Feasibility of Vitrification of Plutonium Together with High Active Waste Concentrate and Fabrication of MOX Storage Rods for Direct Final Disposal Instead of a Use of MOX Fuel for Further Handling of Separated Plutonium

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Some 26 to 29 tons of separated plutonium from German spent fuel are still stored as PuO_2-powder. The Oeko-Institute investigated options for the separated plutonium handling in a project done for the city of Hamburg.

Essential requirements for investigation of different methods

The different methods must fulfil a range of requirements in order to be feasible. Each different method must not only propose a valid solution for the final disposal of plutonium but also satisfy every aspect of the feasibility requirements. It is also important that the necessary safeguard measures to determine protection against proliferation are carried out at each stage; the potential for final disposal is of particular importance.

Achieving a state suitable for final disposal of plutonium is a fundamental aspect to be considered whilst assessing methods proposed for the handling of plutonium.

Methods proposed for the handling of plutonium have limited use if they only offer a temporary solution and not a clear definitive solution for its final disposal. The solutions offered must therefore lead to the safe and final disposal of plutonium. One complete method can comprise stages from different methods, after completing these stages, however, the plutonium must be in a form suitable for final disposal and in keeping with the guidelines.

It is imperitive that each stage of a proposed method be either technically feasible, or — due to similarities with methods already in use — can be easily and quickly put into practice. To apply a method already in use and to adapt it for the purpose of handling plutonium should then ensure the technical feasibility of the method. The further development of a current method, when adapted for the handling of plutonium, should (according to experience) present no particular problems to the technical feasibility of a method. To revert back to methods no longer used in nuclear engineering would mean that the requirements are difficult to fulfil; which is disadvantageous if the technical feasibility could not given for one possible method. Safety is an important part of technical feasibility because every stage of a method must fulfil the safety requirements of nuclear engineering. Technical feasibility also includes the authorisation of the plant, since the different technical aspects must be checked, assessed und decided upon.

The question of time must also be taken into consideration:

- How long would the method take, if all available plutonium were to be handled?
- When would the necessary plants be available?
- Where other material is required (HAWC for vitrification for example): is the right amount of the material available at the right time?

The question of economic viabilty is one aspect which can be of good use during the selection of a method. "Safety comes before economy" is a saying that must be respected in nuclear engineering. It is therefore helpful to have an economic assessment of one or more methods. It is in any case necessary to have at least one method assessed for the handling of the excess separated plutonium, because the current situation (storage as separated PO_) cannot continue; the economical criteria may not therefore lead to the exclusion of all methods.

For every method proposed for the handling of plutonium the safety requirements of nuclear engineering must be fulfilled at each stage of the procedure. Included is the question, whether the safety of a method is seen as guaranteed or whether there is still uncertainty. The detailed safety checks are carried out in corresponding licence procedure.

Where certain stages of the method are already being carried out in existing plants, the official safety standards are first approved when the danger potential of the material to be handled is identical to that of a material already approved. The concentration of fissile material (criticality safety) also plays a part here. The official regulations used in approved plants are sufficient when dealing with materials with not only the same radiological danger potential but also the same radiation protection and emission into the environment.

The relevant safety aspects are above all:

- The criticality safety must be guaranteed for every step of the procedure. For every stage before final disposal this is completed by criticality safe design of components, storage and transport containers as well as the usual method (e.g.: limitation of material amount, limitation of the volume of components for handling fissile material). The criticality safety of final dispose requires checks over a longer period of time.

- Necessary transportation must be carried out in approved transport containers that are designed to cope with criticality safety in the case of an emergency. The requirements result from the internationally applied criteria for nuclear transportation. This also means that certain materials should not be transported- in connection with the handling of plutonium, plutonium nitrate solution and HAWC solution. The transportation of liquid solutions of these substances would bring added risks, which for previous transportations of separated fuels from German nuclear power stations was never and should be avoided.

The methods for the handling of plutonium entail many stages. The last step is always the final disposal of the remaining plutonium. The final disposal "suitablity" arises through the combination of final storage and the plutonium-laden product.

For this reason special attention should be paid to the necessary requirements. These are already present due to the general demands for final storage and its long-term safety. The releasing of plutonium into the biosphere must be prevented as far as possible so that even in view of the extremely long timespan no danger could emerge. This requirement is a special case of the general requirement made on final disposal storage sites–keeping the stored radio active product away from the biosphere. One part of the requirement is the correct choice of a suitable storage site with regard to situation and geology. This report doesn't discuss this aspect of the requirements in detail.

There exist other requirements regarding the form of the plutonium to be disposed of permanently. Of particular relevance here is the avoidance of recriticality during final storage. This comes from the possibility that the fissile Isotope of Plutonium (and Uran-235) through a series of events in the final disposal site could result in a configuration, through which a nuclear chain reaction is possible. Therefore, through different specific measures the possibility of a chain reaction must be avoided.

The disposal container must always be criticality safe before it is brought into final storage because the criticality safety is already checked and approved during the previous stages: interim storage, transport, conditioning for final storage handling. With regard to changes of moderation trough the addition of water, the criticality safety must be guaranteed during the phase of early storage.

Concerning the long-term stay in final storage, only additional mechanisms are important, which could lead to the long-term change of concentration and accumulation of fissile material. Such mechanisms can either result from a segregation of the fuel and subsequent concentration of the fission isotopes, or through specific changes in the composition of the isotopes due to radioactive decay. In order to avoid these effects to ensure that no recriticality occurs, additional measures are required. Such measures could include geometrically designed composition of the plutonium storage containers, which are seen as stable enough under final storage conditions. The additional methods could also include thinning of fissile fuel through material which behaves chemically similar (Uranium-238). Other neutron-absorbing materials like Uran-238 would also come into consideration, the chemical similarity must, however, be guaranteed.

Due to its suitability for weapons plutonium especially must be carefully observed in order to avoid proliferation (misuse of civil plutonium for military purposes), or at least to discover it early on. For this control the so-called safeguard measures are used. Inspectors from international organisations (IAEA/Euratom) control the civil nuclear plants using certain methods. The methods must be adapted to the prevailing technical processes in the observed plant and to the proliferation resistance of each handled material containing plutonium. The isotopic composition is not considered because of plutonium's fundamental suitability to weapons on examination of proliferation resistance.

Safeguards are relatively easy as long as countable items containing fissile materials are handled. For fuel, this is done determined by measuring the fissile material content. As long as this element is not dismantled it can be surveyed easily during handling, storage or transportation, because only the methods of containment and surveillance have to be applied. Sealed containers are, in view of safeguards, similar countable "items" as fuel. Interim storage sites are, therefore, reletively easy to survey as during construction safeguard aspects were considered.

One main weakness of the current safeguard concept is exposed through all the so-called "bulk-handling facilities". These are plants where plutonium (and other fission products) are handled in great amounts in their free-flowing separated form; for example, MOX fuel factories and reprocessing plants. As long as the plutonium is packed and kept together, in fuel assemblies for example, it can be accurately counted using the above methods. If, however, material containing plutonium appears in a non countable form (powder, solution, pellets, abrasive dust and other scrap material) accuracy is not guaranteed during counting - up to 1% inaccuracy is unavoidable with every check carried out. The calculated material balance differs then from stock-take to stock-take. This can lead to considerable amounts of material unaccounted for (MUF). Should there appear a MUF greater than zero, the safeguard organisations are faced with the problem of not being able to distinguish between a real proliferation and a statistical inaccuracy. The bigger the inventory handled in a plant, the greater the inaccuracy. This in turn means that the absolute amount of MUF which can be tolerated is greater- due to this inability to distinguish between real proliferation and inaccuracy.

Due to the fundamental problems of balancing as a measurement technique, the proliferation risk of free-flowing plutonium is high. It is therefore preferable to avoid storing plutonium in such forms and to opt for a speedier conversion into countable items (Itemisation).

The proliferation resistance of handled plutonium can only be understood as gradual criterion, since only a more or less incomplete protection against the abuse of plutonium for military purposes can be achieved. In the US debate, the term ”spent fuel standard” was introduced for the assessment of suggestions made for the handling of plutonium after nuclear disarmament. It states that new plutonium should be converted into a form, like plutonium in spent fuel, which achieves protection against access. The radiological barrier – which is built up over a long period of time from the fission products in the spent fuel – is therefore a deciding factor in the attainment of the “spent fuel standard”. Furthermore, technical and economical factors also have to be taken into consideration. It makes no sense, therefore, to go to great expense and convert the plutonium into a form which claims to be safer than spent fuel element (the form in which most of the plutonium worldwide appearsand will be finally disposed of), since there is no safer form. It seems appropriate then to use the “spent fuel standard” as a yardstick for proliferation resistance for reactor plutonium.

Two important facts are to be noted here:

- Firstly, the “spent fuel standard” would only be a safe reference if reprocessing technology were not available. A safe proliferation resistance would then only be achieved on termination of separating the plutonium from fuel and through the main prohibition of operating plants designed for this purpose..
- Secondly, proliferation resistance standard must not be interpreted too narrow or too formal. Storage forms that do not have certain properties of spent fuel can have a much better proliferation resistance than metallic or oxidic plutonium in its pure forms and could be judged as resistant enough.

Even today there is still no operated site in the world suitable as a final storage site for highly radioactive, heat-producing waste, although planning and preparation is being undertaken in many states that use nuclear power. The developments to date show that, according to their policy, heat-producing waste these states are only prepared to build one final storage for highly active waste and not several storages. There are different technical and financial reasons for this:

The final storage must be planned in such a way as to ensure that all highly active heat-producing waste can be stored. Types of waste are as follows:

- Glass containers from the vitrification of highly active fission products (from reprocessing)
- “normal” spent fuel from the usual plants (direct final disposal)
- special fuel; for example, spent MOX fuel, fuel from specially built reactors.

- scrap containing fissile products from fuel production
- storage containers for final storage of plutonium

The above forms of waste, with the exception of the glass containers, show such a high proportion of plutonium that termination of safeguards is not possible. To investigate the nuclear energy programmes of certain states ascertains that the majority of states have already opted for the direct final storage in earlier years and the final storage of spent fuel must be done in any case – examples are Spain, Sweden and Canada. Germany and Belgium, on the other hand, turn to final storage of spent fuel after having reprocessed over a longer period. Even in those few countries using reprocessing in general – there are in practice again special fuels which must be sent for final storage without having been reprocessed.

All in all it means that the proposed final storage sites in each country for highly radioactive waste either contain only material with considerable plutonium content (in the case of direct final storage strategy) or at least partly contain material with considerable plutonium content (in the case of the reprocessing strategy). Under the specific German conditions it is to be expected that approximately 6000-12000 tons of spent fuel for direct final disposal will have to be disposed, depending on the shut-down of the nuclear power stations. As a result, at least 60 to 120 tons of plutonium will be contained in German storage sites for highly active heat producing wastes.

The safeguards are necessary for practically all of the final storage sites being built internationally, in order to guard against proliferation of nuclear fuel. This requirement is the result of the fear that future generations could use the final storage sites as a plutonium mine and use the stored plutonium for military purposes.

Results of the investigations of the different options for further handling of plutonium

One option for the further use of plutonium is by using MOX fuel elements, which for various reasons are usable with just a portion of recovered plutonium.

The disadvantage of MOX utilisation, besides the technical risks, is also far higher costs in the manufacture as for uranium fuel elements. Also with regard to the spent MOX fuel elements, as compared to the spent uranium elements, there are more requirements to be met.

With regard to the alternatives for the utilisation of MOX fuel elements for the purpose of handling the recovered plutonium, there have already been discussions for some years now.

One of such debates took place in the US regarding 50 tonnes of weapons' plutonium that became surplus after disarmament. The decision-making body at the time designated that a portion of the weapons' plutonium was to be vitrified and another was intended for MOX fuel elements, but possibly even to go for vitrification option for the whole amount.

The vitrification variation that is most popular in the US is the production of plutonium-charged titanium ceramic, which are mounted in high-grade steel containers that are then filled with borosilicate glass, adding HAWC (highly radioactive fissile solution) from military reprocessing.

This produces a product that by its high radiation and chemicophysical shape is considerably better protected in terms of the danger of proliferation than pure plutonium.

In respect to the German situation, the procedure has been adapted in this report, to ensure quick feasibility. The usual mixed-oxide ceramic forms the basis for the ceramic. The product should be the same as glass blocks fabricated from German HAWC (highly radioactive fissile solution). For this method, large-scale technical experience and existing plant can be utilised.

In this report the "direct vitrification of plutonium with fission products" is investigated as a further method of vitrification, where plutonium is vitrified along with HAWC (highly radioactive fissile solution). This obviates the production of mixed-oxide ceramic, thereby steeply reducing the costs of the procedure. Vitrification, using this method can however only be undertaken in a vitrification plant, which would need to be constructed from scratch.

Two other researched alternatives are to be found in the area of the storage rod method. This technique was developed by the Oeko-Institut eV in 1992 in a detailed report.

In the storage rod method, storage rods are manufactured, which in terms of their storage-relevant characteristics correspond to MOX fuel rods, but are designated for direct permanent disposal.

Since there will be no deployment in a reactor, the specifications associated with reactor deployment can be disregarded (internal pressure, level of demand on dimensional stability) when manufacturing the storage rods.

Thereby clear simplifications of the manufacturing process are resultant, which can trickle down as cost reductions.

The storage rod method with storage elements comprises the following discrete steps:

- The manufacturing of mixed-oxide ceramic, storage rods and storage elements, analogous to the conventional procedure in existing MOX fuel element factories

- The transportation of storage elements and corresponding to the transportation of fresh MOX fuel elements to the nuclear power plant
- The mix loading of storage elements and spent fuel elements in a transportation and storage container
- Mixed interim storage of storage elements and spent fuel elements
- Mixed permanent disposal of storage elements and spent fuel elements

The second variation of the storage rod method is different in that the mixing with spent fuel takes place by exchanging individual rods in the spent fuel elements with storage rods in order to produce mixed elements. This would take place in the cooling pond for spent fuel at a nuclear power plant. The other steps in the production and storage processes remain identical to the other variations.

In both cases the spent fuel guarantees protection via its high radiation. The storage rod method is based in both variations on techniques of which Europe has expansive large-scale technical experience. Only existing plants are needed for the treatment of the plutonium.

The capability for permanent storage is guaranteed in all of the four methods that have been examined. This was revealed through the comparison with other materials that would inevitably have to be housed in a permanent storage facility. In respect to the evidence that needs to be supplied for long-term safety and critical safety as well as for safeguard measures there are no additional requirements since the same type of evidence would have to be provided. The future permanent storage facility certainly needs to be equipped in such a way that vitrified plutonium or storage rods can be permanently stored.

The storage rod method with the fabrication of storage elements has displayed the greatest advantages of all the researched methods for permanent storage of plutonium and should be given preference in the continued handling of reactor plutonium.

There is large-scale technological experience available for all the necessary processing steps and all these steps can be carried out in existing plants - which are also accessible to German clients.

The storage rod method leads to a product capable of permanent storage and therefore cannot be seen only as a interim solution. Through the manufacturing of storage rods and elements a figure of at least 3.3 to 7 tonnes can be processed, meaning that by this method, the present plutonium is expected to transformed into a form capable of permanent storage within the shortest time frame in comparison to other methods. A further advantage of the storage rod procedure is that the entire German plutonium could be handled in this way as the number of available spent fuel elements is sufficient for mixing for interim and permanent storage. In comparison to MOX utilisation, this can result in clear economic advantages; the costs are dependent on the achievable amount of fissile materials in the fabricated ceramic.

In comparison to the fabrication of storage elements, the storage rod method with rod exchange entails the additional procedure of exchanging the storage rods for fuel rods in spent fuel elements. Due to a large number of rods to be exchanged, there is a greater risk of rod damage and also a higher collective dosage for personnel. The storage rod method with rod exchange should therefore only be used if, on the basis of far-reaching "physical protection" (because of a more intensive mixture), it is considered to be absolutely necessary.

The realisation for this and for the storage rod method with fabrication of storage elements can be achieved by resorting to existing plants and technical procedures available.

The can in canister method of plutonium vitrification displays fundamental disadvantages in comparison to the storage rod method insofar as technical conformity may be required in a foreign vitrification plant. Using this method would require that an appropriate agreement would need to be reached with the foreign company. With the desired agreement in place the feasibility may be possible without massive additional expenditure.

The direct vitrification of plutonium with fission products should only be considered if the construction of a new vitrification plant was already a necessity. The demand for such a newly planned plant is not foreseeable in Europe's future. If a new plant were to be constructed, direct vitrification would be economically attractive.

All in all, priority should be given to the implementation of the storage rod method with the fabrication of storage elements and thereby provide a practicable opportunity to reduce the existing mountain of recovered (separated) German plutonium.

The main results are summarised in Table 1. In Table 2 costs of the plutonium handling by storage element fabrication and final disposal are given and compared to the costs of fabrication and use of MOX.

Table 1: Results for the different options

	Can-in-canister	Direct vitrification	Storage fuel (fuel assemblies)	Storage fuel (rods)
Technical experience for the steps before final disposal	exists	major part exists	exists	exists
Demand of new facilities	none	new vitrification plant	none	none
Need of new licences	modifications only	new licences	modifications only	modifications only
Conditioned amount of Pu-tot in kg/y	ca. 900 - 1800	ca. 4000	ca. 3300-7000	ca. 3300-7000
Conditioned total amount of Pu-tot in kg	ca. 4000–16000	ca. 3000-9000	no upper limit	no upper limit
Costs in DM/kg	ca. 36.000-115.000	Ca. 23.000-25.000	ca. 42.000-139.000	ca. 43.000-142.000
Product suitable for final disposal	yes	yes	yes	Yes

Table 2: Costs of the storage element option as a function of the total amount of Pu that is processed

Total amount of Pu processed	Costs in DM/kg Pu-tot	
	5% fissile material	**10% fissile material**
1.000 kg	88.700 – 139.400	44.000 – 68.500
25.000 kg	87.300 – 138.100	42.100 – 66.600

Fabrication and use of MOX fuel assemblies:
67.000 – 86.000 DM/kg Pu-tot