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**INTERNATIONALE  
LÄNDERKOMMISSION  
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Baden-Württemberg · Bayern · Hessen



## **ILK Statement**

**on Reprocessing of Spent Fuel Elements**

**Für deutsche Fassung bitte umdrehen!**

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The International Committee on Nuclear Technology (Internationale Länderkommission Kerntechnik, ILK) was established by the three German states of Baden-Württemberg, Bavaria and Hesse in early October 1999. It consists of ten scientists and experts from Germany, France, USA, Sweden and Switzerland. The ILK acts as an independent and objective advisory body to these German states on issues related to the safety of nuclear facilities and radioactive waste management and the risk assessment of the use of nuclear power. In this capacity, the Committee's goal is to contribute to the maintenance and further development of the high, internationally recognised safety standards of nuclear power plants in the southern part of Germany.

Since the beginning of 2001, the ILK has concerned itself with the advantages and disadvantages of the reprocessing of spent fuel elements. In its consideration of this topic, the ILK has also paid attention to the possible future alternatives such as multiple recycling and the transmutation of long-lived fission products and actinides. The conclusions and recommendations of the ILK on reprocessing are presented in the following statement which was adopted on the 14th ILK meeting on November 12<sup>th</sup> 2001 in Strasbourg.

The Chairman



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## Executive Summary

The fact that not only U-235 but also U-238 and Pu-isotopes with uneven mass numbers are valuable fuels resulted in early nuclear energy scenarios incorporating the strategy of a combined use of thermal and fast reactors and reprocessing of spent fuel.

Especially the consistently low costs of natural uranium and the higher costs of fabricating mixed oxide (MOX) fuel elements made single reprocessing of spent uranium oxide (UOX) fuel and the use of MOX in light water reactors (LWR) less attractive and led to a general preference for LWRs.

In Germany, Sec. 9a of the Atomic Energy Act, until 1994, accorded reprocessing of spent fuel (safe recycling) priority over direct disposal (managed disposal). Against this background, the German electricity utilities in the mid-70s signed reprocessing contracts with the French COGEMA and the British BNFL. New reprocessing contracts were concluded between the German electricity utilities and COGEMA and BNFL in 1989/90 after discontinuation of the Wackersdorf reprocessing project. Unlike the old contracts, these new ones allowed the German electricity utilities to be flexible in setting the time and quantity of fuel elements to be reprocessed.

Since the 1994 amendment to the Atomic Energy Act, direct disposal of spent fuel has been legally accepted as an equivalent alternative to reprocessing. The current German federal government intends to limit the management of spent fuel to direct disposal. In the agreement between the federal government and the electricity utilities of June 14, 2000, transports for reprocessing are only allowed until July 1, 2005. This planned ban on reprocessing is to be laid down in the amendment to the Atomic Energy Act.

The current statement compares the radiological impact of the nuclear fuel cycle using direct disposal exclusively on the one hand with single, complete reprocessing of spent uranium fuel elements on the other. Furthermore, the impact of reprocessing on the nuclear fuel cycle costs and the consequences for interim storage and final disposal are discussed. Proceeding from these considerations and with reference to possible future alternatives such as multiple recycling and the transmutation of long-lived fission products and actinides, the ILK arrives at the following conclusions and recommendations concerning reprocessing of spent fuel elements:

Both direct disposal and single reprocessing with subsequent final disposal of the vitrified high-level waste from reprocessing and of spent MOX fuel elements are feasible and can safely be implemented. In particular, it can be shown that repro-

cessing does not lead to significantly higher radiation exposures either of persons occupationally exposed or members of the public than the once-through use of fuel. Moreover, there are no scientific findings calling into doubt that also MOX fuel elements can be safely emplaced in final repositories over very long periods.

Direct final disposal and reprocessing are characterized by specific advantages and disadvantages. Arguments favoring reprocessing, according to the current state of the art, are these:

- Even reprocessing all UOX fuel elements once and using plutonium in MOX will save about 20% of the natural uranium used, compared to direct disposal, for the same amount of energy, which can make a substantial contribution towards resource conservation in terms of sustainability.
- The amount of plutonium arising for final disposal after reprocessing of all UOX fuel elements is reduced by 30%, with the isotope vector of the plutonium contained in the spent MOX improving the resistance of the material to unauthorized use.
- The uranium derived from reprocessing, which may well become an even more valuable nuclear fuel in the future than today, does not need to be emplaced in a final repository.
- The total lower quantities of fissile material to be finally disposed of further reduce the already very small probability of recriticality in the final repository.
- The quantity of high-level waste (HLW) arising from reprocessing of spent UOX fuel is reduced by a factor of 5 compared to direct disposal, and further reductions are possible. However, low and medium-level waste arises during reprocessing which must also be disposed of in a geological repository. Consequently, the balance of waste volumes does not show any advantages for the reprocessing option.

The main arguments against reprocessing include:

- Reprocessing currently engenders fuel cycle costs 10 - 18% higher than those associated with direct disposal. However, it should be noted that the fuel cycle costs only constitute one third of the total energy production costs, and that rising world market prices of natural uranium will further reduce the cost advantage of direct disposal.

- Larger quantities of minor actinides (MA) are produced when MOX is used. However, these minor actinides are not very mobile in the chemical environment of a geological repository.
- Handling separated reactor grade Pu in the fuel cycle entails additional safeguards measures. In contrast to the surveillance of fissile material for a geological repository, these measures are required for a comparatively short time period only until other technologies can replace the use of nuclear fission for power generation.

The radiation exposures stemming from final disposal are determined mainly by a few long-lived fission products, such as Se-79, Zr-93, or I-129. Comparable quantities of these long-lived fission products requiring transfer to a final repository are produced in both alternatives. Accordingly, no significant differences between direct disposal and reprocessing will arise in terms of the radiation exposures resulting from final disposal.

On the whole, the ILK takes the view that there are no convincing technical and economic arguments for or against reprocessing at the current state of the art. A ban on reprocessing would, however, very probably impede research in Germany in the field of partitioning and transmutation of long-lived radionuclides.

This radiochemistry and reactor physics research towards an advanced nuclear fuel cycle is gaining strongly in international significance and can only be conducted on a long-term basis and with sufficient lead time ahead of technological developments. It primarily serves the following purposes:

- Improving fuel efficiency.
- Avoiding or destroying Pu and MA, as well as
- Transmutating long-lived fission products.
- Reducing waste volumes, or
- Further improving confinement of the radiotoxic inventory in a final repository as long as transmutation into stable or short-lived nuclides is not feasible on technical or economic grounds.

It thus ultimately serves to reduce further the radiation exposure to be expected in the long term.

Reprocessing spent nuclear fuel for recycling in LWRs does not represent a suitable means of reducing significantly the radiotoxic inventory of waste because MA are produced by neutron capture in U-238 and in the Pu isotopes. This applies equally to single and multiple recycling in LWR.

The radiotoxic inventory in the waste stream and the potential radiation exposures from final disposal can only be diminished, and the threat of recriticality in final repositories can only be excluded completely, by reprocessing with advanced separation and transmutation technologies. This will require a combined use of thermal and fast reactors and, possibly, also subcritical accelerator-driven systems.

In the light of these potential achievements of long-term research, the ILK recommends that German research institutions participate in international research on reprocessing and transmutation (partitioning & transmutation) to a greater extent than in the past and that this work receives appropriate national support.

Furthermore, the ILK notes that these discernible future developments do not justify delaying the final disposal of radioactive waste in Germany. Even partitioning and transmutation will not achieve complete destruction of all radioactive waste for technical or economic reasons. Instead, they could very greatly reduce waste quantities, shorten the necessary confinement periods, and improve chemical adaptation of the technical barriers of the repository to the different species of radionuclides. In this regard, final repositories will also be indispensable in the future even though the strict geological requirements placed on such a repository could then be lowered.

Considering that these advanced fuel cycles may become available, the ILK recommends a final disposal that allows for the retrieval of radionuclides and fuels for the purposes of transmutation or a further utilization during the so-called operating phase of the final repository (50 - 60 years).

## 1 Introduction

Uranium fuel used in light water reactors (LWR), with an initial enrichment of 3.4% U-235 and after a burn-up of 45 GWd/tHM (tHM: tons of heavy metal), still contains 0.52% U-235; 0.013% U-234, 0.46% U-236, 1.05% Pu; 0.094% of minor actinides (MA); 4.62% fission products, and 93.247% U-238<sup>1</sup>.

The spent U-fuel elements are reprocessed for recovery of the uranium and plutonium as nuclear fuels for thermal reactors. The plutonium is then fed into light water reactors in U-Pu mixed oxide (MOX) fuel with an initial enrichment of 6 - 7% Pu-tot (total plutonium), i.e. 4 - 5% Pu-fiss (fissile plutonium).

The uranium derived from reprocessing (REP U) is stored on the premises of the reprocessing plants. In principle, it can be transferred to final disposal in a suitable form. The residual amounts of U-235, however, can also reenter the fuel cycle via the enrichment procedure in the fabrication of fresh uranium fuel. Some countries, including France, UK and Russia, fabricate fuel elements from reprocessed uranium (REP-U fuel).

Currently, spent MOX fuel elements are not reprocessed but are stored in interim storages and are earmarked for later final disposal. In the single reprocessing of all uranium fuel elements at a medium burn-up rate of 40 GWd/tHM, approx. 20% of the initially employed uranium can be saved compared to the fuel cycle without reprocessing.

In Germany, Sec. 9a of the Atomic Energy Act until 1994 gave priority to reprocessing of spent fuel (safe recycling) over direct disposal (managed disposal). Against this background, the German electricity utilities in the mid-70s signed reprocessing contracts with the French COGEMA and the British BNFL. The quantities of material to be reprocessed under these so-called old contracts are shown in Table 1 [1]. Furthermore, new reprocessing contracts were concluded between the German electricity utilities and COGEMA and BNFL in 1989/90 after discontinuation of the Wackersdorf reprocessing project. Unlike the old contracts, these new ones left the German electricity utilities flexibility in determining the time and quantity of fuel to be reprocessed. However, these contracts also established minimum quantities of material to be reprocessed (see Table 1).

<sup>1</sup> Calculations by TÜV Süddeutschland Bau und Betrieb GmbH using the CASMO code for fuel from pressurized water reactors

	Old contracts (tHM)			New contracts (tHM)		Total (tSM)
	COGEMA	BNFL	THORP	COGEMA	BNFL	
Facility	UP 2	UP 3	THORP	La Hague	THORP	
Contracted	1.541	3.111	884	1.127 <sup>1</sup>	302 <sup>1</sup>	6.965
Reprocessed by- Dec. 31, 1999	1.541	2.483	56	0	0	4.080

Table 1: Reprocessing contracts of the German electricity utilities with COGEMA or BNFL [1].

Since the 1994 amendment to the Atomic Energy Act, the direct disposal of spent fuel has been regarded by law as an equivalent alternative to reprocessing. The current German federal government intends to limit the management of spent fuel to direct disposal. According to the agreement between the federal government and the electricity utilities of June 14, 2000 [2], transports for reprocessing are only permitted until July 1, 2005; quantities delivered may be reprocessed. This planned ban on reprocessing is to be laid down in the pending amendment to the Atomic Energy Act. The federal government justifies the ban by arguing that "the reprocessing of nuclear fuels leads to increased radioactive immissions and to additional accrual of plutonium". It also states that "it creates further risks in the reprocessing facilities in addition to posing a fundamental proliferation risk and makes transports necessary that might otherwise be superfluous" [3].

Proceeding from the radiation impacts of reprocessing, a discussion of fuel costs and the consequences for interim storage and final disposal as well as an outline of future alternatives, such as multiple recycling and transmutation of long-lived fission products and actinides, the ILK takes the following stand on a possible ban on reprocessing in Germany.

<sup>1</sup> Agreed minimum quantities

## 2 Radiological Impact of Reprocessing

The Nuclear Energy Agency of the OECD (NEA) published a comparative study of the radiation impacts of various fuel cycles, comparing direct disposal, on the one hand, and single reprocessing to MOX of all spent uranium fuel elements, on the other hand [4]. In the following, the essential boundary conditions of the study and its results are outlined in terms of occupational radiation exposure as well as the radiation exposure of the public.

The study proceeds from a scenario of a 1000 MWe pressurized water reactor (PWR) and a burn-up of 40 GWd/tHM. For the non-reprocessing option (once-through cycle), the entire spent fuel ends up in the repository for high-level, heat-generating waste, while the reprocessing option results in vitrified waste from reprocessing and spent MOX fuel to be stored in this geological final repository.

The study covers all major steps of the fuel cycle, ranging from mining and milling of the uranium ore to fuel fabrication (including conversion to  $UF_6$  and enrichment), to using the fuel in a nuclear power plant, and to reprocessing.

The uranium derived from reprocessing is assumed not to be processed into fuel, and the tailings of uranium mining and milling to UOC (uranium ore concentrate) are taken to be stabilized over long periods of time so that the emanation of Rn-222 in particular can be permanently kept at a low level. The reprocessing technique chosen is the PUREX process (Plutonium-Uranium Recovery by EXtraction).

The recommendations by the International Commission on Radiological Protection, ICRP (ICRP 1991, Publication 60) [5], constitute the basis for the radiological assessment of occupational radiation exposure and the radiation exposure of the public. The radiation doses received are described as annual individual doses [Sv/a] for occupationally exposed persons or representatives of the most highly exposed groups of the population and as energy-specific collective doses in relation to 1 GWa of electricity [manSv/GWa]. The collective doses are calculated by summation of the effective radiation doses arising over 500 years after release of the activity associated with the production of 1 GWa in a single year.

Individual doses must be viewed against the dose limits recommended by the ICRP of 20 mSv/a over a five-year average for occupationally exposed persons, or 1 mSv/a for the most highly exposed group of the public. These reference levels, in turn, can be compared to the annual mean individual dose to the world population of 2.4 mSv/a due to natural background radiation.

The limitation of the accumulation period to 500 years stems from the consideration that radiation exposures are dominated in any case by the release of the long-lived volatile nuclides C-14 (half-life 5,730 a) as a consequence of energy production and reprocessing, and of Rn-222 with the precursor nuclide Th-230 (half-life 77,000 a) following uranium mining. An extension of the observation period would not significantly change the radiological relations among the various fuel options while introducing further uncertainties into the generic assessments. There are grounds for assuming that this period of 500 years will see no releases worth mentioning from the geological repositories. Releases of long-lived fission products from the final repository only begin to play a role after several 1000 years. Within the framework of the study, these releases therefore are not significant in terms of either magnitude or time period.

It can be noted that only a few radionuclides make noteworthy contributions to the collective dose of the population. There is C-14, on the one hand, which is released during energy generation in nuclear power plants and, in combination with I-129 and Kr-85, during reprocessing. On the other hand, all nuclides of the uranium decay chain associated with the mining and milling of uranium ore should be considered. The main exposures can be traced to the inhalation of Rn-222 and, in the critical group, the ingestion of the long-lived Rn-222 daughter products, Pb-210 and Po-210. The radiation exposure of occupationally exposed persons is dominated by Co-60. Co-60 arises from the activation of steel corrosion products in the reactor water.

The following detailed radiation exposures for the different fuel options with and without reprocessing are estimated, based on the generic analyses of the study or statistical surveys of effective radiation doses in specific plants of the fuel cycle:

Occupational exposures:

- The average measured effective individual doses lie below the ICRP reference levels for all stages of the fuel cycle. Workers in nuclear power plants receive the highest exposures. These exposures do not depend on the use or non-use of MOX.
- Also for energy-specific collective doses, the highest contributions of 1.0 - 2.7 manSv/GWa result from energy generation in nuclear power plants. Differences between fuel strategies with and without MOX do not exist.
- Collective doses from reprocessing and vitrification of high-level waste (HLW) are comparatively low at 0.014 manSv/GWa.
- However, fuel fabrication in a cycle with single reprocessing gives rise to higher collective doses of 0.094 manSv/GWa than direct disposal only (0.007 manSv/GWa).

- This difference is not fully compensated by dose reductions due to the approx. 20% reduction in uranium quantities to be mined, milled and enriched in the reprocessing cycle.
- The exposures induced by transports of spent fuel and waste are negligible in every respect.
- According to the study, the specific collective doses of occupationally exposed persons in sum lie between 1.04 – 2.93 manSv/GWa without reprocessing and between 1.14 – 2.99 manSv/GWa with single reprocessing.

Radiation exposure of the general public:

- The mean annual individual doses in the critical population groups of 0.4 – 0.5 mSv/a lie below the ICRP reference level of 1 mSv/a for all stages of the fuel cycle. The highest individual doses result from uranium mining and milling (0.3 – 0.5 mSv/a) and from reprocessing (0.4 mSv/a).
- The energy-specific collective doses attain levels of 1.2 manSv/GWa for reprocessing and are on the same order of magnitude as those derived from uranium mining and milling (1.0 manSv/GWa without reprocessing, 0.8 manSv/GWa with reprocessing).
- According to the generic study, the (site-specific) mean collective doses due to energy generation in nuclear power plants reach a level of 0.6 manSv/GWa.
- All other contributions from transport, interim storage and similar sources are negligible.
- The study indicates a cumulated specific collective dose to the population of 1.6 manSv/GWa for the option without reprocessing and of 2.6 manSv/GWa with reprocessing.

In summary, it is safe to say that, for both options, radiation exposure is low compared to the ICRP reference levels and to natural background radiation. The differences between the two options are not significant. In particular, they are masked by uncertainties in the assumptions made on the emanation of Rn-222 (3 Bq/m<sup>2</sup>s over 10 – 15 years) from the tailings of uranium ore mining and ore milling. Insufficient containment of the tailings can increase the Rn-release by one order of magnitude.

### 3 Interim Storage and Final Disposal of Spent Fuel

The radiological consequences of single reprocessing and using MOX on wet or dry interim storage of spent fuel on nuclear power plant sites or also in centralized interim storage facilities (as practiced, for example, in Sweden) have been considered in the dose levels mentioned above. The effective doses resulting from interim storage of spent nuclear fuel, on the whole, are very low and the differences between UOX and MOX are not significant.

The higher levels of heat generated by spent MOX fuel compared to uranium fuel do not pose any problem in interim storage. After all, heat generation is dominated by the fission products immediately after unloading from the reactor. The inventory of fission products in MOX and UOX fuels is almost identical for the same burn-up (e.g. fission product inventory at a burn-up of 55 GWd/tHM: UOX fuel –  $1.0 \times 10^{18}$  Bq/tHM at unloading,  $8.1 \times 10^{15}$  Bq/tHM 40 years after unloading; MOX fuel –  $1.0 \times 10^{18}$  Bq/tHM at unloading,  $6.6 \times 10^{15}$  Bq/tHM 40 years after unloading).

The aim pursued worldwide in the management of spent fuel and of medium and high-level waste from reprocessing is final disposal in deep geological formations [6]. However, no geological repository is currently in operation for these types of waste [7]. At the moment, there is no unanimous opinion worldwide on the necessary duration of safe confinement of spent fuel and HLW or on whether the fuel should be emplaced in the repository so as to be retrievable or not. International discussions mention safe confinement periods of up to  $10^6$  years and even longer.

However, a repository evidently must satisfy the following criteria:

- Radiation exposures must be kept low enough over the periods mentioned above to remain low compared to the natural radiation background.
- The repository must be designed and operated in a way preventing unauthorized access to the fissile material emplaced.
- The spent fuel must be emplaced in a way practically excluding criticality in the repository.

The following facts about reprocessing are important from a radiological point of view and with regard to minimizing the fissile material to be emplaced:

Reprocessing 1 tHM of UOX fuel with a burn-up of 33 GWd gives rise to 955 kg of uranium, 10 kg of Pu (all isotopes), and approx. 2 m<sup>3</sup> of vitrified or cemented waste. The uranium can be used in fabricating REP U-fuel or can be placed into a final repository. The residual quantities of U-235 also can be recycled in the enrichment step

in the fabrication of UOX fuel from natural uranium. The decay of U-232 ( $\alpha$ ,  $T_{1/2} = 71.7$  a) and U-234 ( $\alpha$ ,  $T_{1/2} = 2.47 \times 10^5$  a) in uranium separated from reprocessing results in the radiotoxic decay products, Th-228 ( $\alpha$ ,  $T_{1/2} = 1.9$  a) and Tl-208 ( $\beta$ ,  $T_{1/2} = 3.1$  min) when that uranium is emplaced in a final repository. The final disposal of uranium from reprocessing thus not only would represent a waste of valuable nuclear fuel but, at the same time, add to the radiotoxicity and the fissile material inventory of the repository. This observation, of course, in principle also applies to the direct disposal of UOX fuel.

The plutonium separated is processed into MOX. A typical MOX fuel element for a pressurized water reactor contains approx. 35 kg Pu. Seven UOX fuel elements must be reprocessed to produce this quantity. A spent MOX fuel element still contains some 25 kg Pu [8]. With regard to radiotoxicity no benefit can be seen in single reprocessing and using MOX in LWR, as the radiotoxic inventory of spent MOX can be 6 to 8 times higher than that of UOX [8]. Nuclide balances very much depend on the fuel management strategy, i.e. on burn-up and on the local neutron spectrum, among other factors.

The fission products and minor actinides (MA) reach the repository in a vitrified or cemented or compacted form. It is estimated that the amount of material containing the radiotoxic inventory can be reduced by a factor of five by recycling uranium fuel as MOX with current reprocessing technologies. As far as the volume of all waste, including low and medium-level waste, is concerned, the current ratio can be assumed to be close to 1:1. It is stated in [9] that the entire waste volume is some 1.54 times greater for reprocessing than for direct disposal. On the basis of data provided by COGEMA, Hicken and Keßler [10] mention a ratio of 1.2 for the 1992 state of the art and of <0.81 in 2000. Future optimization of reprocessing and waste conditioning is expected to further reduce masses and volumes.

Spent MOX fuel elements are currently not being reprocessed. For the same burn-up, the  $\alpha$ -activity of MOX, compared to uranium fuel 10 years after unloading from the reactor, is about ten times higher, and still four times higher after 100,000 years. The fact that the heat generation of MOX after an interim storage period of 40 years is about 2 – 3 times greater than in spent uranium fuel is also relevant to final disposal. In particular, under the assumption of a maximum permissible temperature in the surroundings of the waste containers in the final repository, the number of waste containers for MOX would have to be up to 3 times higher than for uranium fuel.

On the whole, it can be said that the peculiarities of MOX fuel as a waste form are not as well known as for uranium fuel. However, there are currently no scientific indications that the final disposal of MOX fuel elements might lead to problems in

the final repository. The content of transuranic elements (Pu, Np, Am, Cm...) which is greater by a factor of between 3 and 4 and which dominate the radiotoxic inventory for very long time periods over several 100,000 years does not pose a special difficulty for the final disposal of MOX. According to the current state of knowledge, the transuranic elements are relatively immobile in deep geological formations compared to long-lived fission products. This favors the effective confinement of these nuclides [11].

This explains why the radiation exposures are typically determined by long-lived fission products such as I-129, Cs-135, Se-79 and Tc-99 over very long time periods. Contributions to the annual individual dose from the Np-decay chain worth mentioning only result after about 100,000 years.

Radiological studies [12 - 14] on the whole show very low radiation exposures. Studies of a typical scenario for the Gorleben salt dome [12] demonstrate that individual dose rates do not exceed a level of 0.7 mSv/a for an equivalent emplacement quantity of 73,000 tHM. Radiation exposures from the iodine inventories emplaced dominate up to a disposal period of 3000 years; up until 100,000 years, the main doses are contributed by Se-79. Subsequently, contributions from the Np decay chain dominate. After 200,000 years, the Np decay chain, on the one hand, and the combined effects of Tc-99 and Cs-135, on the other hand, make comparable contributions totaling approx. 0.2 mSv/a. After  $10^6$  years, exposures are determined solely by the decay chains of U and Np. At that time, however, a very low level of 0.01 mSv/a has been reached.

Table 2 illustrates that there are no substantial differences between UOX and MOX in terms of the generation of the relevant fission products. Therefore, the radiation exposures resulting from the final disposal of UOX and MOX will not differ considerably. In any case, these exposures are far lower than natural background exposures and recommended ICRP levels.



Nuclide	UOX-fuel (in Bq/tHM)	MOX-fuel (in Bq/tHM)
Cs-135	$2,6 \times 10^{10}$	$4,0 \times 10^{10}$
Tc-99	$7,3 \times 10^{11}$	$7,4 \times 10^{11}$
I-129	$1,9 \times 10^9$	$2,4 \times 10^9$
Se-79	$2,5 \times 10^{10}$	$2,0 \times 10^{10}$
Zr-93	$1,1 \times 10^{11}$	$7,5 \times 10^{10}$

Table 2: Contents of long-lived fission products in UOX and MOX fuels (burn-up 55 GWd/tHM)

Multiple recycling of LWR fuel, i.e. reprocessing of spent MOX, is possible in principle in the current PUREX process. The spent MOX fuel is dissolved in nitric acid together with spent uranium fuel in order to adhere to the process constraints with regard to dose rate limits and Pu concentration. However, recycling is possible for a limited number of times only, as Pu isotopes accumulate in the MOX which cannot be burnt in the thermal spectrum of a LWR or are highly radioactive (e.g. Pu-238, an  $\alpha$ -emitter with a tendency towards spontaneous fission). Moreover, more Pu is bred continuously as a result of neutron capture in the U-238 in the MOX employed in the reactor. Higher actinides are also bred via neutron capture in the entire Pu inventory in MOX. This makes the destruction of plutonium in LWR very inefficient from a radiotoxic point of view. A decisive reduction of radiotoxic inventories in the repository and of possible radiation exposures ensuing from final disposal can only be achieved by applying advanced partitioning and transmutation techniques (see Sec. 5).

That the amount of fissile material reaching a repository can be decreased by reprocessing and by using MOX is a positive fact as far as criticality safety is concerned, and also makes unauthorized access to such inventories less attractive. The change in the plutonium isotope vector also impedes any misuse of this fissile material.

The uranium separated in reprocessing can be used for other purposes, as outlined above. This includes its possible utilization in a future advanced fuel cycle with the combined use of thermal and fast reactors. Studies by G. Keßler et al. [15, 16] additionally show that complete reprocessing of spent uranium fuel and MOX utilization in LWRs (so-called 22 GWe scenario in Germany) allows the amount of plutonium to be disposed of in repositories to be reduced by 30% compared to exclusive direct disposal (3.1 t/a instead of 4.7 t/a). On the other hand, separated reac-

tor-grade plutonium accumulates in reprocessing which also needs to be safeguarded against unauthorized access. In contrast to fissile material safeguards for a geological final repository, these measures require safeguarding only for a comparatively short time period until other technologies can replace the use of nuclear fission for power generation.

The safeguards problem and questions of physical protection will not be dealt with in detail in this ILK Statement. However, it should be noted that safeguards issues are clearly regulated in Germany since Germany, with all its relevant nuclear facilities, is subject to international nuclear materials safeguards (EURATOM and IAEA) [17]. This also applies to the nuclear installations in France and the UK where German spent nuclear fuels are reprocessed.

The danger of mild nuclear explosions in the repository has been addressed in the technical literature [18 – 20]. Two scenarios are typically discussed. In the first scenario, fuel is emplaced in the repository that has not yet reached full burn-up, and water enters the final disposal containers. Such a scenario can be prevented by selecting configurations that cannot go critical even in case of water ingress, or by lining the containers with neutron-absorbing materials. In the second scenario, water entry into the repository is again assumed. As a possible consequence, the waste disposal containers would be destroyed and Pu-239 - or its decay product, U-235 - would be transported mainly as colloids in the aqueous phase. Transport velocities could be several 10 m/a [21]. Another assumption is that the colloids accumulate in waterlogged cracks, fissures and pores of the repository host rock formation, thereby potentially leading to a critical configuration.

The risk of recriticality in the repository and an associated mild nuclear explosion is estimated to be extremely low for all cases [22, 23].

#### 4 Cost Comparison between Direct Disposal and Single Reprocessing

The economic viability of the two options, direct disposal and single reprocessing, has been compared in various studies since the mid-80s.

In the study by the Nuclear Energy Agency of the OECD dating from 1994 [24], the costs for the entire fuel cycle (including costs of natural uranium, conversion, enrichment, and fuel element fabrication) were considered. Accordingly, a pressurized water reactor with an electric power of 1390 MWe (mean burn-up of the fuel elements: 42.5 GWd/tHM) offers 10% cost advantage for direct disposal over reprocessing. The study concludes that, with underlying uncertainties taken into account, such as the price of natural uranium and the costs of final disposal, the slight difference in costs between direct disposal and reprocessing must be considered insignificant.

The 1995 study by the Energiewirtschaftliches Institut (Institute for Energy Economics) of the University of Cologne [25] shows that direct disposal is the more economical option, even under specific German conditions, under various fuel management strategies for a 1300 MWe LWR. This applies when only considering the spent fuel management costs and also when referring to the entire fuel cycle. According to that study, the cost advantage of direct disposal is around 18% when the entire fuel cycle is considered, and remains constant even if uncertainties are taken into account.

On the whole it can be stated that these comparisons of economic performance are associated with major uncertainties as many detailed cost items to be considered, such as the prices of natural uranium and the costs of final disposal, depend on developments which cannot be estimated accurately. Furthermore, the relative cost advantage of direct disposal loses some of its significance as a consequence of fuel cycle costs constituting only roughly one third of the entire electricity generating costs. Electricity generating costs, in turn, only make up some 15% of the electricity rate to be paid by the individual consumer, while the costs of grid use amount to some 45%, and value added tax and eco-tax account for 25% of the price of electricity [18]. At the present time, however, the stable low uranium fuel costs, the high burn-up attainable, as well as the higher fabrication costs of MOX fuel elements make reprocessing of spent uranium fuel and the use of MOX fairly unattractive to the utilities.

#### 5 Partitioning and Transmutation (P&T)

Natural uranium only contains 0.72% of the U-235 isotope that can be split by thermal neutrons. U-238 can only be split by fast neutrons. Neutron capture and the production of Pu-239 primarily occur in the range of thermal energies. Most trans-uranium elements can be split by fast neutrons; Pu-239 and Pu-241 have high fission cross-sections also in the thermal energy range. Large isotope fractions of the major actinides thus constitute valuable fuels. Because of these facts of physics, earlier nuclear power scenarios, as a matter of principle, proceeded from a joint use of thermal and fast reactors and from reprocessing of spent fuel. The use of U-238 in fast reactors alone could improve by a factor of 50 the energy yield of the uranium mined compared to thermal reactors. The development of fuel costs and the policy of strict non-proliferation of fissile materials over the past few decades have led to a preference for light water reactors and direct disposal of spent fuel.

In 1988, the Japanese government founded the OMEGA (Options for Making Extra Gains from Actinides and Fission Products) initiative and suggested the participation of other OECD countries in related R&D work. The OECD/NEA thus set up an information exchange program (Programme of Technical Information Exchange) to support and promote that international cooperation in the field of P&T. Within the framework of that initiative, the OECD/NEA in 1999 published the report referred to above, "Actinide and Fission Product Partitioning and Transmutation" [8]. Amongst other things, the report presents a concept of an advanced fuel cycle (The Advanced Fuel Cycle with TRU {transuranium} Recycling [AFC]). The report specifically outlines the R&D projects necessary to implement such an AFC. The current state of research was presented in Madrid at the 6th Information Exchange Meeting 2000 on Actinide and Fission Product Partitioning and Transmutation [26].

In any case, the essence of the AFC lies in high-performance reprocessing of spent fuel achieving high standards for partitioning Pu and U, MA and fission products. The concept is also based on the combined use of LWR and fast reactors (FR) and, possibly, accelerator-driven systems (ADS). Some of the concept's cornerstones are outlined below.

- Reprocessing LWR-UO<sub>2</sub> fuel and separation of MA as well as long-lived fission products from the liquid HLW:

The customary technique of reprocessing LWR fuel in use today is the so-called PUREX process. It involves dissolving the spent fuel in nitric acid (HNO<sub>3</sub>) and separating U and Pu by liquid-liquid extraction. Tri-n-butylphosphate (TBP) is used as the solvent [27]. The partitioning rates achieve theoretical levels of

99.9%; in current practice, they reach 99%. Most of the MA are transferred to the liquid HLW. For Am and Cm and the higher actinides with shorter half-lives ( $T_{1/2}$ ) this amounts to a proportion of >99.5%.

Np-237, in contrast, due to different oxidation levels, appears both in the waste stream and in the U-Pu product stream [27]. A clear allocation is desirable, since (as indicated previously) an excessive Np fraction in MOX would lead to radiological problems during multiple recycling because of the production of Pu-238 ( $\alpha$ ,  $T_{1/2} = 86.4$  a) which arises from irradiation of Np-237 in the LWR. A clear allocation to one of the material streams is possible by modifying the PUREX process, and has already been demonstrated on a laboratory scale.

New procedures for separating MA from liquid HLW are currently under development worldwide (TRUEX, DIDPA, and other processes compatible with PUREX). Non-aqueous processes are being investigated in Japan, amongst other places.

A problem thus far has been the removal of Am and Cm from the waste stream. Because of their chemical similarity, these nuclides always appear together with the lanthanides. However, the lanthanide quantity is 10 to 20 times greater than that of Am/Cm. As the lanthanides include strong neutron absorbers, separation of the Am/Cm fraction from the lanthanides is necessary for later transmutation of Am/Cm. This requires sophisticated separation chemistry. Analyses of different transmutation scenarios with varying combinations of LWR and fast breeders [28] all have shown the transmutation of Cm to be absolutely essential to achieve high reduction factors for radiotoxicity in the waste stream. Transmutation of all MA, including Cm, achieves reduction factors of 70 - 110 in the first  $10^4$  years. These levels drop to 7 - 25 without transmutation of Cm. After  $10^4$  years, the influence of Cm transmutation on radiotoxicity is minimal.

In order to reduce potential radiation exposures associated with the emplacement of HLW in the repository, it is particularly important to separate long-lived fission products that are mobile in the geosphere.

Tc-99 in its metallic and oxidic state is an insoluble residue in liquid waste. It is soluble in liquid waste when it appears in the form of a pertechnetate-anion ( $TcO_4^-$ ). The separation of dissolved Tc has been mastered, whereas the separation of Tc from the insoluble residues using pyrochemical procedures is difficult because of its similarity with the platinum metals. Further research is necessary.

I-129 predominantly appears in the washing liquids of the dissolver in the PUREX process. It can be retained in special adsorbents or discharged into seawater. Radiologically, it is one of the problematic nuclides in final disposal.

Zr-93 and Cs-135 each occur with other isotopes (e.g. Cs-137) that are highly radioactive. At the same time, the isotope fractions of Cs-135 and Zr-93 are very small. Thus, isotope separation would have to follow chemical separation if these isotopes are to be transmuted, by irradiation in a reactor, as manageable targets embedded in a suitable matrix material. This seems hard to achieve economically.

- Reprocessing LWR MOX:

The PUREX process in principle can be applied also in reprocessing LWR MOX. Some of the problems arising have been described in Section 3. The separation of MA from the liquid HLW is further aggravated by the high alpha activity caused mainly by Am and Cm. This high alpha activity causes radiolysis of the process fluids. This explains why the storage of LWR-MOX for 50 years or more in an interim storage before reprocessing is being considered

It must be repeated that multiple recycling in LWR is not efficient and, moreover, requires the addition of fresh Pu from LWR  $UO_2$ .

- MOX fabrication for fast reactors (FR MOX):

FR MOX with Pu shares of up to 25% is commercially available, though only for Pu mixes from LWR fuel with low burn-up. At current burn-ups, the mix proportions shift in favor of Pu-238 and Pu-242. These isotopes undergo spontaneous fission and are  $\alpha$ -emitters. This makes them both neutron sources and heat sources and impedes handling fresh FR MOX. In so-called homogeneous recycling (admixture to the fuel), FR MOX would also contain fractions of higher actinides. Am-241 and Cm-244, in particular, would cause more problems due to the further rise in gamma and neutron radiations.

- Manufacturing MA targets for irradiation in LWR and fast reactors:

Heterogeneous irradiation of MA is preferable to homogeneous irradiation. The irradiation targets could be manufactured in small technical facilities with special gamma and neutron shieldings. Making such targets on an industrial scale has not yet been solved.

- Development of "inert" matrix materials for fuels (IMF fuels):

To increase the net burn-up of actinides in the irradiation of fuel in LWR, and in order to reduce the generation of more actinides, materials are being developed which are free from U-238 and have low cross sections for reactions with neutrons. This prevents breeding of transuranium elements on the basis of U-238. Materials currently under study include spinel ( $MgAl_2O_4$ ), yttrium oxide ( $Y_2O_3$ ), and yttrium-aluminium garnet ( $Y_3Al_5O_{12}$ ). Metals, silicon carbide, and nitride fuels are also studied with respect to their radiation behavior and effects on reactor dynamics. In thermal reactors, feedback characteristics are changed substantially by the absence of U-238. The Doppler coefficient, which characterizes the negative feedback on reactor power via the fuel temperature due to neutron capture in U-238, becomes substantially lower in absolute terms. Some matrix materials also have thermal conductivities inferior to those of UOX, thereby impeding heat transfer to the coolant and causing a tendency towards higher fuel temperatures.

The physical and chemical properties of such IMF materials are currently under study in international research programs [29 - 31].

- Fuel use and target irradiation in fast reactors and accelerator-driven systems (ADS):

Experience in the operation of fast breeder reactors cooled by liquid metal is available worldwide. It can also be used in the development and operation of fast burner reactors. In France, the CAPRA program (*Consommation Accrue de Plutonium dans les Rapides*) for burning Pu, and the SPIN program (*Séparation – Incinération*) for burning MA in such a burner, have been conducted since 1992. Similar programs exist in Japan and the USA.

Higher burn-ups than in critical fast reactors can be reached in accelerator-driven systems partly because of the possible higher concentrations of MA (including those with high neutron capture cross sections) and because of the more variable neutron spectrum. Such systems are currently being investigated within the 5th Framework Program of the EU. In this context, especially the concept of the Energy Amplifier [32] developed by C. Rubbia has become known. It is based on the generation of neutrons by spallation of heavy nuclei with high-energy protons from an accelerator. The spallation target as a neutron source is located in a subcritical blanket consisting of fuel and, possibly, of the MA targets. Converting an ADS into a pilot system requires further research and development in accelerator technology, fluid flow engineering (cooling of the proton window), structural materials (stainless

steel and PbBi coolant), and the physical design of the whole system.

Recently, critical and subcritical molten-salt-cooled reactors have been reconsidered for transmutation.

- Transmutation of long-lived fission products:

Effective transmutation is much more difficult to achieve for long-lived fission products than for actinides. The reason lies in the very low transmutation cross sections. Transmutation can only be achieved in a meaningful way in thermal high-flux reactors or accelerator-driven systems (thermal flux around  $10^{18}n/cm^2*s$ ). For Tc-99, transmutation half-lives are around 30 years at best. For I-129, the transmutation process becomes even more difficult since the gaseous irradiation product, Xe-130, must be discharged from the targets. The transmutation of Zr-93 and Cs-135 furthermore requires as a precondition the separation of these isotopes from the other Zr and Cs isotopes.

- Reprocessing spent FR fuel:

Reducing the radiotoxic inventory requires multiple recycling of FR fuel (FR MOX and metal fuels). Burn-ups (up to 180 GWd/tHM) are two or three times higher in fast reactors than in thermal reactors. After 150 GWd/tHM, only some 15 - 20% of the transuranium elements have been burnt up.

Using the PUREX process or a similar aqueous method of reprocessing would require 10 - 12 years of interim storage of the spent fuel in order for the radiation power and afterheat to have subsided sufficiently. A total of 100 - 250 years would be necessary to achieve burn-ups of 90% for the transuranium elements. This is why pyrochemical molten-salt technologies are being considered for reprocessing. The fused salt ( $CdCl_2$ , LiCl, KCl) is much more resistant to high temperatures and alpha radiation. These processes are currently being developed in Japan and other places.

In sum, it can be stated that an advanced fuel cycle can lead not only to an improved fuel utilization but also to a reduction of the long-lived radiotoxic inventory in the final repository and to the decrease in possible radiation exposures. The required safe confinement periods can, under certain circumstances, be shortened to  $10^3$  years by the transmutation of actinides and long-lived fission products [27]. The safety of a repository is achieved and demonstrated much more easily with confinement periods in the order of  $10^3$  years than for  $10^6$  years or even longer. Over a period of  $10^3$  years, confinement could probably be assured by technical barriers only.

Advanced separation techniques would also offer the possibility of chemically designing technical barriers in such a way that optimum retention of the species to be confined would be guaranteed.

Exploiting this potential requires the acceptance of reprocessing. However, it should also be mentioned that the radiotoxic inventory circulating in such an AFC is substantially higher than in the concept currently used. Depending on the specific transmutation scenario, i.e. the mix of LWR and fast breeders, the equilibrium actinide contents (in relation to the generated electric power), i.e., when there is no further net production of actinides, are 6 to 10 times higher than in LWR with UOX fuel [28, 33].

## 6 Conclusions and Recommendations

- The fact that not only U-235 but also U-238 and Pu-isotopes with uneven mass numbers are valuable fuels resulted in early nuclear energy scenarios incorporating the strategy of a combined use of thermal and fast reactors and reprocessing of spent fuel.

Especially the consistently low costs of natural uranium and the higher costs of fabricating mixed oxide (MOX) fuel elements made single reprocessing of spent uranium oxide (UOX) fuel and the use of MOX in light water reactors (LWR) less attractive and led to a general preference for LWRs.

Both

1. direct disposal and
2. single reprocessing with subsequent final disposal of vitrified high-level waste from reprocessing and of spent MOX fuel elements

are feasible and can safely be implemented. In particular, it can be shown that reprocessing does not lead to significantly higher radiation exposures either of persons occupationally exposed or members of the public than the once-through use of fuel. Moreover, there are no scientific findings calling into doubt that also MOX fuel elements can be safely emplaced in final repositories over very long periods.

- Direct disposal and reprocessing are characterized by specific advantages and disadvantages. Arguments favoring reprocessing, according to the current state of the art, are these:
  - Even single reprocessing all UOX fuel elements and using plutonium in MOX will save about 20% of the natural uranium used, compared to direct disposal, for the same amount of energy, which can make a substantial contribution towards resource conservation in terms of sustainability.
  - The amount of plutonium arising for final disposal after reprocessing of all UOX fuel elements is reduced by 30%, with the isotope vector<sup>1</sup> of the plutonium contained in the spent MOX improving the resistance of the material to unauthorized use.
  - The uranium derived from reprocessing, which may well become an even

<sup>1</sup> Composition of the Pu-Isotope mix

more valuable nuclear fuel in the future than today, does not need to be emplaced in a final repository.

- The total lower quantities of fissile material to be finally disposed of further reduce the already very small probability of recriticality in the final repository.
- The quantity of high-level waste (HLW) arising from reprocessing of spent UOX fuel is reduced by a factor of 5 compared to direct disposal, and further reductions are possible. However, low and medium-level waste arises during reprocessing which must also be disposed of in a geological repository. Consequently, the balance of waste volumes does not show any advantages for the reprocessing option.

The main arguments against reprocessing include:

- Reprocessing currently engenders fuel cycle costs 10 - 18% higher than those associated with direct disposal. However, it should be noted that the fuel cycle costs only constitute one third of the total energy production costs, and that rising world market prices of natural uranium will further reduce the cost advantage of direct disposal.
- Larger quantities of minor actinides (MA) are produced when MOX is used. However, these minor actinides are not very mobile in the chemical environment of a geological repository.
- Handling separated reactor grade Pu in the fuel cycle entails additional safeguards measures. In contrast to the surveillance of fissile material for a geological repository, these measures are required for a comparatively short time period only until other technologies can replace the use of nuclear fission for power generation.
- The radiation exposures stemming from final disposal are determined mainly by a few long-lived fission products, such as Se-79, Zr-93, or I-129. Comparable quantities of these long-lived fission products requiring transfer to a final repository are produced in both alternatives. Accordingly, no significant differences between direct disposal and reprocessing will arise in terms of the radiation exposures resulting from final disposal.

- On the whole, the ILK takes the view that there are no convincing technical and economic arguments for or against reprocessing at the current state of the art. A ban on reprocessing would, however, very probably impede research in Germany in the field of partitioning and transmutation of long-lived radionuclides.
- This radiochemistry and reactor physics research towards an advanced nuclear fuel cycle is gaining strongly in international significance and can only be conducted on a long-term basis and with sufficient lead time ahead of technological developments. It primarily serves the following purposes:
  - Improving fuel efficiency.
  - Avoiding or destroying Pu and MA, as well as
  - Transmutating long-lived fission products.
  - Reducing waste volumes, or
  - Further improving confinement of the radiotoxic inventory in a final repository as long as transmutation into stable or short-lived nuclides is not feasible on technical or economic grounds.
- Reprocessing spent nuclear fuel for recycling in LWRs does not represent a suitable means of reducing significantly the radiotoxic inventory of waste because MA are produced by neutron capture in U-238 and in the Pu isotopes. This applies equally to single and multiple recycling in LWR.
- The radiotoxic inventory in the waste stream and the potential radiation exposures from final disposal can only be diminished, and the threat of recriticality in final repositories can only be excluded completely, by reprocessing with advanced separation and transmutation technologies. This will require a combined use of thermal and fast reactors and, possibly, also subcritical accelerator-driven systems.
- In the light of these potential achievements of long-term research, the ILK recommends that German research institutions participate in international research on reprocessing and transmutation (partitioning & transmutation) to a greater extent than in the past and that this work receives appropriate national support.

- Furthermore, the ILK notes that these discernible future developments do not justify delaying the final disposal of radioactive waste in Germany. Even partitioning and transmutation will not achieve complete destruction of all radioactive waste for technical or economic reasons. Instead, they could very greatly reduce waste quantities, shorten the necessary confinement periods, and improve chemical adaptation of the technical barriers of the repository to the different species of radionuclides. In this regard, final repositories will also be indispensable in the future even though the strict geological requirements placed on such a repository could then be lowered.
- Considering that these advanced fuel cycles may become available, the ILK recommends a final disposal that allows for the retrieval of radionuclides and fuels for the purposes of transmutation or a further utilization during the so-called operating phase of the final repository (50 - 60 years).

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## 8 List of Abbreviations

ADS	Accelerator-driven system
AFC	Advanced fuel cycle with TRU (transuranium) recycling
CAPRA	Consommation Accrue de Plutonium dans les Rapides (French research program on P&T)
FR	Fast reactor
HLW	High-level waste
ICRP	International Commission on Radiological Protection
IMF	Inert matrix fuel
LWR	Light water reactor
MA	Minor actinides (Np, Am, Cm)
MOX	Mixed oxide
NEA	OECD Nuclear Energy Agency
OECD	Organisation for Economic Co-operation and Development
OMEGA	Options for Making Extra Gains from Actinides and Fission Products (Japanese research program on P&T)
P&T	Partitioning and transmutation
Pu-fiss	Fissile plutonium
Pu-tot	Total plutonium
PUREX	Plutonium-uranium recovery by extraction (reprocessing technique)
PWR	Pressurized water reactor
REP U	Reprocessed uranium
SPIN	Séparation – Incinération Rapides (French research programme on P&T)
$T_{1/2}$	Half-life
TBP	Tri-n-butylphosphate (organic solvent used in the PUREX process)
THORP	Thermal Oxide Reprocessing Plant (reprocessing plant of BNFL at Sellafield)
tHM	Metric ton of heavy metal
UOC	Uranium ore concentrate
UOX	Uranium oxide
UP 2/3	Usine Plutonium N° 2/3 (Reprocessing plant of COGEMA at La Hague)

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**ILK Publications:**

- ILK-01** ILK Statement on the Transportation of Spent Fuel Elements and Vitrified High Level Waste (July 2000)
- ILK-02** ILK Statement on the Final Storage of Radioactive Waste (July 2000)
- ILK-03** ILK Statement on the Safety of Nuclear Energy Utilisation in Germany (July 2000)
- ILK-04** ILK Recommendations on the Use of Probabilistic Safety Assessments in Nuclear Licensing and Supervision Processes (May 2001)
- ILK-05** ILK Recommendation on the Promotion of International Technical and Scientific Contacts of the Nuclear Safety Authorities of the German States (October 2001)
- ILK-06** ILK Statement on the Draft Amendment dating from the July 5 2001 to the Atomic Energy Act (October 2001)
- ILK-07** ILK Statement on Reprocessing of Spent Fuel Elements (November 2001)
- ILK-CD** CD with all presentations held at the ILK Symposium "Opportunities and Risks of Nuclear Power" in April 2001

**In Preparation:**

- Suitability of the Gorleben Site as a Deep Repository for Radioactive Waste
- Protection of Nuclear Power Plants against Airplane Crashes
- Early detection of deteriorating safety in the operation of Nuclear Power Plants
- Evaluation of the sustainability of power generation systems