Denaturiertes (proliferationsresistentes) Reaktorplutonium lässt sich mit einer Reihe von unterschiedlichen Brennstoffkreislaufoptionen herstellen. Zunächst kann denaturiertes Reaktorplutonium erhalten werden, wenn statt schwach angereichertem U-235 als DWR-Brennstoff wiederangereichertes U-235/U-236 aus wiederaufgearbeitetem Uran verwendet wird (Brennstoff vom Typ A). Auch die erwarteten 2.500 t Reaktorplutonium (die weltweit bis zum Jahr 2010 entstehen sollen), heute zum größten Teil in Zwischenlagern aufbewahrt, ließen sich in einer Übergangsphase mit den Optionen Brennstoff vom Typ B und D in denaturiertes Reaktorplutonium umwandeln. Denaturiertes Reaktorplutonium könnte denselben Sicherheitsstandard aufweisen wie der heutige schwach angereicherte (<20% U-235) LWR-Brennstoff. Man könnte es durch ein- oder zweimaliges Rezyklieren in DWR und danach durch mehrfaches Rezyklieren in Schnellen Reaktoren (vom Typ CAPRA oder IFR) verbrennen. Wenn es einmal denaturiert ist, könnte dieses Reaktorplutonium auch bei mehrfachem Rezyklieren denaturiert bleiben. In einem DWR könnte denaturiertes Reaktorplutonium beispielsweise in einem Umfang von ca. 250 kg/GWe·a zerstört werden. Während denaturiertes Reaktorplutonium unter weniger strengen *IAEO*-Sicherheitsauflagen rezykliert und verbrannt werden könnte, müsste Neptunium auch in Zukunft in allen Fällen von der *IAEO* überwacht werden, in denen es in größeren Mengen entsteht.

The Generation of Denatured Reactor Plutonium by Different Options of the Fuel Cycle

C.H.M. Broeders and G. Kessler, Karlsruhe

1. Introduction

We demonstrated [1, 2] that reactor plutonium with an increased isotopic content of 6% or somewhat more Pu-238 can be regarded as proliferation resistant. The utilization of such denatured plutonium would become unsuitable for a Nuclear Explosive Device (NED), because the high explosive lenses surrounding the plutonium and its reflector would partially melt or their elevated temperature would lead to self ignition or self explosion of the high explosives (chemical pre-ignition).

However, this will require the production of Pu-238 within the isotopic mixture of reactor plutonium in sufficient quantities. This can be done – as we will show – in an adapted fuel cycle [3]. This will – because of the high α-heat production of Pu-238 – require modern reprocessing and re-fabrication techniques [4–6] as they are being investigated and developed for actinide transmutation and incineration. Earlier studies – which were published around 1980 [7–10] – had indicated already that the Pu-238 production can be enhanced, e.g. by

– using U-235 / U-236 by re-enriching reprocessed spent fuel,
e.g. applying centrifuge enrichment,
– recycling of recovered minor actinides: neptunium, americium, curium in the proportion as they appear after a burn-up of 50 GWd/t in spent LWR fuel,
– reduction of the U-238 content in the fuel by substitution of thorium.

More recent publications [11– 16] confirm these tendencies.

For our fuel cycle calculations we applied the KARBUS / KAPROS system [17–21] which was verified by recalculation of the ICE experiments [22, 23] within the German pressurized water reactor *Obrigheim*. In the next section we will show first how denatured reactor plutonium can be generated directly from re-enriched U-235 / U-236 from reprocessed spent LWR fuel. In a subsequent section we will then show 3 different options how the present reactor plutonium in existing stored spent LWR fuel could be transformed to denatured reactor plutonium by first reprocessing and re-fabricating it to mixed Pu / U oxide (MOX) fuel and recycling it. When the denatured reactor plutonium will be generated, it could be treated by *International Atomic Energy Agency* (*IAEA*) like low enriched (< 20% U-235) LWR fuel and be recycled for further incineration, e.g. in LWRs or Fast Reactor (FRs) [24, 27]. This is displayed later in *Figure 1* going from a "transition phase" to a "civil denatured reactor plutonium cycle". The question then arises how the denatured reactor plutonium changes its isotopic composition when it is multirecycled in LWRs or CAPRA type FRs. We will investigate this question in a following section.

Anschriften der Verfasser:
C.H.M. Broeders
Senior Scientist of the former Institute for Neutron Physics and Reactor Technology
Research Center Karlsruhe
Grenzstrasse 12
76351 Linkenheim-Hochstetten
G. Kessler
Director Emeritus of the former Institute for Neutron Physics and Reactor Technology
Research Center Karlsruhe
Jägersteig 4
76297 Stutensee

Paper presented during an International Workshop on Future Nuclear Systems and Fuel Cycles, Karlsruhe, Sept. 1-2, 2005

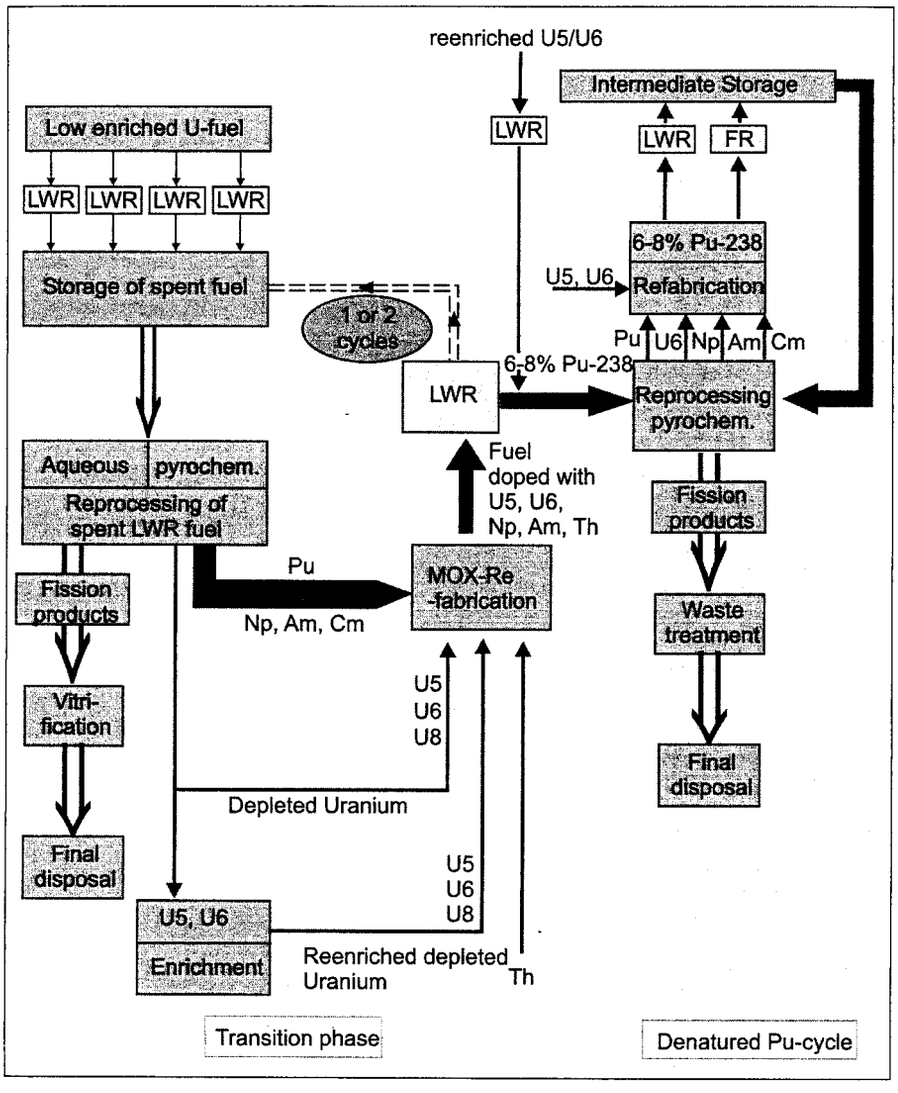

Fig. 1. Transition from present intermediate storage of spent LWR fuel through reprocessing and re-fabrication of dedicated fuel to denatured plutonium fuel cycle and plutonium incineration.

Since also neptunium and americium – when chemically separated in sensitive quantities during actinide transmutation and incineration scenarios – will have to be monitored by *IAEA* we will discuss safeguards concerns in the last section.

Before we report the results of our fuel cycle calculations in the next section, we want to emphasize again that present aqueous PUREX reprocessing and the glove box based MOX re-fabrication techniques would not be applicable any more for denatured reactor plutonium with a Pu-238 content of 6% or somewhat more. The α-heat and neutron radiation of the Pu-238 restrict present MOX re-fabrication plants to about < 4% Pu-238 in the plutonium (licensing requirement). Therefore, the new pyrochemical reprocessing techniques – which will operate inherently at high temperatures – or adapted aqueous reprocessing techniques and the corresponding re-fabrication techniques for metal or MOX-fuel will have to be applied [4–6].

2. Different Fuel Cycle Options to Generate Denatured Reactor Plutonium

We assumed for all our calculations the fuel in form of dioxides of 96% of their theoretical density. The theoretical densities are 10.5 g/cm^3 for uranium, 9.6 g/cm^3 for thorium and 11 g/cm^3 for the minor actinides. The calculations are only performed for PWRs.

2.1 Denatured Reactor Plutonium from Re-enriched U-235 / U-236 of Reprocessed Uranium Fuel

Spent fuel contains about 0.7% U-235 and 0.6% U-236 after a burn-up of 50 GWd/t (about 0.7% U-236 after 60GWd/t burn-up). Such reprocessed uranium can be re-enriched, e.g. by centrifuge technology in a ratio U-235 / U-236 equal 4:3 [28, 29]. In addition, also fresh enriched U-235 / U-238 can be added to attain a mixture as we show it in *Table 1*. Low enriched uranium fuel with 5.52% U-235, 3.0% U-236 and 91,48% U-238 can attain a discharge burn-up of 60 GWd/t and a Pu-238 content in the plutonium isotopic mixture of 11,9% and 7% Pu-242 (*Table 2*). This plutonium in the spent fuel elements is already denatured. These results, however, are valid only for a P/D ratio of 1,443 to assure an acceptable negative coolant temperature and void coefficient in PWRs (see [3] for more details).

2.2. Denatured Reactor Pu from Re-enriched Reprocessed Uranium and Reactor Plutonium from Present Spent LWR Fuel

In this section we assume that re-enriched reprocessed uranium with 5% U-235, 3% U-236 and 92% U-238 is mixed with 6,1% reactor plutonium which would come from reprocessing of the present LWR spent fuel with a burn-up of 50 GWd/t (*Table 1*). The isotopic composition of this plutonium is given in *Table 3* for 10 years after unloading of the irradiated fuel. Again a maximum burn-up of 60 GWd/t can be reached with this fuel composition. The plutonium would have an isotopic composition with 6% Pu-238 and 9% Pu-242 (*Table 2*). This plutonium would be

Fuel type	pitch /diameter P/D	moderator /fuel ratio MF	Fuel composition	Th wt %	U wt %		Plutonium wt %		MA wt %
					Total	Fissile Fraction	Total	Fissile Fraction	
A	1.4427	2.2039	Re-enriched recycled U	0	100	5.52	0	0	0
B	1.3389	1.7132	Re-enriched recycled U + Pu Table IV	0	93.9	5.00	6.1	64.5	0
C	1.4068	2.0302	Re-enriched recycled U + Pu -MA Table VI	0	92.5	5.00	6.5	64.5	1.0
D	1.5926	2.9780	Enriched U + Th + Pu-MA Table VI	52.6	35.1	6.00	10.7	64.5	1.6

Tab. 1. Dedicated fuel compositions for fuel cycle calculations.

Fuel type	p/D	Fuel composition	denatured Pu-238 in %	Pu composition Pu-242 in %
A	1,443	Re-enriched reprocessed U	11,9	7,0
B	1,339	Re-enriched reprocessed U + Pu Table III	6,0	9,0
C	1,407	Re-enriched reprocessed U + Pu + MA Table IV	11,0	9,0
D	1,593	Enriched U + thorium + Pu + MA Table IV	10,2	10,6
			in addition	2,8 % U-233

Tab. 2. Results for Pu-238 and Pu-242 percentage in plutonium after a burn-up of 60 GWd/t of fuel type A, B, C, D.

Isotope	wt %
Pu-238	3.2
Pu-239	56.4
Pu-240	26.6
Pu-241	8.0
Pu-242	5.8

Tab. 3. Plutonium composition from spent nuclear fuel with 50 GWd/t, 10 years after unloading.

at the lower limit which we have defined in [1, 2] for denatured reactor plutonium. A P/D ratio of 1,339 would guarantee an acceptable negative coolant temperature and void coefficient (see [3] for more details).

2.3 Denatured Reactor Plutonium from Re-enriched Reprocessed Uranium and present Reactor Plutonium as well as Minor Actinides from Spent LWR Fuel

In this section we extend the possibilities for Pu-238 generation and mix the re-enriched reprocessed uranium with 5% U-235, 3% U-236 and 92% U-238 with 6,5% reactor plutonium and 1% minor actinides in the composition as they appear altogether after reprocessing of present LWR fuel with a burn-up of 50 GWd/t (10 years after unloading). *Table 1* and *Table 4* display the different contents of this fuel mixture.

Isotope	wt %
Np-237	6.65
Pu-238	2.75
Pu-239	48.73
Pu-240	23.02
Pu-241	6.94
Pu-242	5.04
Am-241	4.64
Am-242m	0.19
Am-243	1.48
Cm-243	0.00
Cm-244	0.50
Cm-245	0.06
Cm-246	0.00

Tab. 4. Plutonium and minor actinides composition from spent nuclear fuel with 50 GWd/t, 10 years after unloading.

Again a discharge burn-up of about 60 GWd/t can be attained. The plutonium composition at discharge would contain 11% Pu-238 and 9% Pu-242. This discharged plutonium would be denatured. A P/D ratio of 1.407 would guarantee a negative coolant temperature and void coefficient of such a PWR (see [3] for more details).

2.4 Denatured Reactor Plutonium and Denatured Uranium from Low Enriched Uranium, Thorium, Present Reactor Plutonium and Minor Actinides from Spent LWR Fuel

In this section we extend the previous possibilities by replacing U-238 by Th-232. Only low-enriched uranium from natural uranium is used (*Table 1*). In this case also U-233 is generated in addition to Pu-238. Again a discharge burn-up of 60 GWd/t can be attained. The plutonium composition at discharge would contain 10.2% Pu-238 and 10.6% Pu-242. In addition U-233 is generated and the discharged fuel would contain 2,8% U-233 and still 3,5% U-235 as fissile uranium isotopes. A P/D = 1,59 would guarantee a sufficiently negative coolant temperature and void coefficient for such a PWR.

These results show that denatured reactor plutonium can be generated from re-enriched reprocessed uranium (option A). In addition, reactor plutonium from discharged fuel with a burn-up of 50 GWd/t can be converted during one single burn-up cycle of 60 GWd/t (options B,C,D) into denatured reactor plutonium as defined in [1, 2]. Such denatured reactor plutonium would be, according to [1] – unsuitable for a Nuclear Explosive Device.

3. Long Term Behavior of Denatured Reactor Plutonium in PWRs or FRs

As mentioned already in Section 1 it is important to analyze the long term behavior of reactor plutonium during recycling in PWRs and FRs, because plutonium and actinides should be incinerated by multi-recyling strategies [24, 27]. We selected one isotopic composition of denatured reactor plutonium as shown by *Table 5* which is mixed with re-enriched reprocessed uranium with 5% U-235, 3% U-236 and 92% U-238 and followed its isotopic change during burn-up up to 60 GWd/t discharge burn-up. (*Table 5*). It can be concluded that the isotopic content of both Pu-238 and Pu-242 is slightly increasing, i.e. the denatured reactor plutonium remains denatured.

We also followed the isotopic change of denatured reactor plutonium, when subjected to a typical fast reactor neutron spectrum (e.g. SNR-300). In this case the denatured reactor plutonium was mixed with natural uranium only. As could be expected – due to the higher fission /absorption ratio in the fast reactor neutron spectrum – the content of Pu-238 decreased from, e.g. 11,7% to 10%, over a burn-up of 120 GWd/t. This means that either one has to load fast reactor denatured plutonium mixed with natural

Isotope	wt % begin of cycle	wt % end of cycle 60 GWd/t
Pu-238	7.7	9.0
Pu-239	44.0	36.0
Pu-240	31.0	26.5
Pu-241	10.3	18.0
Pu-242	7.0	10.5

Fig. 5. Plutonium composition of denatured PWR MOX fuel at begin and end (60 GWd/t) of burn-up cycle.

uranium with a somewhat higher Pu-238 content in order to account for the about 1,7% decrease over a typical burn-up of about 120 GWd/t or one can still reduce the decrease of Pu-238 by the admixture of minor actinides to the fresh fuel.

In any case PWRs will operate in a certain symbiosis with FRs in the future. In this case the PWRs can then generate some Pu-238 for the FRs. The incineration rate of denatured reactor plutonium when loaded in a PWR core is about 10 kg/t_{HM}, i.e. for a 1 GWe PWR with a loading /unloading rate of 25 t per year about 250 kg / GWe·y would be incinerated.

4. Strategy for a Proliferation Resistant Fuel Cylce

In Sections 2 and 3 we showed how denatured reactor plutonium with 6% Pu-238 or somewhat more can be generated directly from re-enriched reprocessed uranium or by conversion of present plutonium from spent fuel of LWRs. This is shown on the left side of *Figure 1* (transition phase).

After the reactor plutonium will be converted to denatured reactor plutonium it will go into the denatured plutonium fuel cycle (right side of *Figure 1*) and remain there for incineration by multi-recycling.

The present civil nuclear energy economy in the world will have produced about 2,500 t of reactor plutonium by the year 2010, about 80% of which will be in intermediate storage and 20% of it will already be reprocessed and stored in MOX fuel (mainly in Europe and Japan) [30]. This reactor plutonium together with the accompanying neptunium, americium and curium could be incinerated by multi-recycling [24–27, 31, 34] in reactors instead of storing the spent fuel elements in deep repositories (direct fuel disposal) which would lead to long term accumulation of all plutonium, neptunium and americium (curium-244 has a decay half time of about 18y).

The use of re-enriched reprocessed uranium (fuel type A) would not require a change in fuel fabrication technology or in core design of PWRs. But the enrichment cost might be somewhat higher. There is extensive experience available for MOX re-fabrication and irradiation behaviour in Europe (fuel type B) [31, 36]. Fuel cycle cost for MOX fuel including reprocessing are somewhat higher than present LWR uranium fuel [35]. Only aqueous reprocessing technology (PUREX) is presently available on a large scale at *La Hague* (France), *Sellafield* (UK) and *Tokai-mura* (Japan) [32]. Therefore, advanced aqueous or pyrochemical reprocessing technology and the corresponding re-fabrication technology will have to be developed for fuel types C and D (*Table 1* and *Table 2*) as it would also be the case for actinide transmutation scenarios [33–35]. Additional irradiation testing of such fuel rods would become necessary [31, 33]. The PWR core design for fuel type C and D would need small changes and adaptation for their control systems, e.g. more control rods, more poison rods, high enriched boron [25–27]. Also, a higher P/D ratio, as e.g. in case of fuel type D, (*Table 1*) would need changes by either

- reducing the fuel pin diameter, or
- replacing a certain number of fuel rods by water rods [27].

In the latter cases this could lead to an increase of the pressure vessel diameter or to a reduction of the power output [3].

The incineration of denatured plutonium and minor actinides would need the deployment of sufficient reprocessing and re-fabrication capacities and suitable reactors (MOX-PWRs and CAPRA-FRs or IFRs [26, 27, 32, 34]). The optimum strategy would be to convert the present reactor plutonium in PWRs to denatured proliferation resistant reactor plutonium first by recycling once or twice and then incinerate the denatured reactor plutonium and minor actinides in CAPRA-type FRs or IFRs (because of too high build-up of americium in PWRs during multirecycling and better incineration efficiency of FRs) [34, 35]. This would be similar to the present strategy followed by the U.S. [33, 34] and France [32].

If the 2,500 t of reactor plutonium in spent fuel elements in the world after reprocessing would be used in PWR MOX fuel elements with, e.g. fuel type C (*Table 1*) this would be sufficient for about 38,500 t of MOX fuel elements or the first core inventory of about 320 GWe MOX PWRs.

5. Safeguards Issues

5.1 Safeguards for Denatured Plutonium

In [1, 2] it was demonstrated that reactor plutonium with an isotopic content of about 6% or somewhat more Pu-238 can be regarded as denatured or proliferation resistant plutonium. Such denatured plutonium could be treated by *IAEA* safeguards like low enriched uranium with <20% U-235 or <12% U-233.

In this context the question is interesting to discuss whether such denatured reactor plutonium as e.g. given in *Table 5* with 7.7% Pu-238, 44% Pu-239, 31% Pu-240, 10.3% Pu-241, 7% Pu-242 could be modified by enrichment techniques to, e.g., weapons plutonium [37]. We discuss here only centrifuge enrichment and LASER enrichment technology:

For the utilization of centrifuge enrichment technology this question can be clearly answered [29]. The theory for centrifuge enrichment and its experimental verification show, as an example, for a 4 component isotopic mixture of uranium isotopes the following different enrichment factors U-232 (11), U-234 (9), U-235 (7), U-236 (5) (enrichment factors in brackets). The enrichment factors are monotonically decreasing with higher mass of the different isotopes. This means that the isotope with the lowest mass (U-232) is being enriched with the highest enrichment factor of 11. If we apply these results to our above 5 component isotopic mixture of Pu-238, Pu-239, Pu-240, Pu-241, Pu-242 of *Table 5* it leads to the conclusion that Pu-238 as the lowest mass isotope will have the highest enrichment factor of all plutonium isotopes and cannot be depleted as it would be needed for weapons plutonium with only 0.01% Pu-238 [37].

For LASER isotope separation not sufficient scientific information is available in the open literature to answer this question directly. But it is known that the different isotopes can be selectively excited by LASER beams and be separated. However, if the argumentation of [38, 39] is applied to our denatured plutonium, one can conclude that LASER enrichment would become very costly, if technically possible at all. All earlier efforts of enriching uranium by LASER technology were not pursued to

technical scale in the USA and elsewhere up to now.

In any case it would be technically much easier and less costly to enrich uranium by centrifuge technology to weapons uranium with ≥93% U-235.

5.2 Safeguards for Minor Actinides

The minor actinides, e.g. neptunium, americium, curium, have similar critical masses as high enriched uranium (>90% U-235). Neptunium is considered usable for nuclear explosive devices [40, 41]. It has a bare critical mass of about 57 kg. With a reflector, e.g. BeO, this critical mass can be reduced to about 45 kg. It has no α-heat production and essentially no spontaneous fission neutron production, like e.g. Pu-238, Pu-240, Pu-242.

The critical mass of Am-241 was calculated to be about 34 to 45 kg and for Am-243 a critical mass of between 111 and 193 kg was determined, if reflected by steel in both cases [47]. Pure Am-241 can originate from the decay of Pu-241 in aged plutonium [38]. But in spent reactor fuel both americium isotopes Am-241 and Am-243 always appear, e.g. in a ratio 4:1 after a burn-up of about 50 GWd/t and a cooling period of 10 years. The critical mass of both Am-241 and Am-243 together in a mixture of 4:1 will be slightly higher than 34 kg if we take the lowest calculated values [47] to be conservative. Am-242m has a considerably lower critical mass, but is produced in spent fuel in only negligible amounts.

It is difficult to separate americium and curium chemically from each other. In addition, Am-241 has an α-heat production of 106 W/kg. Am-241 also has a spontaneous fission neutron source rate of 800 n/s·kg, whereas Am-243 has an additional spontaneous fission neutron source rate of 1.5·1E05 n/s kg. This high spontaneous fission neutron rate would require an implosion type Nuclear Explosive Device and lead to serious pre-ignition problems [42, 43]. Such an implosion type NED would produce the extremely high α-heat power of about 3.6 kW and this would lead to even more serious problems for the high explosive lenses as described for denatured reactor plutonium with more than 6% Pu-238 in the plutonium [1,2]. We, therefore, exclude Am-241 and Am-243 in a mixture of 4:1, as it appears in spent fuel from safeguards concerns.

The *IAEA* has begun to adopt measures to monitor neptunium [44, 45]. Unfortunately neptunium cannot be denatured. Therefore, it should be incinerated as soon as it is generated. Reprocessing techniques which keep plutonium, neptunium, americium and curium together as they exist in spent fuel after a certain burn-up can only alleviate (with respect to plutonium and neptunium) the safeguards concerns to a certain extent. But only denatured reactor plutonium with 6% Pu-238 or somewhat more can assure proliferation resistance [1, 2]. Also, if the denatured plutonium is combined with U-233 (fuel type D or as discussed below) the production of neptunium in spent fuel can be minimized. In *Table 6* and *Table 7* we show the different fuel cycle options of Sections 2 and 3 for the generation of denatured plutonium (*Table 6*) as well as for plutonium incineration (*Table 7*) and their safeguards implications. *Table 6* displays the different options discussed in Sections 2 and 3. The generation of denatured reactor plutonium via fuel type A needs re-enriched U-235/U-236 fuel which can be treated, regarding *IAEA* safeguards, like present low enriched (<20% U-235) LWR fuel. It generates denatured reactor plutonium which would not be of special safeguards concern anymore and can be treated regarding safeguards like low enriched LWR fuel as well. But the neptunium in the spent fuel must be monitored by *IAEA*. This neptunium together with americium and curium and the denatured reactor plutonium can be used further (in fuel type C or D) for multi-recycling in FRs.

fresh fuel type	A	B	C	D
safeguards	●	✱	✱	✱
pyro-chemical reprocessing	Np^+	Np^+	Np^+	Np^+

● as low enriched LWR fuel at present
✱ as MOX fuel at present
\+ monitoring of neptunium in future

Fig. 6. Safeguards implications for fuel type A, B, C, D.

Destruction of denatured plutonium		
fresh fuel	in LWRs denatured Pu	in FRs denatured Pu
Safeguards	●	●
reprocessing	(Np^+)	(Np^+)

● as low enriched LWR fuel at present,
\+ monitoring of neptunium, if considerable amounts are produced

Fig. 7. Safeguards implications for denatured plutonium.

The option fuel type B with re-enriched U-235/U-236 and plutonium from present spent fuel needs the same safeguards as presently required by *IAEA* for MOX fuel. It would have to be recycled probably twice in PWRs to obtain denatured reactor plutonium. The neptunium together with americium and curium which is separated by advanced aqueous or pyro-chemical reprocessing from the spent fuel can also be used for multi-recycling of TRU (containing denatured reactor plutonium) in FRs.

The options C and D with plutonium from present spent fuel as well as neptunium and americium need the same *IAEA* safeguards measures as for present MOX fuel (fuel type B), but in addition monitoring of neptunium by *IAEA* would be required. The neptunium, americium and curium together with the denatured reactor plutonium which are separated during advanced aqueous or pyro-chemical reprocessing from the spent fuel can be used for multirecycling of TRU in FRs.

In *Table 7* we show the safeguards implications for the incineration phase of denatured plutonium. The fuel with denatured reactor plutonium should preferably be mixed with U- fuel containing denatured U-233, U-238 fuel in order to minimize the generation of neptunium via the build-up from U-235 and U-236. One such example could be denatured U-233 fuel from PWRs as described by [46] based on plutonium/thorium fuel and U_{nat} in a seed blanket geometry. In this proposal the blanket subassemblies are loaded with plutonium/thorium and U_{nat} in a carefully selected amount such that denatured uranium with 0.1% U-232, 11.25% U-233, 3.69% U-234, 0.75% U-235, 0.19% U-236, 79.62% U-238 (including 5% Pa-233) is generated. Such denatured uranium fuel could be blended with U_{nat} to

reach the desired enrichment level for U-233.

Both, in case of LWR and FR fuel with denatured reactor plutonium, the safeguards implications would then be the same as for present fresh low enriched LWR fuel. *IAEA* monitoring of neptunium, however, would become necessary, for all cases in which considerable amounts of neptunium are produced.

6. Conclusions

It was shown that denatured (proliferation resistant) reactor plutonium can be generated in a number of different fuel cycle options. First denatured reactor plutonium can be obtained if, instead of low enriched U-235 PWR fuel, re-enriched U-235/U-236 from reprocessed uranium is used (fuel type A). Also the envisaged existing 2,500 t of reactor plutonium (being generated world wide up to the year 2010), mostly stored in intermediate fuel storage facilities at present, could be converted during a transition phase into denatured reactor plutonium by the options fuel type B and D. Denatured reactor plutonium could have the same safeguards standard as present low enriched (<20% U-235) LWR fuel. It could be incinerated by recycling once or twice in PWRs and subsequently by multi-recycling in FRs (CAPRA type or IFRs). Once denatured, such reactor plutonium could remain denatured during multiple recycling. In a PWR, e.g., denatured reactor plutonium could be destroyed at a rate of about 250 kg/GWe·y. While denatured reactor plutonium could be recycled and incinerated under relieved *IAEA* safeguards, neptunium would still have to be monitored by the *IAEA* in future for all cases in which considerable amounts of neptunium are produced.

Acknowledgement

The authors wish to thank *Mrs. M. Wettstein* for typing the paper, *Mrs. A. Veser* for preparing the figure and *Dr. E. Kiefhaber* for his critical review.

References:

[1] *G. Kessler*: Plutonium denaturing by Pu-238, to be published in Nucl. Sci. Eng.

[2] *G. K*essler: Analysis for a future proliferation resistant plutonium fuel cycle. *atomwirtschaft,* 51, 5, pp. 33 (2006)

[3] *C.H.M. Broeders, G. Kessler*: Fuel cycle options for the production and utilization of denatured plutonium, to be published in Nucl. Sci. Eng.

[4] *J.J. Laidler* et al.: Development of pyro-processing technology, Progress in Nuclear Energy, 31 (1/2), 131 (1997)

[5] *O. Conocar, N. Douyere* et al.: Promising Pyrochemical Actinide/Lanthanide Separation Processes Using Aluminum, Nucl. Sci. Eng., 153, pp. 253-261 (2006)

[6] *R. Herbig, O.V. Skiba* et al.: Vibrocompacted fuel for the liquid metal reactor BOR 60, J. of Nuclear Materials, 204, 93 (1993)

[7] INFCE – Summary volume and reports of INFCE working groups, *IAEA*, Vienna, 1980

[8] *C.D. Heising-Goodman*: An evaluation of the plutonium denaturing concept in an effective safeguards method, Nucl. Techn., 50, 242 (1980)

[9] *D.O. Campbell, E.M. Gift*: Proliferation resistant nuclear fuel cycles, ORNL/TM-6392 (1978)

[10] *J.V. Massey, A. Schneider*: The role of Plutonium-238 in nuclear fuel cycles, Nucl. Techn., 56, 55 (1982)

[11] *Y. Ronen, Y. Kimhi*: A non-proliferation nuclear fuel for LWR, Nucl. Techn., 96, 133 (1991)

[12] *E. Shwageraus* et al.: Use of thorium for transmutation of plutonium and minor actinides in PWRs, Nucl. Techn., 147, 53 (2004)

[13] *M. Saito* et al.: Innovative nuclear energy systems for inherently protected plutonium production, Int. Conf. on Innovative Technologies for Nuclear Fuel Cycles and Nuclear Power, Vienna (June 23-26, 2003)

[14] *M. Saito*: Advanced core concepts with enhanced proliferation resistance by transmutation of minor actinides, paper No. 172, Proceedings of Global 2005, Oct. 9-13, 2005, Tsukuba, Japan

[15] *M. Saito*: The technical barriers for proliferation resistance in nuclear material management, *IAEA* technical meeting on fissile material management strategies for sustainable nuclear energy, Sept. 12-15, 2005, Vienna, Austria

[16] *M. Osaka, M. Saito* et al.: Aspects of Pu-238 production in the experimental fast reactor JOYO, Annals of Nuclear Energy, 32, 1023 (2005)

[17] *C.H.M. Broeders*: Entwicklungsarbeiten für die neutronenphysikalische Auslegung von Fortschrittlichen Druckwasserreaktoren (FDWR) mit kompakten Dreiecksgittern in hexagonalen Brennelementen. PhD Thesis *TU Karlsruhe* and *KfK* 5072 (1992)

[18] *C.H.M. Broeders* et al.: Modular program system for nuclear reactor analysis, status and results of selected applications, Jahrestagung Kerntechnik, Düsseldorf, 2004

[19] *J.R. Askew* et al.: A general description of the lattice code WIMS, Journal of the *British Nuclear Society,* 5, 564 (1966)

[20] *D. Woll*: Aufbau der Gruppenkonstantenbibliothek GRUBA und ihre Verwaltung durch das Programmsystem GRUMA, *KFK* 3745 (1984)

[21] *C.H.M. Broeders*: Neutronic investigations of an equilibrium core for a tight lattice light water reactor, Kerntechnik 57, No. 1, 23 (1992)

[22] *L. Koch, S. Schoof* (eds.): The Isotope Correlation Experiment ICE, ESARDA 2/81, EUR 7766EN, *KFK* 3337 (1981)

[23] *L. Send*: Investigations for fuel recycling in LWRs, Diploma Thesis *TU Karlsruhe,* October 2005 (available on http://inrwww.fzk.de)

[24] *C.H.M. Broeders*: Investigations related to the build-up of transurania in pressurized water reactors, FZKA 5784, Dec. 1996

[25] *S. Aniel-Buchheit* et al.: Plutonium recycling in a Full-MOX 900-MW (electric) PWR: Physical analysis of accident behaviors, Nucl. Techn., 128, 241, Nov. 1999

[26] *J. Tommasi* et al.: Long-lived waste transmutation in reactors, Nucl. Techn., 111, 113, July 1995

[27] *P. Barbrault*: A plutonium-fueled high-moderated pressurized water reactor for the next century, Nucl. Sci. & Eng., 122, 240, 1996

[28] *F.J. Müller*: Beitrag zur Theorie der Mehrkomponenten-Isotopentrennung und deren Anwendung insbesondere auf die Untersuchung des U-236-Aufbaus im Brennstoffkreislauf, Dissertation *RWTH Aachen,* Brosch. 1540 (1975)

[29] *H.O. Hoppe*: Urenco personal communication, Oct. 2005

[30] *K. Fukuda*: *IAEA* scenario of MA transmutation in LWR, COES-INES Topical Forum on Protected Plutonium Utilization for Peace and Sustainable Prosperity, *Tokyo Institute of Technology*, March 1-4, 2004

[31] *M. Salvatores*: Scenarios using P/T and major challenges, Nuclear Fuel Cycle Workshop, Karlsruhe, Sept. 1-3, 2005

[32] *J. Bouchard*: The enclosed fuel cycle and nonproliferation issues, Global 2005, Tsukuba, Japan, Oct. 2005

[33] GNEP roll out means big jump for fuel cycle, Nuclear News, *American Nuclear Society*, March 2006

Advanced fuel cycle initiative, www.ne.anl.gov/research

[34] *D.C. Wade, R.N. Hill*: The design rationale for the IFB, Progress in Nuclear Energy, Vol. 31, No. 1/2, pp. 13-42, 1997

[35] *P.J. Finck*: Answering questions on reprocessing explaining the closed fuel cycle, Nuclear News, p. 10, September 2005

[36] *J. Krellmann*: Plutonium processing at the *Siemens* Hanau fuel fabrication plant, Nucl. Techn., 102, 18, April 1993

[37] Management and disposition of excess weapons plutonium, reactor related options, *National Academy of Sciences,* National Academic Press, Washington DC, 1995

[38] *St. Fetter* et al.: Detecting Nuclear Warheads, Science and Global Security, Vol. 1, pp. 225 (1990)

[39] *R.W. Solarz*: A physics overview of AVLIS, UCID-20343, Febr. 1985

[40] *D. Loaiza* et al.: Results and analysis of the spherical Np-237 critical experiment surrounded by highly enriched uranium hemispherical shells, Nucl. Sci. & Eng., 152, 65 (2006)

[41] *D. Loaiza, W. Stratton*: Criticality data for spherical U-235, Pu-239 and Np-237 systems reflector moderated by low capturing-moderator materials, Nucl. Techn., 146, 143, May 2004

[42] *W. Seifritz*: Nukleare Sprengkörper – Bedrohung oder Energieversorgung für die Menschheit?, *Karl Thiemig,* München, 1983

[43] *A. deVolpi*: Proliferation, Plutonium and policy/institutional and technological impediments to nuclear weapons propagation, Pergamon Press, New York, 1979

[44] *D. Albright, L. Barbour*: The challenges of fissile materials, Chapter V: separated neptunium-237 and americium, *Institute for Science and International Security,* Press, Washington D.C., March 1999

[45] *A. Morgenstern* et al.: Analysis of Np-237 in spent fuel solutions, Radiochim. Acta, 90, 389 (2002)

[46] *A. Galperin* et al.: A Pressurized Water Reactor plutonium incinerator based on thorium fuel and seed-blanket assembly geometry, Nucl. Techn. 132, 214, Nov. 2000

[47] *M. Diaz* et al.: Critical mass calculations for Am-241, Am-242m and Am-243, 7th International Conference on Nuclear Criticality Safety (ICNC 2003), Oct. 24-24, 2003, Tokai-mura, Japan □