWRs, PWRs and MSRs [17]. As an example, at the end of eighties, a light water core in a commercial power plant, Shippingport, used mixed oxides of thorium - uranium-233 and natural uranium in combination with small amounts of very highly enriched uranium in metallic form. It proved the concept by reaching a breeding ratio of 1.01 during a 1200 days of effective full-power operation [32], [33].

Fuels that are definitely non-conventional by its form and possibly even composition are molten salt fuels. While most of the nuclear fuel designs relied on solid fuels cooled by moving coolants, the Molten Salt Reactor (MSR) core consisted of fuel dissolved in coolant. The main objective of the first MSR program was to develop a compact, light reactor usable for aircraft propulsion [34], [35]. The interest in the nuclear powered airplane eventually dropped, however, the molten salt concept immediately received a new objective. The possible ongoing element extraction from the fuel salts made the MSR a very promising candidate for a breeder, which was a popular idea in the light of that times’ pessimistic estimations of fossil resources. In total, two MSR were built and operated. These experiments revealed some problems resulting from the very non-conventional form of fuel, like construction steel radiation hardening and cracking. Solutions to these problems had however later been demonstrated [34]. The MSR used NaF, ZrF$_4$, LiF and BeF salts and the fuel was in the form of UF$_4$ and ThF$_4$. The moderator was first BeO and later graphite. The MSR program was eventually abandoned after the U.S. Atomic Energy Commission decided to support a proposal of liquid metal cooled fast breeder project rather than the molten salt concept, however, nowadays it receives revived attention as a potential future system.

The concept of liquid fuel was also employed in the Aqueous Homogeneous Reactors (AHR) as the one depicted in figure 1.5. These reactors used uranium sulfate or uranium nitrate fuels mixed with water, which functioned as a moderator. Up to this day, AHR reactors are used for radioisotope production, for example the
ARGUS reactor at the Kurchatov institute in Russia [36], [37].

Organic-cooled and moderated reactors, which were suggested and operated in USA, utilized enriched uranium in form of UO₂ similar to standard LWRs. The concept of organic-cooled reactor suffered from several issues like poor heat transfer properties of the coolant, difficulties in the reactor control due to very strong negative coolant temperature feedback and last but not least, radiation-induced damage to the coolant. The problems were later addressed in the heavy water moderated, organic cooled reactor, WR-1, which ran successfully for 15 years. WR-1 used uranium carbide and U₃Si fuels [29], [39].

Another class of nuclear reactors developed in the past were space propulsion reactors, illustrated in figure 1.6. From the fifties to the seventies, 20 such reactors were built and ground-tested in USA. The fuels were often coated particles of uranium carbide or dispersion of uranium and zirconium carbide in a graphite matrix material [40]. The only American reactor launched into space was SNAP-10 with uranium-zirconium-hydride fuel and NaK as coolant. It was flown in 1965. After a short operational period it was shut down and it remains on the orbit.
Russia, on the other hand, launched over 30 reactors into space with the main aim of surveillance. For example the TOPAZ-1 system, utilizing very highly enriched $\text{UO}_2$, was launched in 1987. Both Russia and USA continued the work on space propulsion until nineties. [17], [41], [42].

![Nuclear thermal propulsion engine](image)

**Figure 1.6.** Nuclear thermal propulsion engine, reproduced with permission of [40].

The idea of fuel in form of coated micro particles that could handle very high temperatures, emerged in the late fifties [43]. The helium cooled, graphite moderated DRAGON Reactor Experiment operated in UK with uranium, thorium and plutonium oxides and carbides in coated particles. Coated particles, with a total diameter of the particle 1.1 mm, had fuel kernels of 0.4 mm radius, wrapped by a layer of inner pyrocarbon, silicon carbide and an outer pyrocarbon layer. These particles were mixed into the graphite matrix of the fuel rods and placed into the core. The history of the high temperature reactors continued with the Arbeitsgemeinschaft Versuchsreaktor (AVR), which was successfully operated in Germany for 22 years [44]. This concept introduced innovative fuel assemblies in the form of 6 cm spheres (pebbles). The graphite matrix of the spheres contained one layer pyrocarbon coated BISO particles, approximately 0.5 mm in radius. Later on, TRISO coated particles with an additional SiC layer were also tested. As fuel, uranium and thorium carbides, and oxides were used. A prototype of the next pebble bed reactor - the Thorium High Temperature Reactor (THTR), was delivering electricity to the grid from 1985 to 1989 [17]. The reason for shutting the power plant down was rather a political pressure than any operational problems. The fuel compacts consisted of a mixture of graphite and coated fuel particles with uranium and thorium oxide kernels.

In the USA, a 110 MWth experimental reactor at Peach Bottom was operated from 1967 to 1974 [17]. This reactor was using coated particles with highly enriched uranium and thorium carbides in annular graphite fuel elements. Both BISO and TRISO coated particles were tested. The first commercial scale HTR in the U.S., Fort Saint Vrain, operated from 1976 to 1989. The fuel in Fort Saint Vrain consisted of thorium-uranium carbide TRISO particles embedded in hexagonal prismatic blocks [45].
1.3 Non-conventional thermal reactor fuels in future

The future of nuclear power is based on satisfying the requirements of sustainable, safe, reliable and economical operation with minimal risks for proliferation. The sustainability requirement favors fuels that allow very high burnups and thereby efficient resource utilization. A sustainable nuclear fuel can as well be one that can carry sufficient amount of TRU elements for Pu and MA transmutation, giving the possibility of a closed fuel cycle with high utilization of uranium. Thorium-bearing fuels open the way to new fuel, U-233. The safety requirement aims at fuels with good thermal conductivities, high compatibility with the other elements present in the core, good stability at elevated temperatures, durability in transients and possibly a good ability to retain the fission products.

1.3.1 Generation IV systems

The Generation IV Roadmap [47] is a document issued by the U.S. DOE Nuclear Energy Research Advisory Committee and the Generation IV International Forum. It lists some of the most promising nuclear energy systems deployable in near future and also recommends research efforts focused on developing these systems. Three out of the six selected systems are thermal reactors and two out of these three rely...
on the use of non-conventional fuels.

- The VHTR reactor that uses fuel in form of TRISO particles, graphite as moderator and helium as the coolant.

- The Supercritical Water Reactor (SCWR) that is cooled and moderated by water, which however features higher pressures and temperatures and thereby works with increased thermal efficiency.

- The MSR with uranium and thorium fuel, moderated by graphite. The MSR concept is recently also being developed as a fast spectrum reactor.

The first Generation IV reactors are aimed at commercial deployment around the year 2030. From a technical point of view, they are successors of the advanced generation III systems that are being built now.

1.3.2 Other non-conventional concepts for future

There are of course, ongoing research efforts regarding other innovative reactors or fuels outside of the Gen IV frame. For example, uranium-based CERMET fuels for enhanced safety of commercial VVERs are being investigated in Russia [48], [49], [50]. French research on CERMET fuels in PWRs highlights the possibility of very high burnups in a once-through uranium cycle and enhanced proliferation resistance of the spent CERMET fuel. [51]. In USA, CERMET fuels are investigated for use in thorium fuel cycle [52]. The use of CERMET fuels in LWR power plants is however not entirely new, zirconium matrix fuels were at some point employed in Shippingport [53]. Composite fuels of the METMET type for LWR are being discussed in Russia, [54]. The advantages of METMET fuels, as compared to the traditional oxide fuels, consist in elevated contents of uranium, higher burnups and also lower operational temperatures of fuel.

Another interesting concept that emerged in the end of the nineties, is the use of TRISO particles in LWRs [55]. In the context of the recent loss of coolant accident in Fukushima, TRISO fuels are expected to provide higher safety in transients thanks due to their retention abilities [56], [57], [58], [59]. In China, they are also looking into a combination of TRISO fuel in pebbles, graphite as moderator and molten salt as coolant. The missions to Mars planned by NASA will likely use nuclear thermal propulsion systems derived from the NERVA program [40] rather than conventional fuels. Last but not least, scientists investigate the possibilities of plutonium and minor actinides transmutation in both thermal and fast spectrum reactors [60], [61], [62], [63].
1.4 Non-conventional thermal reactor fuels in this thesis

Paper I presents a neutronics analysis of an Advanced High Temperature Reactor (AHTR). This concept employs Pu or U oxide fuels in form of TRISO particles embedded in graphite blocks. In this concept, the heat is removed by non-fueled molten salts. The advantage of the salt coolant is the low-pressure, high-temperature operation, while the TRISO particles provide a good barrier for release of fission products at operational and accident conditions.

Paper II investigates the voiding feedback in a BWR fueled with U-Pu-Am-Cm oxide fuels. The reactivity feedback to voiding degrades in both fast and thermal reactors once the Trans-Uranium elements (TRU) are introduced into the fuel. It is of vital importance to understand the origins of the voiding feedback in order to design void-safe transmutation fuels.

Paper III is dedicated to the use of nitride fuel in BWRs. The augmented fissile density in the UN fuel as compared to UO$_2$ fuel allows for an extended in-core fuel cycle. In order to be employable in light-water environment, the nitride fuels must prove their stability in water and steam. A new method of fabrication aimed at addressing this issue is being developed. A patent application has recently been submitted [64]. While UN fuels have previously been used in fast reactors, their deployment in BWRs is rather innovative.

Paper IV analyses multirecycling of Pu, Am and Cm in BWRs. Based on the previous study of the voiding feedback, we introduce several transmutation fuels with a negative total void worth. The innovative thorium-based transmutation fuels can accommodate higher amounts of minor actinides as compared to uranium based fuels without encountering a positive feedback to voiding.

Table 1.1 summarizes the discussion on the present, past and future fuels in thermal systems.
## 14 CHAPTER 1. OVERVIEW OF ADVANCED FUELS IN THERMAL REACTORS

### Non-conventional fuels in current thermal reactors

<table>
<thead>
<tr>
<th>Reactor Type</th>
<th>Fuel Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Magnox reactors</td>
<td>Natural U in form of metal</td>
</tr>
<tr>
<td>LWR</td>
<td>METMET, CERMET</td>
</tr>
<tr>
<td>PHWR</td>
<td>Mixed-OXide (MOX)</td>
</tr>
<tr>
<td>Research LWR</td>
<td>Thorium</td>
</tr>
<tr>
<td>VHTR</td>
<td>Enriched uranium-aluminum dispersion</td>
</tr>
<tr>
<td>Marine propulsion reactors, PWR</td>
<td>Homogeneous mixture of uranium and zirconium hydride</td>
</tr>
</tbody>
</table>

### VHTR

- TRISO fuel with U and Th

### Marine propulsion reactors, PWR

- CERMET, metallic fuels, U

### Non-conventional fuels in thermal reactors in past

<table>
<thead>
<tr>
<th>Reactor Type</th>
<th>Fuel Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pile-1 graphite reactor</td>
<td>Natural uranium in oxide and metal</td>
</tr>
<tr>
<td>Pile-1 graphite reactor</td>
<td>Enriched uranium plates</td>
</tr>
<tr>
<td>Submarine PWR</td>
<td>in zirconium alloy</td>
</tr>
<tr>
<td>PTRR</td>
<td>MOX</td>
</tr>
<tr>
<td>PHWR</td>
<td>Thorium</td>
</tr>
<tr>
<td>HTR</td>
<td>TRISO, BISO, thorium</td>
</tr>
<tr>
<td>MSR</td>
<td>Thorium</td>
</tr>
<tr>
<td>PWR</td>
<td>MOX, Thorium, CERMET</td>
</tr>
<tr>
<td>BWR</td>
<td>MOX, Thorium</td>
</tr>
<tr>
<td>AHR</td>
<td>Uranium sulfate, uranium nitrate fuels mixed with water</td>
</tr>
<tr>
<td>AHR</td>
<td>Uranium sulfate, uranium nitrate fuels mixed with water</td>
</tr>
</tbody>
</table>

### Organic cooled and moderated reactors

- Uranium carbide and U₃Si fuels
- Uranium carbide coated particles
- Dispersion of U and Zr carbide in a graphite matrix
- Uranium - zirconium hydride

### Space propulsion reactors

- Uranium carbide coated particles
- Dispersion of U and Zr carbide in a graphite matrix
- Uranium - zirconium hydride

### Non-conventional fuels for future thermal reactors

<table>
<thead>
<tr>
<th>Reactor Type</th>
<th>Fuel Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>VHTR</td>
<td>TRISO</td>
</tr>
<tr>
<td>MSR</td>
<td>Molten fluoride salt, U, Th</td>
</tr>
<tr>
<td>LWR</td>
<td>TRISO</td>
</tr>
</tbody>
</table>

### Advanced fuels in this work

<table>
<thead>
<tr>
<th>Reactor Type</th>
<th>Fuel Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>AHTR</td>
<td>TRISO, U, Pu</td>
</tr>
<tr>
<td>BWR</td>
<td>Uranium nitride</td>
</tr>
<tr>
<td></td>
<td>U, Pu, MA</td>
</tr>
<tr>
<td></td>
<td>U, Th, Pu, MA</td>
</tr>
</tbody>
</table>

**Table 1.1.** Examples of current, past and future thermal reactors and their fuels.
Chapter 2

Reactivity feedbacks

A substantial part of the work presented in the included articles is related to the safety parameters, especially to the coefficients of reactivity and reactivity feedbacks in general. This chapter presents a short overview over the reactivity feedbacks relevant to the investigated systems, namely feedback to voiding, feedback to fuel and to moderator temperature change.

In general sense, a reactivity feedback describes how the reactivity of a system changes as a consequence of some phenomenon or an action. Phenomena influencing the reactivity may be for example temperature or density changes of the fuel, moderator, coolant, control rods, etc. It can as well be burnup of the fuel or accumulation of xenon. An example of an action influencing reactivity may be a Control Rod (CR) drop, CR ejection or variation of the circulation pumps speed in a BWR.

In reality, all the phenomena and their effects on the system are interconnected. For example a decrease of the speed of the circulation pumps in a BWR leads to decreased cooling of the core. Consequently, the coolant void fraction in the core increases. This causes an increase in the fuel temperature followed by an automatic drop of reactivity as a consequence of the fast-acting Doppler effect. Also, in a system with standard UO$_2$ fuel, the worsened moderation due to the water density decrease causes the thermal fission reaction rate to drop. These negative feedbacks cause the reactivity of the system to decrease. A decreased reactivity however means lesser heat generation. The lower fuel temperature and consequently lower void percentage in the core leads to a boost of the reactivity...

In the Monte-Carlo neutronics simulations it is common to investigate the reactivity feedback separately as if they were independent on each other. While this approach does not provide a complete description of the transient behavior, it is a fast method for gaining an insight into a design without the need of modeling the whole system and possibly also to obtain parameters for a transient-behavior study. For example, a large, positive reactivity feedback to coolant voiding creates a potential for problems. It is not straightforward to determine the consequences
without a full coupled neutronics-thermal hydraulics and possibly also material, simulation. It is however for instance possible to reveal which reaction rates are the ones that contribute to the voiding feedback and suggest design modifications in order to minimize or even reverse the effect.

2.1 Void coefficient, feedback to voiding

Void coefficient quantifies the change in the reactivity which occurs as a consequence of a changed density of the coolant. During the reactor operation, the change of the coolant density is often caused by thermal expansion. The vast majority of the existing thermal reactors have one liquid acting as both coolant and moderator and that is water. Graphite moderated reactors (GCR, LWGR, AHTR), on the other hand have a solid moderator - graphite, and gaseous or liquid coolant. The coolant in the graphite moderated cores may be CO$_2$ (GCR), helium (VHTR), water (LWGR also called RBMK) or molten salt (AHTR). What the response of the system on coolant voiding will be depends on several aspects. Gaseous coolants are in general transparent to neutrons and their voiding does not directly affect the neutronics of system to a larger extend. In all other systems, the voiding of the coolant brings about below – listed phenomena:

- Decreased efficiency of neutron moderation. As a consequence of the less efficient moderation the energy spectrum of the neutrons hardens. In general, depending on the fuel composition and on the presence of other absorbers, such spectral hardening can be either in favor of the fission reaction or the opposite.

- Drop of capture rates on the actual coolant. This is a consequence of the decreasing coolant density and it contributes positively to the reactivity. An absorber dissolved in the coolant - like boric acid in PWRs makes this effect stronger.

- Increased leakage from the fuel region followed by absorption on the construction materials, reflectors or leakage from the reactor. With the increasing void, the resonance capture rate in the construction materials grows slightly, contributing marginally negatively to the reactivity.

- If the core contains sufficient amount of some other strong thermal absorber, for example, if there is Hf in the cladding [65], the capture rate in such component decreases upon voiding, contributing significantly positively to the reactivity.

Figure 2.1 illustrates the case of a BWR core fueled with transmutation fuel. The reaction rate shares show, how the varying void percentage influences the distribution of the absorption (capture and fission) in fuel, coolant, construction materials and radial reflector. Note that the leakage from the entire reactor (bordered by the reactor vessel) is rather negligible.
Since the feedback to voiding is usually the first to turn positive upon transition to non-conventional fuels, we payed it special attention through all the work. In Paper I we investigated AHTR, which is a graphite moderated, molten salt cooled system. In Papers II-IV we studied water cooled and moderated BWRs. In the AHTR core, it is the combination of graphite moderator and molten salt coolant that creates a potential for positive feedback to voiding. This is because the main moderator - graphite, always remains in the core which efficiently limits the negative part of the feedback the voiding could bring about. In the BWRs, on the other hand, the moderator always voids at the same time as the coolant, since it is the same water. Current BWRs would therefore need to employ significantly modified fuels - like fuels with relatively high minor actinide contents (Papers II and IV), to produce a positive feedback to voiding.

Moreover, the feedback to voiding may be sensitive to the instantaneous void percentage, as illustrated in figure 2.2 for a BWR core fueled with transmutation fuel. If that is the case, it would be incorrect to represent the void feedback by a single void coefficient for the whole range of voids. Instead of presenting a void coefficient for each interval of void, we chose to present the feedback to voiding as a $k_{eff}$ versus the void percentage. The details of the methodology we used in investigation of the void feedback are given below.
It is worth noting that a reactor may be operated with a small positive feedback to voiding, as long as all other negative feedbacks prevail and the total feedback to increased power (temperature) remains negative. A class of thermal reactors that have demonstrated long-term, safe operation with a positive void feedback is CANDU [66].

2.2 Total void worth

The total void worth presents a limit case of voiding. It refers to a situation where all the coolant disappears from the core, as a consequence of - for example a Loss of Coolant Accident (LOCA). In such case, it is of course highly desirable that the chain reaction ceases without action of an operator. In quantitative terms this means that the total void worth of a critical reactor is required to be negative and preferably large.

It is straightforward that if the total void worth is positive, the void coefficient must be positive as well, at least in some regions of void. A negative total void worth, on the other hand, does not exclude a positive feedback to voiding in some regions of void. Figure 2.2 shows a negative feedback to voiding up to about 83% void. Further voiding however causes the $k_{\text{eff}}$ to increase. The total void worth associated with this core is negative, -4600 pcm.

While the total void worth clearly does not contain a complete information about the voiding feedback, it presents a very useful indicator of the voiding be-
behavior, which does not require more than two calculations. In Paper IV we conveniently used the total void worth to indicate the evolution of the feedback to voiding through the multirecycling and to identify the moments of the multirecycling with the highest probability of problematic voiding behavior. In these calculations, we employed an infinite assembly model. As shown in Paper II, the infinite assembly model does not take into account any leakage from the fuel region, hence it overestimates the total void worth by several thousands of pcm, providing an overly conservative result.

2.3 Doppler

The nuclear Doppler effect refers to the increased probability of absorption of neutrons on the fuel nuclei due to thermal broadening of the resonances. If the fuel temperature increases, the resonance peaks of the nuclei broaden and decrease. The total area of the resonance is conserved, however, the broadening on the higher-energy side of the peak and the consequent decrease of the flux in the region of the somewhat decreased top of the peak causes the absorption reaction rate to increase. In general, the resonance in question may be a fission or a capture resonance. In all the fuels discussed in this thesis and the included articles, the Doppler effect results in a negative reactivity feedback. This is because the fuels we employed feature large thermal capture resonances and the investigated systems not suffer from significant depletion of neutron flux in the thermal region. Quite different situation occurs in cores that feature low thermal flux. For example, in fast reactors with high Am contents the thermal resonances become so unimportant due to the suppressed thermal flux that the Doppler feedback significantly decreases [67], [68], [69]. Moreover, if the fuel contains sufficient share of nuclei with large thermal fission resonances, like Pu-239, the broadening of these resonances may turn the Doppler effect positive [67], [70]. Figure 2.3 illustrates the Doppler broadening of the large capture resonances of Pu-240 and U-238, Paper I.

In the literature, it is a habit to quantify the Doppler feedback by a Doppler constant or a Doppler coefficient. We used this representation in Papers III and IV. In Paper I, we chose to represent the Doppler feedback by a plot of \( k_{\text{eff}} \) versus fuel temperature. Section 3.3 describes the details of the methodology we employed when calculating the Doppler constant in BWRs.

2.4 Moderator temperature coefficient

The increasing temperature of the moderating environment causes the energy of the neutrons, which are in thermal equilibrium with the environment, to shift towards higher values. In water-cooled reactors there is only a limited potential for the water temperature to rise before the density of the water significantly changes. The reaction of the system to the density change- which is of major significance,
is described as the feedback to voiding. The feedback to an isolated moderator temperature change has not been investigated for water.

However, in a system with solid moderator - like the AHTR in Paper I, the situation is quite opposite. The thermal expansion of graphite is rather negligible, the temperature ranges are much larger and it therefore becomes useful to investigate the reactivity changes connected to the increased temperature of the moderator. In MCNP [1], [2], [3], this is realized by selecting appropriate library with corresponding $S(\alpha, \beta)$ treatment. The $S(\alpha, \beta)$ data replace the library data generated with free-gas models to account for the energy of the chemical bonds between the carbon atoms in the thermal region. Energies, where the effect of the bonds becomes significant and needs to be counted with are in order of several eV. Besides carbon, the $S(\alpha, \beta)$ treatment is used also in conjunction with other elements and chemical bonds such as beryllium metal or oxide, benzene, hydrogen in water or zirconium hydride, etc [2].

Figure 2.4 shows the spectral shift which is a result of the increasing graphite temperature in AHTR. The vertical lines in figure 2.4 indicate the peaks of the capture to fission ratio of the fuel. For instance, the line passing the visible dip the spectrum shown in left subplot reflects the presence of the large resonance of Pu-240. In Paper I, the legend in the figure showing these spectral shifts for the respective fuels contains a typo. The correct temperatures labels are, as in figure 2.4 below: 800, 1200, 1500 K.
Figure 2.4. Spectral shift (averaged over the whole core) that occurs as a result of increased temperature of graphite, AHTR, Paper I.
Chapter 3

Computation tools and methodology

Use of every computational method is connected with some possibilities and limitations. This chapter discusses the codes and data libraries employed in the calculations presented in the included articles as well as the methodology of the calculations developed for investigating the voiding behavior and Doppler feedback in BWRs.

3.1 Codes and data libraries

Neutronics studies form a central part in the nuclear reactor analysis. Whether standing alone or coupled to other computational modules, neutronics codes typically provide neutron flux distributions, fission power densities, reaction rates and $k_{eff}$ - the eigenvalue of the neutron balance equation. The requested physical quantities are obtained by means of solution of the neutron transport problem, which in the most general form exists in six dimensional space. Such solution for a whole reactor model presents a great computational challenge. In practice, a user must always judge carefully the level of detail relevant for the problem he or she is attempting to addresses. Based on these considerations, the user must chose appropriate method of solution, along with applicable approximations.

Essentially, there are three approaches to the solution of the neutron transport problem. Deterministic methods of solution, stochastic (Monte Carlo) methods and hybrid methods, which combine the previous two. While codes based on the deterministic approach strive to calculate a solution of the transport equation, the stochastic Monte Carlo methods obtain results by simulating individual particles behavior. From large number of such simulations, they derive average properties and required quantities.

The deterministic codes are in general challenged by the large numbers of equations they need to solve, as a result of which they must resort to various approximations. Assuming that each of the independent variables - $x$, $y$, $z$, $\Omega$ and $E$ is discretized on a grid of - say 100 points, the number of unknowns becomes $10^{12}$. Already representing a whole reactor – from the details of the fuel elements in
range of mm to the reactor vessel in range of meters on such grid calls for major simplifications. Yet, such system of equations would still be beyond the capacity of current computers. It proofs necessary to reduce the number of unknowns in the problem. A way of reduction of the number of unknowns used by deterministic codes is spatial homogenization. In this process, averaged data describing concerned region of space are generated to replace the original, detail representation. Homogenization is usually performed on several levels. A user may also opt for fewer spatial coordinates or fewer energy groups in order to simplify the problem. This however assumes that a detailed neutron spectra of the original system is known and the reduction of the number of groups does not distort the fundamental physics of the system.

Deterministic codes are often employed in coupled neutronics–thermal hydraulics codes, enabling a description of time—dependent behavior of the whole system. These so called systems codes are often suited for a specific system (SCORPIO-VVER with neutronics module MOBY-DICK used in Dukovany nuclear power plant)[71]. They are well benchmarked for their purposes and widely used within both industry and research.

Monte Carlo codes use a different philosophy; rather than solving a system of equations for integral parameters, they attempt to solve the neutron transport problem from the point of view of a single particle which undergoes series of events during its lifetime. The processes are governed by probabilities of various events—captures, fissions, scatterings and probabilities of outcomes of these events—fission, n,xn reaction, scattering to an angle, path length, etc. All the possible events and outcomes are supplied to the calculation in form of probability density functions, which are sampled with help of random numbers. Monte Carlo solutions rely of the fact that with sufficiently large number of such one-particle experiments a statistically significant behavior stands out.

Monte Carlo codes typically use continuous—energy cross—section data and they enable high—fidelity modeling of virtually any system. Detailed Monte Carlo calculations however tend to be very time consuming, which is why Monte Carlo codes are still waiting for their wide deployment within system codes. The current Monte Carlo codes do not contain any time dependence of the problems either, therefore, unless coupled to a kinetic module, they cannot offer a description of any transient behavior. The increasing computational power together with growing request for the best-estimate solutions is expected to give rise to the deployment of Monte Carlo codes in future system and multi-physics codes.

The flexibility of the geometry modeling together with the high energy resolution are major advantages in investigations of innovative systems with advanced geometries and fuels. Such systems and fuels are the objects of interest in this thesis and Monte Carlo code MCNP [1], [2], [3] has been used for vast majority of the work presented.
3.1.1 MCNP/MCNPX

MCNP [1], [2], [3] is the most widely used Monte Carlo code with application not only in reactor analysis, but also in general particle transport, radiation protection and dosimetry within for example space or medical physics. The name is an abbreviation which stands for Monte Carlo N-Particle Transport Code. The code exists in several versions and it may be coupled to any evaluated data library. In the articles included in this thesis, we used MCNP in two versions with two sets of cross-sections. In Paper I, we used MCNP version 4c3 [1] coupled with the nuclear data library JEFF-3.0 [72]. In all subsequent studies we utilized the MCNPX code [2] together with the continuous energy cross-section library ENDF B-VII.0 [73]. MCNPX stands for Monte Carlo N-Particle eXtended and for the purpose of the work presented in Papers III and IV it is important that this version is capable of depletion calculations.

3.1.2 Nuclear data

The probabilities of particles interactions with matter as well as possible outcomes of these interactions are summarized in nuclear data libraries. In order to be useful for the computational codes, the experimental data that form the basis of these libraries, undergo a process of evaluation and they are complemented by results of theoretical calculations. Currently, there are six general purpose, up-to-date evaluated nuclear data libraries which are prepared to be used by neutronics codes [74]:

- BROND-2.2
- CENDL-3.1
- ENDF/B-VII.0
- JEFF-3.1.1
- JENDL-4.0
- RUSFOND-2010

These libraries contain huge amounts of data in order to describe the particle interactions with hundreds of nuclides. The neutron cross-sections in these libraries generally exhibit low level of uncertainties for the isotopes that have been used in the nuclear reactors for decades and for low energies of neutrons [75]. However, for more exotic isotopes— for example those that have been considered waste rather then fuel for the longer part of the nuclear history as well as for higher neutron energies, there is an apparent lack of consistent data.

This lack of data demonstrates itself in both large error margins reported together with the cross-section data and in disagreement among the respective libraries. It is not clear, whether the discrepancy between two evaluations always
necessarily lies within the error range. An example of the error on the data reported in ENDF/B−VII.1 [73] obtained with software JANIS 3.2 [76] is illustrated in figure 3.1. This figure shows the values of the relative standard deviation associated with the total cross−section of Am-241 and Cm-244 as function of neutron energy.

![Figure 3.1. Relative standard deviation associated with the total cross−section of Am−241 and Cm−244. ENDF B−VII.1 [73].](image)

The relative standard deviation of the total cross−section ranges from 2% up to 33% with rather high values through all the thermal region. These values may be compared to the relative standard deviation associated with the total cross−section of for example U-235, which reaches maximum of 1.5% or Pu-239 with maximal value of 2.5%. The fuels we used in Papers II and IV feature minor actinide contents up to 4% and it is relevant to ask, what can be the effect of the cross−sections on our results. While a detailed analysis would be out of scope of this work, some indications may be found in the literature.

The effect of the data spread on safety parameters calculation has been investigated by Pandikumar et al. [77] for a Fast Breeder Reactor (FBR) fueled with 5% MA. A FBR features a harder spectrum than the BWR investigated in our studies (figure 3.2) and the results of Pandikumar et al. [77] are therefore serve merely as an indication.

In their study, Pandikumar et al. [77] compared ENDF, JENDL and JEFF evaluated data. The results of the calculations with different cross−section data sets exhibit a reasonably good agreement, for instance the reported difference on the Doppler worth lies within 5%. The authors however warn, that higher content of MA would likely lead to larger discrepancies. Figure 3.2 shows a comparison of the neutron spectrum inside the fuel of a BWR fueled with thorium−based trans−
mutation fuels and a FR spectrum with 26% Pu \cite{78}. The BWR transmutation fuel contains 12% Pu and 3.45% MA, according to Paper IV. As visible from figure 3.2, the spectrum in our BWR is somewhat softer than one of a FBR, which means that more neutrons are likely to be influenced by the relatively high uncertainties on the Am-241 and Cm-244 cross-sections in the thermal region, figure 3.1.

![Neutron energy spectrum in fuel at Xe-equilibrium of the fifteenth cycle of multirecycle in BWR (Paper IV) compared to neutron spectrum of a FR with 26% Pu \cite{78}](image)

**Figure 3.2.** Neutron energy spectrum in fuel at Xe-equilibrium of the fifteenth cycle of multirecycle in BWR (Paper IV) compared to neutron spectrum of a FR with 26% Pu \cite{78}.

### 3.2 Feedback to voiding

The usual way to describe the reactor’s feedback to voiding is a Void Coefficient of Reactivity (VCR) defined as the difference of the reactivities over the density change. In terms of $k_{\text{eff}}$, this is:

$$
VCR = \frac{1}{k_{\text{eff}}} - \frac{1}{k_{\text{eff}}^{\text{void}}} \frac{\Delta \rho_{\text{coolant}}}{\Delta \rho_{\text{coolant}}}
$$

(3.1)

This assumes that the reactivity change is proportional to the density change, which is valid only in some special cases and in small intervals of $\Delta \rho$. In general, the reaction to voiding may vary with the percentage of void already present in the coolant, as illustrated in figure 2.2. Therefore, when studying the BWR in Papers II and VI, we chose to represent the voiding feedback as a dependence of $k_{\text{eff}}$ on
void fraction in the core. This representation becomes especially useful with high Pu and MA loadings, as it unambiguously reveals the void intervals with positive feedback to voiding.

### 3.2.1 Density profile in BWR

A BWR core during normal operation contains water of various densities, because of the boiling that takes place in the fuel assemblies. In order to account for the axial moderator density change in the BWR core we divided the water inside the fuel into 25 axial zones with decreasing density. The shape of the operational density profile marked red in figure 3.3 was derived from a representative void profile of an uprated BWR. The water outside the fuel assemblies was assumed to have an axially invariant density, same as the density at the inlet.

In order to investigate the feedback to gradual voiding, we studied the dependence of $k_{eff}$ on the void percentage in Papers II and IV. The density decrease for the detailed void feedback calculations has been realized in ten steps in Paper IV, upper part of figure 3.3 and in seven steps in Paper II. We started from the operational density profile and kept lowering the density in each axial segment according to figure 3.3 until the density reached 0.045 g cm$^{-3}$. This number represents a saturated vapor density at 70 bar [79]. In order to investigate the void effect in the whole range of the possible moderator densities at constant pressure, we also increased the density profile in seven steps (lower subplot of figure 3.3), until reaching the density of saturated liquid at 70 bar, 0.755 g cm$^{-3}$ [79].
In the total void worth calculations, the water and vapor from the fuel assemblies and from the top reflector is instantly voided. The total void worth is defined as the difference between $k_{eff}$ of a core with the operational water densities and $k_{eff}$ of the evacuated core. The result is multiplied by $10^5$ to be expressed in pcm.

\[
\text{total void worth} = 10^5 \cdot (k_{\text{voided}}^{\text{eff}} - k_{\text{eff}})
\] (3.2)

Note, that if this calculation is performed in an infinite lattice rather than a full-core model (Paper IV), the $k_{eff}$ in equation 3.2.1 becomes $k_{inf}$. In MCNPX [2], the evacuation of the coolant is realized by introducing empty cells. In the simulations in Paper IV, the water in the gap between the assemblies as well as the out-core water is remains in place during voiding. This yields the most conservative estimate of the total void worth.

### 3.2.2 2D versus 3D model in voiding calculations

During the review process of Paper II, we came across an interesting question. How (in)correct is it to use an infinite (2D) model for investigation of the voiding behavior?

Assuming no correction for the leakage, we showed that the use of 2D model for voiding behavior substantially overestimates the value of the total void worth and the feedback to voiding. The over-estimation of the feedback to voiding is shown in figure 3.4. The discrepancy between the 2D and 3D model becomes greater at greater shares of void. In the limit case of total void worth, an infinite model may yield as much as 8000 pcm over-estimate (Paper II). Since the total void worth is sometimes used to judge the applicability of new fuels and designs, it is important to be aware of this over-estimation. This is not to say that use of 2D models is entirely incorrect - we found them very useful for the scoping studies in Paper IV when we needed to obtain some indication of the voiding behavior change during multirecycling. Due to the number of steps in the multirecycling and number of fuels investigated, a use of a 3D model for every fuel and every step would be very time consuming.

### 3.3 Doppler coefficient in BWR

In Paper III we evaluated the Doppler coefficient for several types of fuel. To calculate the coefficient, we utilized the empirical relation for thermal and epithermal spectrum reactors, given in [80].

\[
\alpha_{Fuel} = -\ln \left( \frac{1}{\rho(300K)^{1.5}} \right) \beta \sqrt{T_{Fuel}}
\]
Figure 3.4. $k_{eff}$ variation with the in-core void. Difference between the 2D and 3D model is demonstrated on an example of uranium-based fuel with 15.05%Pu and 2.45% MA. The red dash-dot line indicates the operational void percentage, 54%. A three-sigma confidence interval is depicted., Paper II.

\[(3.3)\]

Where $p(300 \text{ K})$ is the resonance escape probability at 300 K, $\beta$ is a constant that depends on the fuel and geometry and $T_{\text{fuel}}$ is the temperature of the fuel. While this relation seems to hold for a cold, unpoisoned reactor fueled with UO$_2$, we have observed that for some other fuels and states the reactivity dependence as $1/\sqrt{T_{\text{fuel}}}$ represents the MCNP [1], [2], [3] values with worse precision. For each fuel configuration, we had four pairs of values ($T$, $\rho(T)$) from the MCNP calculation; out of those, usually three to four were matched by the $1/\sqrt{T_{\text{fuel}}}$ dependence outside two-sigma confidence interval.

The resonance theory shows how the Doppler feedback changes with spectrum - for instance, for fast spectrum reactors with oxide fuels the reactivity change is inversely proportional to $T_{\text{fuel}}$, i.e. $\rho \approx 1/T_{\text{fuel}}$ [81]. In this section we would like to see whether the fit to the calculated values could be improved by optimizing the exponent rather than assuming that it is -1/2 as in [80]. Let us therefore assume the dependence of $\alpha_{\text{fuel}}$ on $T$ in more general form:

$$\alpha_{\text{fuel}} = E \cdot C \cdot T^E$$
3.3. DOPPLER COEFFICIENT IN BWR

(3.4) Where \( E \) is an exponent and \( C \) is a constant. By integration of the definition of the Doppler coefficient of reactivity:

\[
\alpha_{\text{Fuel}} = \frac{\partial \rho}{\partial t}
\]

(3.5) with a boundary condition \( \rho(T_0) = \rho_0 \) we obtain for the reactivity:

\[
\rho(T) = \frac{E \cdot C \cdot T^{E+1}}{E + 1} - \rho_0
\]

(3.6) From this expression we calculated the \( C \), \( E \) and \( \rho_0 \) by finding the best fit to the \( \rho(T) \) values obtained from MCNP [1], [2], [3] calculations for each configuration. Then we evaluated the Doppler coefficient by plugging these values along with the operational temperature into 3.5. Fuels from Paper IV - for the convenience listed in table 3.1, present an interesting material for this calculation. Table 3.2 lists the exponents and Doppler coefficients for these fuels. Figure 3.5 shows an example of fitting for U-1 fuel.

<table>
<thead>
<tr>
<th>Fuel name</th>
<th>Composition</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-1</td>
<td>U-based 12.55% Pu 2.45% MA</td>
</tr>
<tr>
<td>U-2</td>
<td>U-based 12.55% Pu 3.00% MA</td>
</tr>
<tr>
<td>U-3</td>
<td>U-based 12.00% Pu 3.45% MA</td>
</tr>
<tr>
<td>Th-1</td>
<td>Th-based 12.68% Pu 4.08% MA</td>
</tr>
<tr>
<td>Th-2</td>
<td>Th-based 12.25% Pu 4.15% MA</td>
</tr>
<tr>
<td>Th-3</td>
<td>Th-based 12.00% Pu 3.45% MA</td>
</tr>
</tbody>
</table>

Table 3.1. Fuel compositions from Paper IV. Uranium enrichment in the U-fuels in the XV\textsuperscript{th} cycle ranges between 3 and 4.6%.

From table 3.2 it can be seen that the presence of Th generally improves the Doppler coefficient. A comparison of the results for U-3 and Th-3 configurations moreover shows that the thorium based fuel features 1.5 times higher Doppler coefficient than the uranium-based fuel with the same shares of Pu and MA. Even though the increasing MA share is expected to worsen the Doppler coefficient, a serious degradation of the Doppler feedback as a consequence of the elevated concentration of MA reported by [67] and [68] is not observed at the present MA percentages. The second column of table 3.2 also shows that the exponents reside between the literature value for an LWR with \( \text{UO}_2 \) fuel [80] and that of a fast reactor [81].
Figure 3.5. Search for the exponent in the Doppler coefficient calculation, U-1 fuel, Paper IV, table 3.1.

<table>
<thead>
<tr>
<th>Fuel</th>
<th>Calculated E</th>
<th>Doppler coefficient</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-1</td>
<td>-0.62</td>
<td>-2.35</td>
</tr>
<tr>
<td>U-2</td>
<td>-0.73</td>
<td>-2.20</td>
</tr>
<tr>
<td>U-3</td>
<td>-0.72</td>
<td>-2.25</td>
</tr>
<tr>
<td>Th-1</td>
<td>-0.68</td>
<td>-3.42</td>
</tr>
<tr>
<td>Th-2</td>
<td>-0.63</td>
<td>-3.51</td>
</tr>
<tr>
<td>Th-3</td>
<td>-0.67</td>
<td>-3.32</td>
</tr>
</tbody>
</table>

Table 3.2. Exponents and Doppler coefficients [pcm/K] for fuels from Paper IV. The error on the Doppler coefficient is below 0.05 pcm/K.
Chapter 4

Selected problems not covered in the articles

Being slightly out of the scope of the included articles, the topics discussed in this chapter have never been submitted for publication. Yet they are relevant to the presented work and I find them interesting and worthwhile to share in my thesis. The selected problems are relevant either to the Monte Carlo simulation or to the use of transmutation fuels in BWRs. Namely they concern the source convergence in Monte Carlo calculations, He and decay heat production in transmutation fuels.

4.1 Source convergence in MCNP calculations for VHTR

In large, thermal cores a user of MCNP [1], [2], [3] may observe that some quantity, for example power or neutron flux profile, calculated across a perfectly symmetrical system appears to be unsymmetrical. This problem persists even with very high statistical precision - the difference between two points that are physically identical and therefore are expected to show the same values, lies well outside a three - sigma confidence interval, figure 4.1. This non - symmetry moreover propagates to other calculations, like burnup or thermal hydraulics, anytime a coupled computational system is utilized.

The flux profile depicted in figure 4.1 is a consequence of slow redistribution of the fission sites to the fundamental mode during the Monte Carlo calculation. Sometimes the redistribution may be so slow that the source appears to be invariant and the related quantities like $k_{eff}$ or source entropy exhibit a (false) convergence.

4.2 The source convergence problem has been a recognized issue in the scientific community and in the literature, it is often discussed in connection with systems that suffer from inadequate sampling or with loosely coupled systems.

The essence of the problem with the inadequate or incorrect sampling of the modeled system is related to the fact that in a Monte Carlo calculation the fission source is defined only in a limited number of positions. This can result in a situation
where the sampling of an important, or even the most reactive part, is omitted. This in return leads to an incorrect fission source distribution. Such calculation also estimates $k_{eff}$ wrongly. An example of improper sampling is presented in “the
4.1. SOURCE CONVERGENCE IN MCNP CALCULATIONS FOR VHTR

$k_{eff}$ of the world problem” [82]. In this sample problem, there is 9x9 - 1 identical spheres placed in a regular cubic lattice. One sphere, the one located in the center of the field contains a super-critical amount of fuel. Despite the presence of this super-critical element in the array, the calculated $k_{eff}$ of the system appears lower than one.

A loosely coupled system is a system in which there are two or more reactive regions decoupled by a less reactive part. Examples of such systems are fuel storages [83], [84] or a reactive slabs, [85], which is often used by authors as a sample problem to demonstrate the source (non) convergence and possible solutions. In these loosely coupled systems, the neutron transport and resulting redistribution of the fission sites is hindered by the less reactive part(s). The efficiency of the information transfer between the parts of the system can as well be diminished by the distance, if the system is sufficiently large. An example of a large system suffering from poor source convergence may be the AHTR from Paper I and the VHTR, depicted in figure 4.3, which was investigated in some of the articles that are not part of the thesis. The problem with source convergence has also been reported for PWRs [83] and BWR[86].

This section describes the source convergence problem in the large, thermal core of the VHTR and shows the situation in the BWR models used in the included articles. The motivation to describe the problem for VHTR is that 1/ it has not been done before 2/ a lot effort is being devoted to high-fidelity modeling of this system, while an unrecognized false source convergence may be jeopardizing the results on a much more basic level.

Figure 4.3. The VHTR core as modeled in MCNP [1], [2], [3].

The underlying difficulty with the poor information transfer in a system may be expressed by a high Dominance Ratio (DR). DR is an inherent property of a
system and a characteristic of the calculation. It is the ratio of the first and
the fundamental mode eigenvalues, $k_1/k_0$, which comprise the solution to the
eigenvalue equation, written in operator notation as:

$$k\phi = L\phi$$

(4.1)

Where $k$ is an eigenvalue, $\phi$ is a corresponding eigenfunction and $L$ is an
operator representing the system. The solution to equation 4.1 is composed of many
eigenfunctions with their eigenvalues, out of which only the zeroth mode eigenfunc-
tion is positive in the whole system and therefore bears a physical meaning. This
mode is called the fundamental mode and its eigenvalue is $k = k_{eff} = 1$. In a power
iteration, which is a method adopted by Monte Carlo methods for computation of
the fundamental source distribution, the fission source converges to its fundamental
mode as $O((k_1/k_0)^n)$. Consequently, in systems, where $k_1/k_0$ is close to unity, the
convergence may be very slow.

To illustrate the effect of the higher modes in the VHTR simulation, we simpli-
fied the model into a homogeneous reflected reactor. Figure 4.4 shows flux profiles
calculated for this simplified VHTR core fueled with Pu. One of the profiles is
calculated by deterministic code SCALE 5.1 [87], while the other one was obtained
from an MCNP 5 [3] simulation. In both SCALE and MCNP results, the homog-
enized system exhibits a reversed flux profile with a dip in the fuel region. This
is a consequence of increased resonance absorption due to homogenization. In the
homogeneous reactor, the thermal neutron flux peaks in the reflector, while the fast
peak is highest in the vicinity of the reflector, inside the fuel. Both thermal and
fast flux exhibit a depression towards the center of the fuel.

As visible from figure 4.4, the flux profile computed by SCALE is symmetrical,
while the MCNP calculation clearly exhibits a convergence problem. In the MCNP
simulation, we used 250 iteration cycles, out of which 50 were inactive. This number
of cycles had been chosen to achieve a converged value of $k_{eff}$, which was - from
the point of view of the diagnostic tools available in MCNP [3] , fulfilled.

We used the diffusion code Dalton based on the use of the ARPACK library [88],
[89], [90] to calculate the DR, the fundamental and first modes for the homogeneous
core fueled with Pu. The fundamental and first modes decomposed into seven
energy groups are shown in figures 4.5 and 4.6. Apparently, the shape of the MCNP
flux depicted in figure 4.4 reassembles some sort of combination of the fundamental
mode from figure 4.5 and the first mode from figure 4.6.

The diminishing influence of the first mode is illustrated in figure 4.7. Two full
-size, homogenized VHTR cores, a quarter of a PWR core [91] and two hypothet-
ical cases of DR 0.9999 and 0.9000 are presented. The DR for the VHTR cores
obtained by Dalton calculation [90], [88], [89] are 0.9908 and 0.9993, for uranium
and plutonium fueled system, respectively. Note, that there are also methods how
Figure 4.4. Comparison of the flux profile calculated with SCALE [87] and with MCNP [3].

Figure 4.5. The fundamental mode computed by Dalton code [90], [88], [89].

to assess DR with Monte Carlo codes [92], [93], however so far these methods are not a part of the standard calculation routines.

The curves in figure 4.7 indicate that a homogenized VHTR core fueled with Pu requires many more cycles to converge than the 250 which we used in our
The first mode computed by Dalton code [90], [88], [89].

Figure 4.6. The effect of the higher modes on the solution decreases with the number of cycles in the power iteration according to DR.

Figure 4.7. The effect of the higher modes on the solution decreases with the number of cycles in the power iteration according to DR.

calculations. At the same time, figure 4.7 does not provide a complete guideline for the convergence of the fission source to the system’s fundamental mode. If the system is poorly sampled, the number of cycles needed for convergence of the fission source increases. Moreover, it has been shown that a very poor sampling
causes the source to converge to a biased spatial distribution that fails to represent
the physics of the simulated system, page 26 in [94]. According to Brown et al.
[91], the number of neutrons per cycle needed to remove the sampling bias from a
calculation performed on a core model is in order of 10K.

Thousands of iteration cycles with large enough batches of neutrons however
present a challenge to the current computational possibilities of most users. The
sampling can be somewhat improved by simulating only a part of the system, if the
symmetry of the problem allows it. More importantly, this reduction of the system
dimensions also decreases the DR and thereby it helps to eliminate the higher
modes [91]. In case of the VHTR full-core analyses, with both axial and radial fuel
shuffling and control rod holes modeled, there is no symmetry that would allow for
use of reflective boundaries. In the BWR cases, we always utilized 1/4 of the core
in conjunction with reflective boundaries.

Several methods aiming at acceleration of the source convergence have been
suggested. Importance sampling [95], stratified sampling [96], [97], superhistory
powering [98], [99], the Wielandt method [100], [101], a class of methods based
on the fission matrix [102], [103], [104], [105], the Sandwich method [106], Zero -
variance method [107], [108], etc. Some of the methods rely on knowledge obtained
from a deterministic calculation, like the importance sampling [95] or Coarse-Mesh
Finite Difference (CMFD) method [109], while the other take sole advantage of
Monte Carlo solutions. The acceleration of the source convergence is a hot topic
among the Monte Carlo code developers, especially in light of the increasing de-
mands for multiphysics codes and best estimate results. We users may therefore
hope that some of the methods for the acceleration of the source convergence will
soon become available in the standard Monte Carlo codes.

Apart from the solutions to the problem with the source convergence, a user may
be interested in the real error that is associated with the Monte Carlo simulation and
that is partly contributed by the correlations between the fission generations. The
computation of the standard deviation that is a part of the Monte Carlo result omits
these correlations and therefore underestimates the error. According to Brown et
al., the uncertainties calculated by a Monte Carlo code in a model of a quarter of
a PWR core are 1.7 to 4.7 times smaller than the true uncertainties [91]. Methods
have been proposed to improve the estimate of the error in the simulations [110],
[98], [111], [112].

4.1.1 Convergence diagnostic with random numbers

In order to assess the quality of the convergence, especially for non - symmetrical
BWR cores, we used a simple method based on multiple independent runs achieved
by use of different random numbers sets. We show the results of this test for a
reflected pile of VHTR fuel blocks and a quarter of a BWR core as used in Papers
II - IV.

Random numbers utilized in the statistical sampling are one of the factors
determining the commencement and the course of the calculation. Random numbers
together with probability density functions are used in the Monte Carlo method to select the initial source points, collision sites, types of collision events and outcomes, direction of the neutron’s motion, its energy, etc. Obviously, the result of a well-converged calculation must be independent of the chosen random numbers set.

In our calculations, we tested the use of both the Debug Information (DBCN) and the Random Number Generation Cards (RAND) card [2] to influence the initial random number generator seed and stride- the number of random numbers between source particles. The 13th and 14th entry on the DBCN card are identical to the second and third entry on the RAND card. In the present simulation we used the DBCN card, however, modification of the RAND card yields a similar result.

Figure 4.8 reveals an obvious strong dependence of the calculation output on the random numbers set for the pile of VHTR fuel blocks. In the first calculation, both DBCN and RAND cards were absent and the code took the default values. In the second calculation, we modified the 13th entry on the DBCN card. The difference between the two flux-to-average profiles is significant, yet the difference in the eigenvalue is not overwhelmingly large; it is 64 pcm, which translates to 2.6 sigma in the current calculation.

![Figure 4.8. Influence of the random number parameters on the result. VHTR fuel pile, one - sigma confidence interval depicted.](image)

The result for the BWR core model (without CR) shown in figure 4.9 reveals some small differences between the axial flux profiles calculated with and without a DBCN card, which remain within two-sigma confidence interval.
4.2 Helium and fission gas production in transmutation fuels in BWR

The elevated content of MA in the transmutation fuels lead to increased neutron production and heat generation as a result of nuclear decay. In Paper IV we calculated the neutron source and alpha-heating originating from Pu and MA in the transmutation fuels. An alpha particle emitted in the fuel however does not only cause heating of the fuel, but it is also one of the production paths for He. Other modes of helium production are (n, alpha) reactions on O-16 and ternary fissions.

Helium exists in the ceramic fuel in form of gas with limited solubility. It tends to migrate through the fuel, forming bubbles, causing swelling of the fuel and increasing the pressure inside the fuel pins, possessing a risk of the clad rupture.

This section of the thesis compares the production of helium and some fission gases in standard and transmutation fuels. There are some important differences between helium and the other fission gases, hence we show the results both as total production and production of He and other gases separately. One difference between He and the other gases is the lower migration rates of the other gases in the fuel. Therefore, during normal operation, they are likely to be retained within the fuel pellets. It is only at elevated temperatures during a transient that they can be released into the gas plenum of the fuel, contributing to the stress on the cladding. This study takes into account gaseous and volatile fission products Br, Kr, I and Xe.

In collaboration with Kamil Tucek from JRC Petten, we used the MCB code
[113] and our MCNP [2] inputs to calculate the amounts of He, Br, Kr, I and Xe that accumulate in the BWR fuel during one year of storage, six years of Full Power Days (FPD) operation and four years of cooling. This is the deployment scheme we considered in our multirecycling study (Paper IV). For this calculation, we utilized an infinite assembly model (described in the first part of Paper IV) and three fuels:

- standard UO$_2$ fuel with two burnable poison pins per assembly (as described in Paper III)
- uranium - based transmutation fuel U-1 (Paper IV) with 12.55% Pu and 2.45% MA
- thorium - based transmutation fuel Th-3 (Paper IV) with 12.00% Pu and 3.45% MA

The transmutation fuels (table 3.1) do not need to contain any pins with burnable poison, because the very use of MA provides a sufficiently tame reactivity swing. At time 0, in figure 4.10, the UO$_2$ fuel is loaded fresh as it would be in a standard fuel cycle in a BWR. The transmutation fuels, on the other hand, originate from the fifteenth cycle of multirecycling (Paper IV). Figure 4.10 shows the evolution of the amount of He and fission gases in each fuel.

The results reveal that the maximal amount of the gases accumulated in the fuel is reached at the end of the four years of cooling. As expected, the amounts of He are higher for the transmutation fuels with elevated amounts of MA. The uranium - based transmutation fuel U-1 with 2.45% MA accumulates about three
times more He than the reference UO$_2$ fuel. The thorium - based transmutation fuel Th-3 with 3.45% MA accumulates four times more He than the reference UO$_2$ fuel. Assuming an unchanged volume of the gas plenum, one can use the ideal gas law to estimate that the use of transmutation fuels increases the gas pressure in the fuel pins by 3-4 times.

In a scenario where all other gases get entirely released into the fuel rod, the pressure in the transmutation fuel would be 1.5 and 1.7 times higher as compared to UO$_2$ fuel. This is because the production of the other gases is very similar for all the compared fuels and it is essentially only He that makes up the difference between the standard and the transmutation fuel, as visible from figure 4.10.

It is important to note that this study takes advantage of an radial - infinite assembly model. The above listed values therefore represent the oldest assembly that would exist in a reactor core. A real reactor core is always composed of fuel assemblies of various ages.

### 4.3 Decay heat in transmutation fuels

Another issue that emerges as a consequence of the use of the transmutation fuels is an increased decay heat. In Paper IV we investigated the elevated amounts of decay heat from the fuel handling and fuel reprocessing point of view. This means that the results were derived from the fuel composition that goes to reprocessing, i.e. after four years of cooling. Also, the decay heat was assumed to be entirely originating in the alpha decay of Pu and MA isotopes; no fission products were taken into account.

This section of the thesis quantifies how the use of the transmutation fuels from Paper IV influences the power decrease that follows after a sudden shutdown. A scenario with an abrupt stop of the fission reaction would be relevant in an emergency situation similar to Fukushima, where a core on full power gets suddenly shut down and emergency cooling must be established.

Similarly to the previous section, even these calculations use the radial - infinite assembly model. The results hence represent a centrally located fuel assembly at the maximum age an assembly may reach inside the core. The fuels used in the comparison are similar to those in the previous section, i.e. a UO$_2$ fuel from a once - through cycle and two transmutation fuels U - 1 and Th - 3 from the fifteenth cycle of multirecycle (table 3.1). The composition of the fuel corresponds to the end of FPD.

This analysis was accomplished with our MCNP [2] inputs and MCB code [113] ran by Kamil Tucek from JRC Petten.
As figure 4.11 shows, the use of transmutation fuels increases the residual heat after shutdown. Moreover, the decay heat is higher in the fuel with higher content of MA, which is to be expected. U-1 with 12.55% Pu and 2.45% MA exhibits about 0.5% higher decay heat after shutdown as compared to the UO$_2$ fuel. Th-3, which contains 12.00% Pu and 3.45% MA lies about 1% above the standard UO$_2$. One percent of the total reactor power represents in this case about 25 MW.

In more detail, the residual heat is about two times higher for both the transmutation fuels directly after the core shut down and becomes three to four times higher after 30 days. The slower decrease in the residual heating in the transmutation fuels is caused by presence of the longer - living MA.
Chapter 5

Summary of the included articles

According to [4], the vast majority of the reactors operated around the world are thermal reactors. Therefore a clear reason exists for interest in investigating innovative designs and fuels for these systems.

In connection with the GEN IV road map published in 2002, the high temperature reactors received much interest as candidates for future generation of nuclear power plants. The concept of AHTR, a molten salt cooled, very high temperature reactor has been proposed by ORNL in 2005 [114] with the aim of low-pressure, high temperature operation that could ensure a heat source for efficient hydrogen production. At the same time, hydrogen propulsion for vehicles was investigated in many laboratories [115]. **Paper I** elaborates on the coefficients of reactivity in an AHTR fueled with reactor-grade Pu and enriched U. A special focus has been put on the properties of the coolant candidates, out of which FLiBe turns out as the optimal choice. We showed, that an AHTR fuel can be designed with a negative void worth.

The feedback to voiding is traditionally seen as a one of the show-stoppers to efficient transmutation of spent nuclear fuel. Meaning that the feedback turns largely positive with risk of overriding other negative feedbacks present in the core as soon as the amount of the Pu and MA in the fuel exceeds a certain level. The vision of incineration of the spent fuel in future or current systems is however too attractive to be abandoned, as it would not only reduce the amount of nuclear waste that needs to be stored but also increase the energy derived from the initial uranium. Leaving off other issues like economy or proliferation, we focused on the possibility of the transmutation of Pu and MA in current BWRs from a purely neutronics point of view. What are the limitations on Pu and MA content? Can one use hafnium cladding in order to harden the neutron spectra and thereby increase the fission probability of some higher minor actinides [65]?

**In Paper II** we showed that with uranium - based transmutation fuels, a BWR core may accept as much as 3.45% MA (without Np) without encountering problem with the voiding behavior. In order to design a fuel with as much MA as possi-
ble that still maintains a negative void coefficient, we identified which reactions in which isotopes are the ones responsible for the positive feedback to voiding. This was an important foundation for the work in Paper IV. We also showed the importance of full core models in the investigations of the voiding behavior. An infinite assembly model overestimates the feedback to such a degree, that it can lead to faulty decisions on feasibility of concepts. Last but not least we explained, why the presence of Hf causes such large positive feedback to voiding.

Paper IV is a logical continuation of Paper II, however, meanwhile Hallstadius, Lahoda, Wallenius, Jolkkonen et al. patented a new nitride fuel for light water reactors [64]. More precisely, the concept of nitride fuels has been known before, but in exclusive connection with fast reactors. In the patent application, possible methods for stabilizing UN fuels in water and steam at 300° C were suggested. The topic of Paper III hence became the burnup of nitride fuels in BWR. We showed that the higher amounts of heavy metal that can be fitted into the fuel thanks to the UN stoichiometry increase the fuel residence time by 1.4 years. There is no major deterioration of any safety parameters connected with the use of nitride fuels.

Returning to the topic of the transmutation of spent fuel in BWR, in Paper IV we investigated the multirecycling of Pu and MA (without Np) in BWR. We showed the evolution of the potentially problematic feedback to voiding during the multirecycling and identified the maximal MA loadings that provide a negative total void worth during the whole multirecycling. We introduced a new type of fuel in which the uranium is entirely replaced by thorium. The thorium-supported fuel allows for higher MA contents, 3.45% as compared to 2.45% in uranium-supported fuels. In thorium-based fuels, the limitation to the MA loading is the low reactivity of the fuel. We show that in the uranium-supported fuels the need for increasing enrichment in the multirecycling drives the deterioration of the feedback to voiding, while in the thorium-supported fuels the maximal available reactivity is limited by the breeding. We also showed, that it is better to exclude Np from multirecycling due to its deteriorating effect on the feedback to voiding.
Bibliography


