FINAL REPORT ON THE IAEA COORDINATED RESEARCH PROGRAM ON THE POTENTIAL OF THORIUM-BASED FUEL CYCLES TO CONSTRAIN PLUTONIUM AND TO REDUCE THE LONG-TERM WASTE RADIO-TOXICITY

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ABSTRACT

In 1995, the IAEA launched a Coordinated Research Project (CRP) on the "Potential of Thorium-based Fuel Cycles to Constrain Plutonium and to Reduce Long-term Waste Toxicity." The following Member States participated in the CRP: China, Germany, India, Israel, Japan, Republic of Korea, Netherlands, Russia, and the United States of America. The research program was divided into three stages:

- Stage 1: Benchmark calculations
- Stage 2: Optimization of the incineration of plutonium in various reactor types.
- Stage 3: Assessment of the resulting impact on the waste toxicity.

The present paper reports the results of Stage 3 and the overall conclusions from the CRP.
Generally, there is a remarkable potential to effectively constrain the production of plutonium and to reduce existing plutonium stockpiles by implementing the thorium fuel cycle in a large number of current reactors. The results of the research offer a promising, possible near-future solution in view of the proliferation concern combined with the plutonium. However, plutonium-incineration in thermal reactors turns out to be less effective from the point of view of the reduction of the long-term radio-toxicity of the nuclear waste, which has to be finally disposed.

1. INTRODUCTION

Large stockpiles of civil plutonium have accumulated worldwide from nuclear power programs in different countries. There is a serious public and political concern in the world about misuse of this plutonium and about accidental release of highly radiotoxic material into the environment. It is therefore necessary to safeguard the plutonium under strong security. One alternative for the management of plutonium is to incinerate it in reactors. However, if the reactors are fuelled with plutonium in the form of U-Pu-mixed oxide (MOX), second-generation plutonium is produced. A possible solution to this problem is to incinerate plutonium in combination with thorium. The thorium cycle produces ^{233}U which, from a non-proliferation point of view, is preferable to plutonium for two reasons. Firstly, it is contaminated with ^{232}U, which decays to give highly active daughter products. This would make handling and diversion difficult. Secondly, in case this is not sufficient a deterrent, the ^{233}U could be denatured by adding some ^{238}U to the thorium. The quantity of ^{238}U could be fine-tuned so as to be sufficient to denature the ^{233}U, but not so much as to produce a significant quantity of plutonium. The thorium option not only produces electricity, but also replaces the plutonium with denatured ^{233}U, which can be used in other reactors at a later date. All this can be done in existing reactors.

The activities carried out by the IAEA within its Program on Emerging Nuclear Energy Systems for Energy Generation and Transmutation of Actinides include preparation of status reports on advanced technologies development, conduct of technical information exchange meetings and co-operative Coordinated Research Programs (CRPs).

CRPs are tools which are effectively used in the Agency activities to promote exchange of scientific and technical information and implement collaborative research and development in nuclear power reactor technology. Apart from CRPs allowing to share the efforts on an international basis and to benefit from the joint experience and expertise of researchers from the participating institutes, CRPs are fostering international team building.

In 1995, the Agency initiated the CRP on "Potential of Thorium-based Fuel Cycles to Constrain Plutonium and to Reduce Long-term Waste Toxicity". The scope of this CRP was discussed and agreed upon by the participants of the Consultancy on “Thorium-based Fuel Cycles”, held from 6 to 9 June 1995 at the Agency's Headquarters in Vienna. The Member States joining into the CRP were: China, Germany, India, Israel, Japan, Republic of Korea, the Netherlands, Russian Federation and the United States of America.

This CRP examines the different fuel cycle options in which plutonium can be recycled with thorium to incinerate plutonium. The potential of the thorium-matrix has been examined through computer simulations. Each participant has chosen his own cycle, and the different cycles are compared through certain predefined parameters (e.g., annual reduction of plutonium stockpiles). The radio-toxicity accumulation and the transmutation potential of thorium-based cycles for current, advanced and innovative nuclear power reactors are investigated.

The research program was divided into three stages:

- Stage 1: Benchmark calculations
- Stage 2: Optimization of the incineration of plutonium in various reactor types.
- Stage 3: Assessment of the resulting impact on the waste radio-toxicity.

The results of Stage 1 were presented at ICENES 98 /1/ and the results of Stage 2 at ICENES 2000 /2/. The present paper – besides a short summary of the results of Stage 2 - reports the results of Stage 3 and the overall conclusions of the CRP. It was prepared by H. J. Rütten, Scientific coordinator of the CRP, and representatives of IAEA on behalf of all the Chief Scientific Investigators of this CRP, namely:
Zhu Yongjun (China), R. Srivenkatesan (India), A. Galperin (Israel), Tomohiko Iwasaki (Japan), Won Seok Park (Republic of Korea), H. T. Klippel (The Netherlands), V. Ilynin (Russia), and M. Todosow (USA).

2. EVALUATION OF THE POTENTIAL OF LWRS, HTRS, HWRS AND MSRS FOR PLUTONIUM-INCINERATION (SUMMARY)

The aim of the research during the second stage of the CRP was to find fuelling strategies, which – on the basis of proven reactor technology - are suitable to burn plutonium most effectively on the one hand, and to minimize the amount of plutonium to be disposed, on the other hand. Only plutonium of the first generation, namely

- typical LWR-plutonium and
- weapons-grade plutonium,

was to be regarded in the scope of this CRP. Four types of reactors were investigated in view of their potential to burn plutonium, each by one group of countries. Israel, Korea, Russia and the USA have done research on LWRs, China, the Netherlands and Germany have studied plutonium-burning in Modular HTRs, India studied the respective potential of the PHWR and Japan that of the MSR. Two characteristic values –aiming at two different goals of optimization- may describe the effectiveness of plutonium-burning in the different reactors:

(1) The amount of plutonium, which is burned per unit of produced electric energy. Maximization of this item optimizes the speed of the reduction of existing plutonium-stockpiles.

(2) The relation between the amount of plutonium, which is burned during the lifetime of the fuel elements, and the amount of plutonium, which is residual in the unloaded fuel. Maximization of this item minimizes the plutonium-quantity, which either has to be finally disposed or has to be re-fabricated a second time.

Fig. 1 summarizes the results of the research, which has been done for the use of LWR-grade plutonium and of weapons-grade plutonium, respectively, during stage 2 of the CRP.

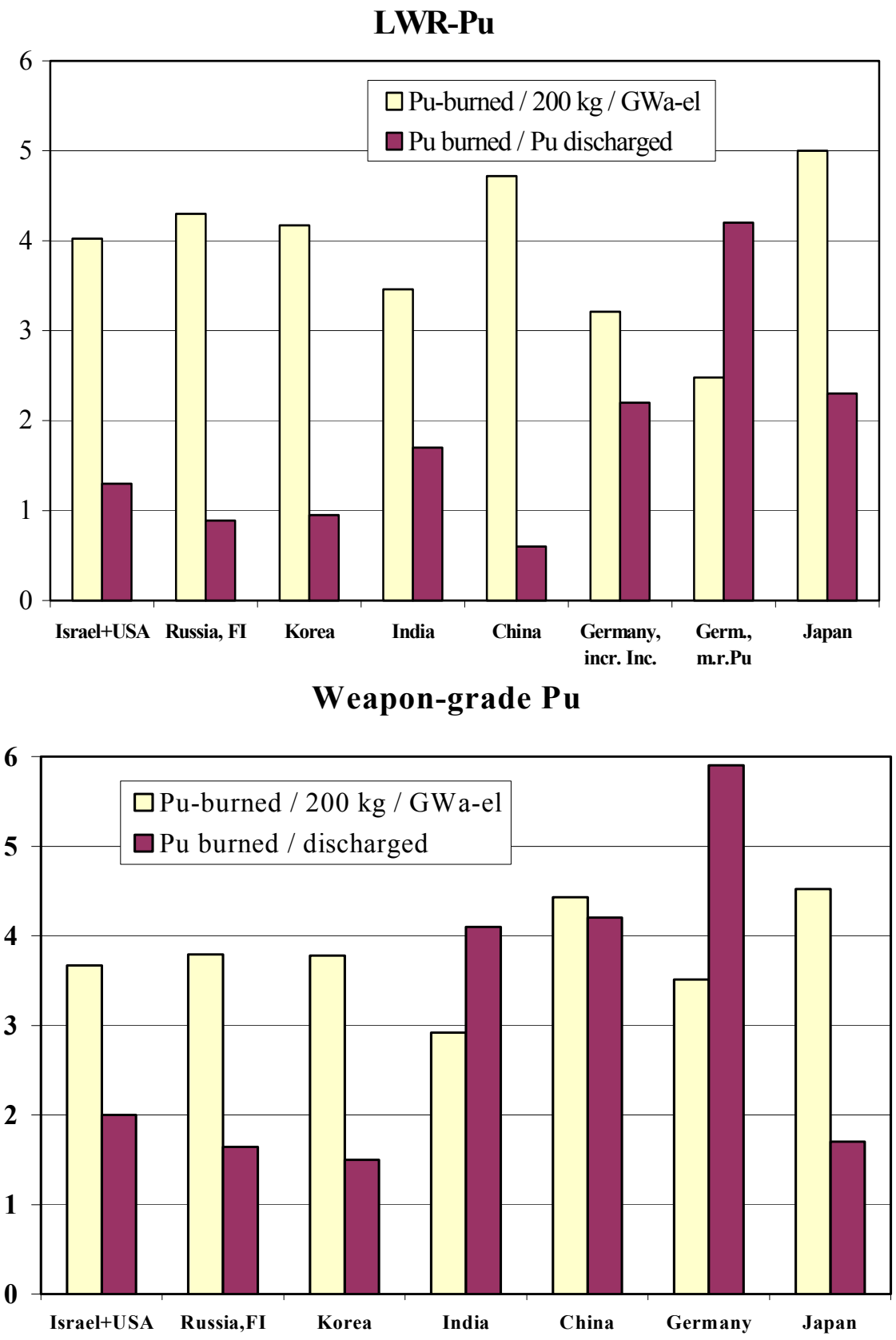

Fig. 1: Burning LWR- and Weapons-grade Plutonium

The plutonium incineration rate (ordinates of Fig. 1) is given in units of 200 kg / GW_{el} a, which is (very roughly) the plutonium amount produced by current LWRs. In other words, this number indicates how many units of LWRs could get rid of their spent plutonium with the help of one unit of the considered plutonium-burner. It is concluded that

- Incinerating LWR-plutonium:
  Water-cooled reactors (LWR and HWR), having a relatively low heavy-metal burnup (40-50 GWd/to), reach a large plutonium-incineration rate in the range of about 700 –850 kg/GW_{el} a. On the other hand, this amount of incinerated plutonium is small compared to that remaining for disposal or reuse at the end of burnup (Pu burned / Pu-discharged equals 0.8 – 1.7, which corresponds to a residual plutonium fraction in the range of 56% to 37%). Achieving a large heavy metal burnup (e.g., in HTR-fuel), results in a smaller amount of incinerated plutonium per unit of produced energy (500 – 650 kg/GW_{el} a), but distinctly reduces the fraction of residual plutonium down to 19 %.

- Incinerating weapons-grade plutonium:
  The higher neutronic value of weapons-grade plutonium and the strongly reduced build-up of minor actinides out of ^{242}Pu generally makes weapons-grade plutonium incineration more effective than for LWR-grade plutonium. While the amount of incinerated weapons plutonium per unit of produced electric energy is comparable to the incineration rate of LWR-plutonium, the quantity of residual plutonium can be strongly reduced in case of weapons-grade plutonium. The ratio "Pu burned / Pu-discharged" equals 1.5 to 2 (40% to 33% residual plutonium fraction) for LWRs, about 4 (20% residual plutonium fraction) for the HWR and up to 6 (14% residual plutonium fraction) in case of the HTR. Here, the advantage of reactors having high burnups, becomes obvious once more.

3. EFFECT OF PLUTONIUM-INCINERATION ON THE RADIO-TOXICITY OF DISPOSED NUCLEAR WASTE

3.1 INCENTIVES AND DATA BASE

The research summarized in chapter 2 primarily aims at the minimization of the proliferation risk by minimizing the plutonium-production and maximizing the plutonium-incineration. The question still remains, whether and to which degree the incineration of plutonium furthermore is an appropriate tool to significantly reduce the hazard potential of the nuclear waste, which in the end remains for final disposal.

The most common procedure in order to assess the radio-toxicity of nuclear materials is based on the recommendations of the *International Commission on Radiological Protection*, defining "Annual Limits on Intake" for the radiotoxic isotopes. Recent recommendations are given in Sv/Bq and are called "Dose Coefficients for Intake (DCI)". It was agreed to use the values according to *ICRP Publications 68 /3/ and 61 /4/* as the common data base within the frame of this CRP. A comparison between the waste of uranium-fuelled LWRs (providing no

reprocessing of the discharged fuel), on the one hand, and the waste produced in a scenario applying plutonium incinerating reactors, on the other hand, helps to assess the effect on the radio-toxicity.

3.2 RADIO-TOXICITY BENCHMARK

In order to ensure that the computer codes used in the different countries have been correctly updated with regard to the boundary conditions indicated in chapter 3.1, the first step of this evaluation was a benchmark calculating the radio-toxicity of the spent fuel resulting from the one-year operation of a 1 GW_{el} reference-PWR. The composition of unloaded heavy metal isotopes is defined in Table I.

Table I: Discharge rate of heavy metal isotopes (kg/year) for a typical PWR

U-234	4.51 E 00
U-235	2.70 E 02
U-236	1.07 E 02
U-237	2.70 E -01
U-238	2.69 E 04
Np-237	1.11 E 01
Np-239	2.27 E 00
Pu-238	3.12 E 00
Pu-239	1.43 E 02
Pu-240	5.78 E 01
Pu-241	3.11 E 01
Pu-242	1.02 E 01
Am-241	8.87 E -01
Am-242m	1.66 E -02
Am-242	2.12 E -03
Am-243	1.77 E 00
Am-244	6.41 E -05
Cm-242	2.38 E -01
Cm-243	5.17 E -03
Cm-244	4.56 E -01
Cm 245	1.66 E -02
Σ	2.75 E 04

All data normalized to the electric power output
1000 MW_{el} and 300 full-power days

The task to be commonly performed consisted in evaluating:

- The ingestion hazard of the complete heavy metal waste
- The inhalation hazard of the complete heavy metal waste
- The ingestion hazard of the heavy metal waste remaining after separation of 99 % of all plutonium-isotopes.
- The inhalation hazard of the heavy metal waste remaining after separation of 99 % of all plutonium-isotopes.

Dose coefficients of Intake (DCI) for the heavy metal isotopes and for the fission products, to be commonly used are given in Table II.

Table II: Dose coefficients of intake (DCI)

Unit: **"Sv / Bq "**
NUCL: Isotope identification number =Z*10000 + W*10 + IS, being
Z: the atomic number
W: the atomic weight
IS: equal 0 or 1 for ground or metastable state, respectively.
DCI-W: DCI-value for water
DCI-A: DCI-value for air
References:
ICRP Publication 68 (1994)
ICRP Publication 61 (1991), DCI calculated for reference dose 20 mSv / a

NUCL	DCI-W	DCI-A	NUCL	DCI-W	DCI-A	NUCL	DCI-W	DCI-A
20040	0.E+00	0.E+00	882250	1.E-07	6.E-06	942360	9.E-08	2.E-05
812070	5.E-10	5.E-09	882260	3.E-07	2.E-05	942380	2.E-07	4.E-05
812080	5.E-10	5.E-09	882280	7.E-07	3.E-06	942390	3.E-07	5.E-05
812090	5.E-10	5.E-09	892250	2.E-08	8.E-06	942400	3.E-07	5.E-05
822060	0.E+00	0.E+00	892270	1.E-06	6.E-04	942410	5.E-09	9.E-07
822070	0.E+00	0.E+00	892280	4.E-10	3.E-08	942420	2.E-07	4.E-05
822080	0.E+00	0.E+00	902270	9.E-09	1.E-05	942430	9.E-11	1.E-10
822090	6.E-11	3.E-11	902280	7.E-08	4.E-05	942440	2.E-07	4.E-05
822100	7.E-07	1.E-06	902290	5.E-07	1.E-04	942450	7.E-10	7.E-10
822110	2.E-10	6.E-09	902300	2.E-07	4.E-05	952410	2.E-07	4.E-05
822120	6.E-09	3.E-08	902310	3.E-10	4.E-10	952421	2.E-07	4.E-05
822140	1.E-10	5.E-09	902320	2.E-07	4.E-05	952420	3.E-10	2.E-08
832090	0.E+00	0.E+00	902330	5.E-10	5.E-09	952430	2.E-07	4.E-05
832100	1.E-09	8.E-08	902340	3.E-09	7.E-09	952440	5.E-10	2.E-09
832110	9.E-10	3.E-08	912310	7.E-07	1.E-04	952450	6.E-11	8.E-11
832120	3.E-10	4.E-08	912320	7.E-10	1.E-08	962420	1.E-08	5.E-06
832130	2.E-10	4.E-08	912330	9.E-10	4.E-09	962430	2.E-07	3.E-05
832140	1.E-10	2.E-08	912341	5.E-10	5.E-09	962440	1.E-07	3.E-05
842100	2.E-07	3.E-06	912340	5.E-10	6.E-10	962450	2.E-07	4.E-05
842110	9.E-10	3.E-08	922320	3.E-07	4.E-05	962460	2.E-07	4.E-05
842120	9.E-10	3.E-08	922330	5.E-08	9.E-06	962470	2.E-07	4.E-05
842130	9.E-10	3.E-08	922340	5.E-08	9.E-06	962480	8.E-07	1.E-04
842140	9.E-10	3.E-08	922350	5.E-08	8.E-06	962490	3.E-11	5.E-11
842150	9.E-10	3.E-08	922360	5.E-08	8.E-06	962500	4.E-06	8.E-04
842160	9.E-10	3.E-08	922370	8.E-10	2.E-09	972490	1.E-09	2.E-07
842180	9.E-10	3.E-08	922380	4.E-08	7.E-06	972500	1.E-10	1.E-09
852170	9.E-10	3.E-08	922390	3.E-11	4.E-11	982490	4.E-07	7.E-05
862190	3.E-11	7.E-11	922400	1.E-09	8.E-10	982500	2.E-07	3.E-05
862200	3.E-11	7.E-11	932360	2.E-08	3.E-06	982510	4.E-07	7.E-05
862220	3.E-11	7.E-12	932370	1.E-07	2.E-05	982520	9.E-08	2.E-05
872210	9.E-10	3.E-08	932380	9.E-10	2.E-09	982530	1.E-09	1.E-06
872230	2.E-09	1.E-09	932390	8.E-10	1.E-09	982540	4.E-07	4.E-05
882230	1.E-07	7.E-06	932401	5.E-10	5.E-09	992530	6.E-09	3.E-06
882240	7.E-08	3.E-06	932400	8.E-11	1.E-10			

On this basis the benchmark definition is:

- *Calculate the total radio-toxicity of the given heavy metals and of their daughter products for a decay period ranging from 10^0 years through 10^6 years.*
- *Exclude the bulk of plutonium and its daughter products by a reduction of all initial plutonium-isotopes by the factor 0.01. Calculate the remaining radio-toxicity.*

The results of the benchmark are displayed in Fig.2 and Fig. 3. The ingestion hazard and the inhalation hazard, respectively, resulting from the given initial isotope mixture, is plotted as a function of the decay time. The agreement between the participants is quite satisfying except that Israel and USA generally evaluate somewhat higher radio-toxicity values for a decay time of about 10^6 years and longer. However – as can be seen from the figures – this fact does not appear to be important for the assessment of plutonium-separation, because the impact of plutonium and its daughter nuclides on the radio-toxicity of the waste tends to vanish at such a decay time, anyway.

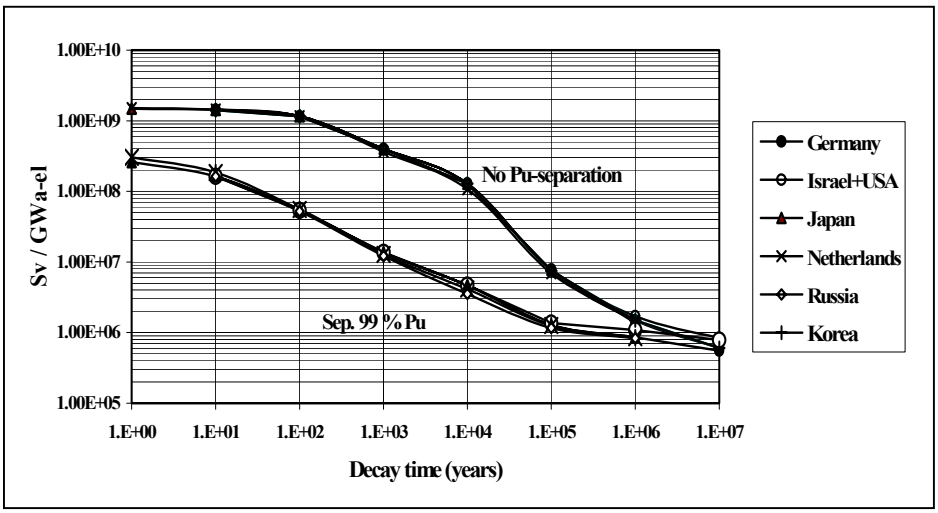

Fig. 2: Benchmark: Ingestion hazard of the Heavy metal isotopes

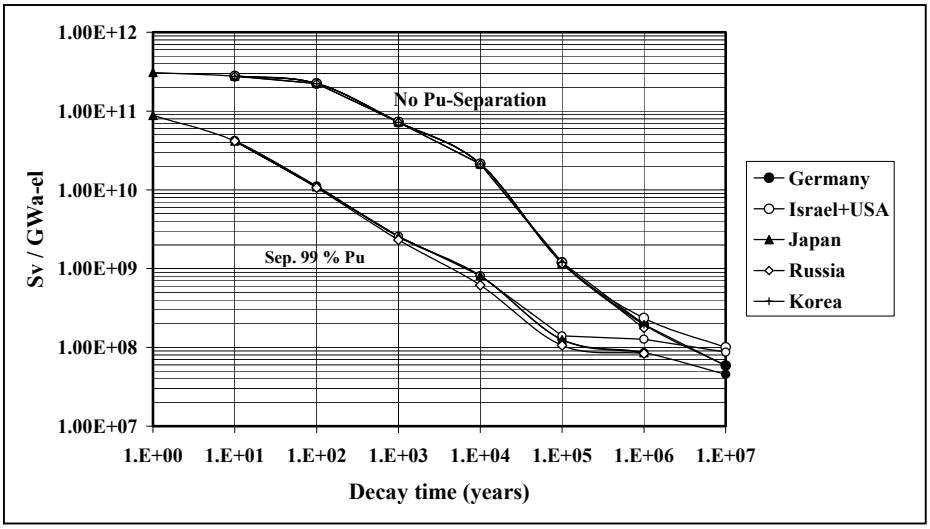

Fig. 3: Benchmark: Inhalation hazard of the heavy metal isotopes

3.3 POSSIBLE REDUCTION OF THE WASTE RADIO-TOXICITY

3.3.1 Procedure for the assessment

In the realistic scenario of the worldwide population of nuclear reactors we are faced with an increasing number of reactors producing plutonium at an increasing yearly amount. Once a decision will be made, to build plutonium-incinerators, the yearly rate of incinerated plutonium would have to increase with time, too. In order not only to compensate future production of plutonium but to also reduce current stockpiles, the number of incinerators would have to increase as rapidly as possible until an equilibrium between plutonium-production in uranium-fuelled reactors, on the one hand, and plutonium-burning in thorium / plutonium-fuelled reactors, on the other hand, could be reached. Switching over from uranium to thorium in as many existing reactors and as soon as possible could limit the necessary number of plutonium incinerators.

Future scenarios are hard to evaluate. So we restrict ourselves to a relatively simple model, comparing two alternative reactor scenarios:

Scenario 1:
Assume a given population of which we name "conventional reactors"(e.g. typical PWRs). These are all operated by use of uranium fuel, and their waste is disposed without separation of any isotopes. These reactors produce a certain, yearly amount of radio-toxicity, which we measure in $Sv/GW_{el}\,a$.

Scenario 2:
Alternatively, we assume a nuclear scenario of the same size (in GW), where we have an equilibrium combination of "conventional", uranium fuelled reactors and of a certain fraction of "plutonium-burners" using thorium based fuel. The principle mass flow of the fuel is illustrated in Fig. 4:

The bulk waste of "conventional" reactors is disposed and it produces a certain, yearly amount of radio-toxicity (Tox. (a)). 99 % of the unloaded plutonium, however, are separated to be used as feed fissile material (or part of it, respectively) of the "plutonium-burner". The spent fuel of these burners is finally disposed, representing another yearly amount of radio-toxicity, Tox. (b). We add the amounts of radio-toxicity disposed from both the "producers" and the "burners", normalized to 1 GWa_{el}.

The sharing of the power production between the two reactor types depends on the amount of plutonium, which the respective incinerator (according to each country's proposal) is able to load each year:

be

PDR: the plutonium-discharge rate of one "conventional reactor, commonly assumed to be equal 245 kg/GWa_{el} in this study,

PCR: the plutonium-charge rate of the regarded plutonium incinerator,

X: at equilibrium, the power share of the plutonium-producer,
Y: at equilibrium, the power share of the plutonium-incinerator, Y=1-X

Then it follows (see Fig. 14):

$$X = \frac{PCR}{PCR + PDR}$$

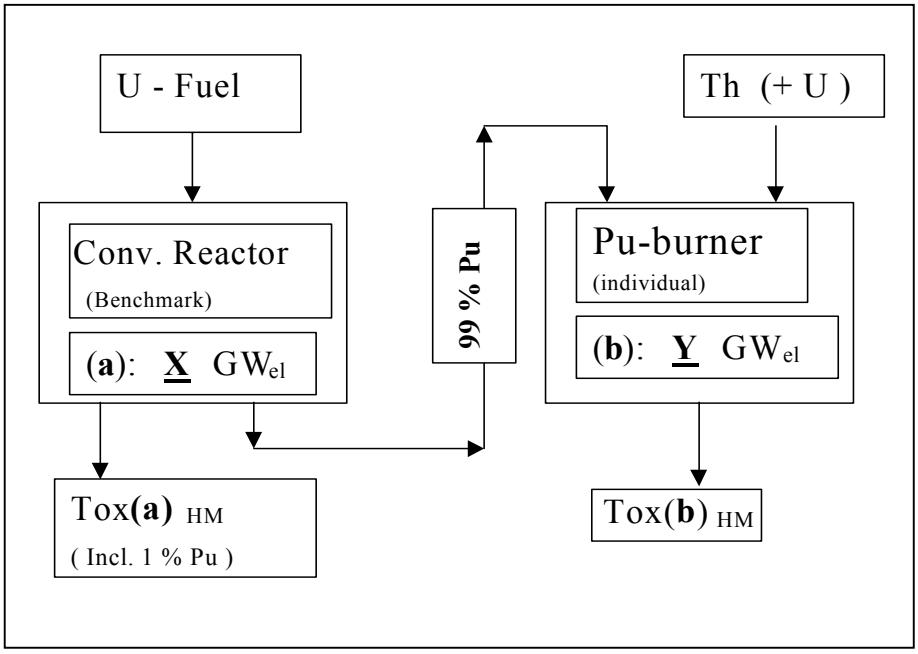

Fig. 4: Symbiosis of plutonium-producers and plutonium-burners

The comparison of the radio-toxicity produced by this combined system of reactors (Tox.(a) + Tox.(b)) to the radio-toxicity produced by scenario 1 is an indication of the potential of plutonium incineration with the help of thorium fuelled reactors, not only in view of the proliferation concern, but also in order to reduce the long-term waste radio-toxicity.

3.3.2 Results and conclusions, stage 3

Fig. 5 and 6 elucidate the principal impact of plutonium-incineration on the amount and on the time dependence of the waste radio-toxicity. Here, the German results (HTR, increased incineration) have been taken as an example; all other contributions show similar characteristics:
A certain reduction of the radio-toxicity occurs at a decay time of the waste between some 10^2 and 5 x 10^4 years as the consequence of the strongly reduced amount of plutonium and of its

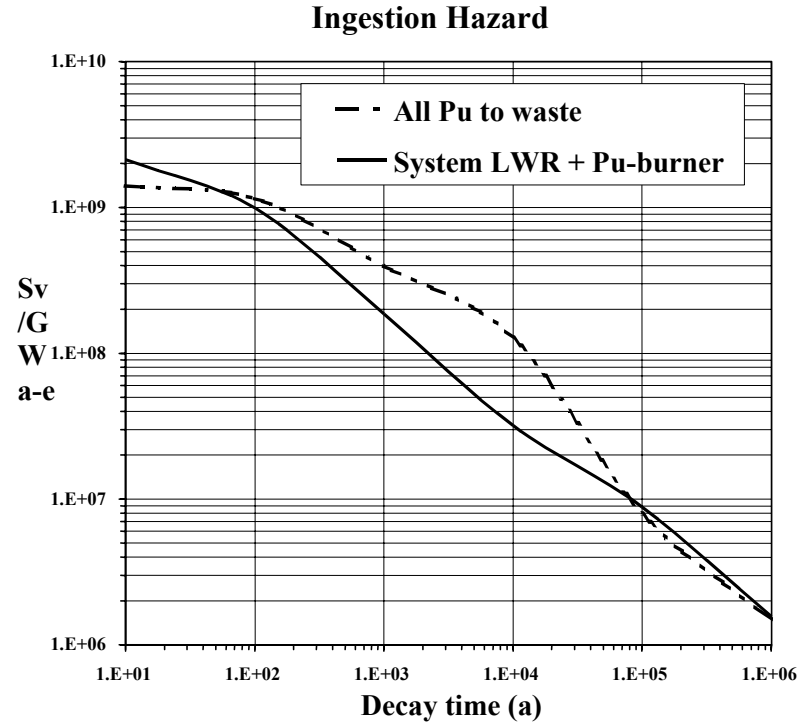

Fig. 5: Comparison: Ingestion hazard of heavy metal waste with and without Pu-incineration (Example, German burner variant)

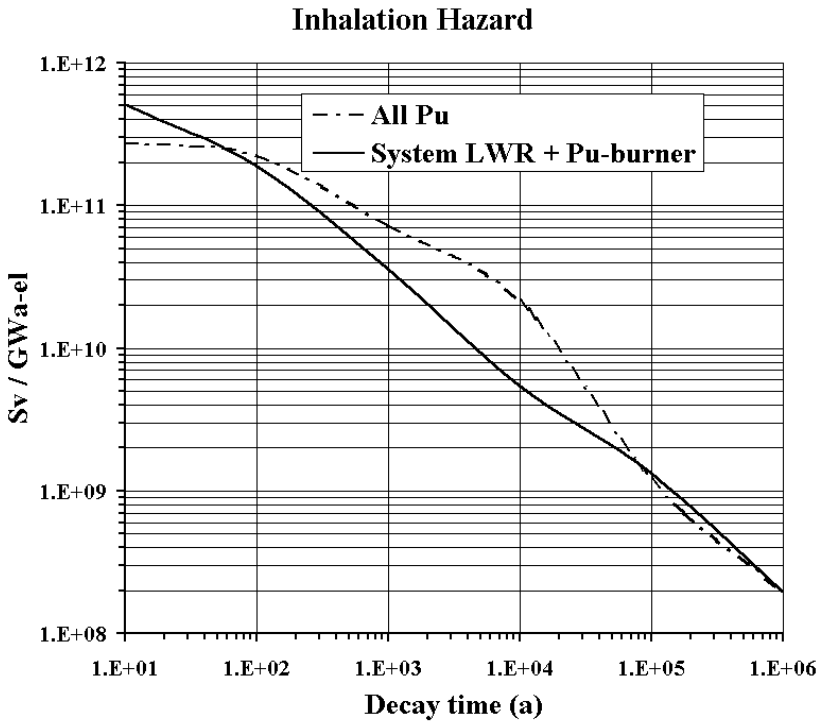

Fig. 6: Comparison: Inhalation hazard of heavy metal waste with and without Pu-incineration (Example, German burner variant)

daughter nuclides in the disposed waste. However, during the first two decades, the radio-toxicity of the heavy metal waste is even increased due to the highly toxic Minor Actinides, which are produced by neutron capture in ^{242}Pu (since plutonium is recycled). This is true for the ingestion hazard (Fig.5) as well as for the inhalation hazard (Fig. 6).

Fig. 7 and Fig. 8 give a comparison of the investigation results, which have been worked out by the participating countries, making use of their individual, favored reactor concept for plutonium-incineration. A "Decontamination-factor (DF)" is plotted versus the decay time. The factor DF is equal to the radio-toxicity of the heavy metal waste, which appears after the (partial) incineration of the LWR-plutonium in the various burner concepts, divided by the radio-toxicity resulting from direct disposal of the heavy metal waste.

With the exception of the common results of Israel and USA, the results of all participants show the same principal effects:

- For the first decades after disposal, the radio-toxicity of the waste is increased (up to a factor ≈ 2). It is obvious, that the initial increase of the radio-toxicity is the more distinct, the higher the heavy metal burnup of the plutonium-incinerator (particularly HTR and MSR) is.

- The radio-toxicity is also increased at decay times larger than 10^5 years (up to a factor ≈ 2).

- It is decreased for the period between about 10^2 years and about 0.5-1.0 x 10^5 years by at maximum the factor 2 to 4. Here, the high burnup applied in the HTR causes by far the strongest reduction.

A reduction of the waste radio-toxicity by an order of magnitude or more seems not to be achievable by any of the considered concepts.

4. OVERALL CRP CONCLUSION

In the course of the IAEA CRP, participants from different Member States performed three benchmark tasks for different reactor concepts (benchmarks 1 and 2 see /1/). The results obtained are very satisfactory and deemed to constitute a sufficiently reliable basis for overall conclusions on the potential of thorium-based fuel cycles to constrain plutonium and to reduce the long-term potential radiotoxic hazard of the waste.

An assessment of thorium fuelled thermal reactors in view of their potential for the utilization of plutonium and for a possible, combined reduction of the waste radio-toxicity has been performed. The analyses looked at the utilization, on the one side, of first generation reactor-grade plutonium, i.e., plutonium typically discharged from current reactors (LWRs), and, on the other side, of weapons-grade plutonium. Plutonium utilization may be looked at from the point of view of two main optimization goals: firstly, to achieve a large incineration rate in relation to the amount of electricity produced, and, secondly, to minimize the amount of

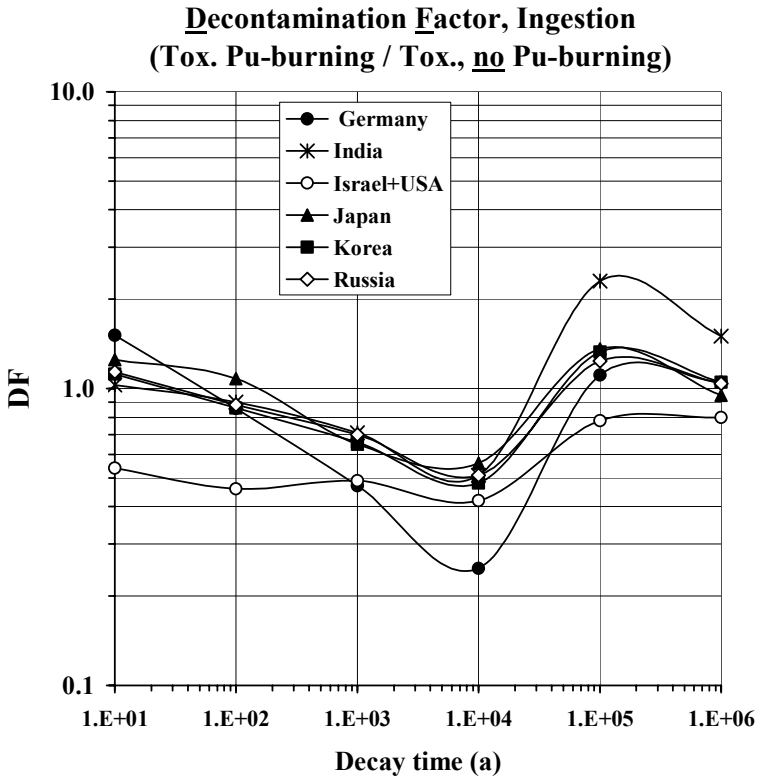

Fig. 7: Relative change of the ingestion hazard of heavy-metal waste achieved by Pu-incineration in the various burner concepts

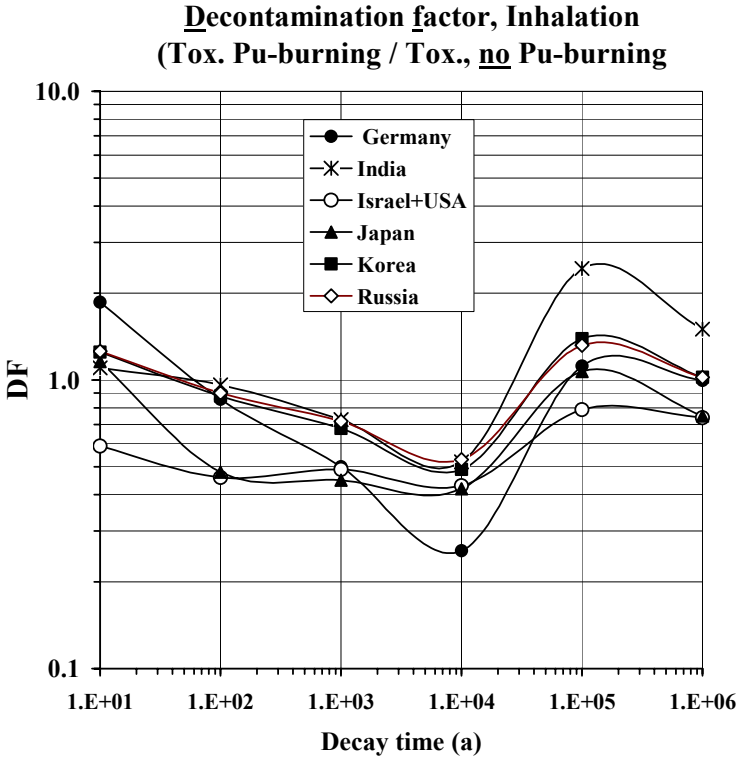

Fig. 8: Relative change of the inhalation hazard of heavy-metal waste achieved by Pu-incineration in the various burner concepts.

plutonium, which is still residual in the discharged fuel elements after their use in a plutonium incinerating reactor. For each of the two plutonium types, the overall conclusions to be drawn with regard to both optimization goals mentioned above can be summarized as follows:

(a) LWR plutonium:
Water-cooled reactors (LWR and HWR), characterized by a relatively low heavy metal burnup, attain a large plutonium incineration rate. On the other hand, the amount of plutonium incinerated is low compared to the remaining plutonium inventory in the spent fuel (to be disposed off or recycled). Achieving a large heavy metal burnup (e.g., in the HTR case) results in a smaller amount of plutonium incinerated per unit of produced energy, but distinctly reduces the fraction of residual plutonium. Typically, one plutonium incinerating reactor burns approximately 2.5 to 4 times the amount of plutonium which is produced by an LWR of the same power.

(b) Weapons-grade plutonium:
The higher neutronic value of weapons-grade plutonium, and the strongly reduced build-up of minor actinides starting from ^{242}Pu, generally makes its incineration more effective than in the case of LWR plutonium. While the amount of weapons-grade plutonium, which is burned per unit of produced electricity, is comparable to that of LWR plutonium, the quantity of residual plutonium in the spent fuel assemblies can be strongly reduced in the case of weapons-grade plutonium. In view of the minimization of the amount of plutonium remaining for final disposal, there is a clear advantage for reactors having an especially high heavy metal burnup.

Summing up, there is a remarkable potential to effectively constrain the production of plutonium and to reduce existing plutonium stockpiles, by implementing the thorium fuel cycle in a large number of current reactors. This path offers a promising near-future plutonium management solution in view of, e.g., the proliferation concerns linked to plutonium. However, plutonium incineration in thermal reactors turns out to be less effective from the point of view of the reduction of the long-term radio-toxicity of the nuclear waste. A reduction by an order of magnitude or more of the potential long-term radiotoxic hazard of the waste seems not to be achievable by any of the considered plutonium incinerating thermal reactors. Most of the calculations performed for LWR plutonium indicate that the waste radio-toxicity will be decreased by not more than a factor of 2 to 4, and only for the period between approximately 10^2 to 10^5 years after disposal. The waste radio-toxicity is even increased during the first decades and for extremely long times after disposal.

5. References

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