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TRANSMUTATION OF LONG-LIVED NUCLEAR WASTE

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Recently the possibility to convert nuclear waste into harmless or even valuable material has been used as an argument in the public discussion on new waste treatment strategies. The NEA as well as the EC encourage national research institutes to organize the study of new transmutation methods in an international network. This new exercise in modern alchemy was initiated by Japanese efforts in the OMEGA project, which means: "Options for Making Extra Gains from Actinides". This is not considered as a substitute for studies of management and disposal of radioactive waste, but rather to develop a complementary policy. By now it is generally recognized that an integration of transmutation methods in the nuclear fuel cycle could reduce the long-term toxicity of commercial waste and also allows for a cleaner transmutation of plutonium from the expected decommissioning of weapons. An extra benefit of this more economic use of fuel could be a reduction of risks due to uranium mining.

In ancient times compounds of uranium were applied as an additive in a process to produce yellow glass. Chance only will favour the prepared mind, and it was Becquerel who "accidentally" discovered radio-activity of uranium (in 1896) by means of newly developed photographic techniques. This discovery was followed by the rapid advancements in nuclear physics and technology, which culminated in the introduction of electro-nuclear power by neutron-induced fission. In this process heat is liberated by transmutation of uranium into waste products. Spent fuel mainly consists of the uranium remains (usually as an oxide), contaminated with fission products and traces of actinides from capture of neutrons in the uranium. Fissile actinides like plutonium and uranium can be recovered, whereas the remaining radio-active waste can be vitrified into boron-silicate glass and permanently stored in repositories. After all, fission products and trace actinides stay in glass for a long time, and repositories are reliable for many millions of years. Gas and oil enclosed in old gas-tight geological structures demonstrate the reliability of geological barriers, but on the other hand the fact that fossil fuel can be extracted, also demonstrates that human intrusion cannot entirely be ruled out.

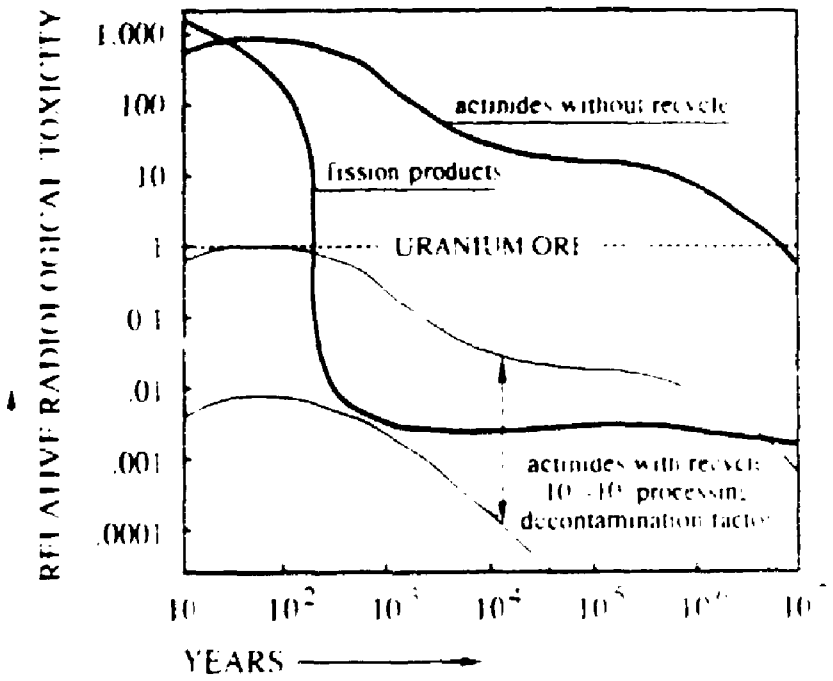
Long-lived nuclear waste emits radiation for thousands of years, and the moral issue is being raised that the long-term risks should be reduced. Our descendants may neither be in a position to profit from electro-nuclear power nor be aware of the repositories, which may be ancient by then. Therefore radio-toxicity of waste should be "ALARA" (as low as reasonably achievable), and it ought to be worthwhile to find out whether one could develop some clean, safe, and economic methods to eliminate some risky components. Just like

transmuting uranium, one could in principle learn to transmute waste, and reduce its long-term radio-toxicity again to the level of the original uranium ore. Aspects of risk, energy, and of resources should be considered before decisions are taken how far one should go.

A problem is that estimates of the radio-toxicity for some of the critical elements in waste have changed by an order of magnitude during the last decade. Usually the discussions of risk are being focused on "high-level waste", like spent fuel or toxic leftovers from the fuel cycle. Any comparison to the risks of the original uranium ore emphasizes radio-toxic transuranium elements. Fig.1 (from ref.1) shows how the radio-toxicity of high level waste is thought to decrease by decay. Actinide toxicity will remain high for a very long time, unless the waste actinides are removed and transmuted.

Figure 1:

VIEW ON THE DECAY OF RELATIVE TOXICITY OF SPENT FUEL.

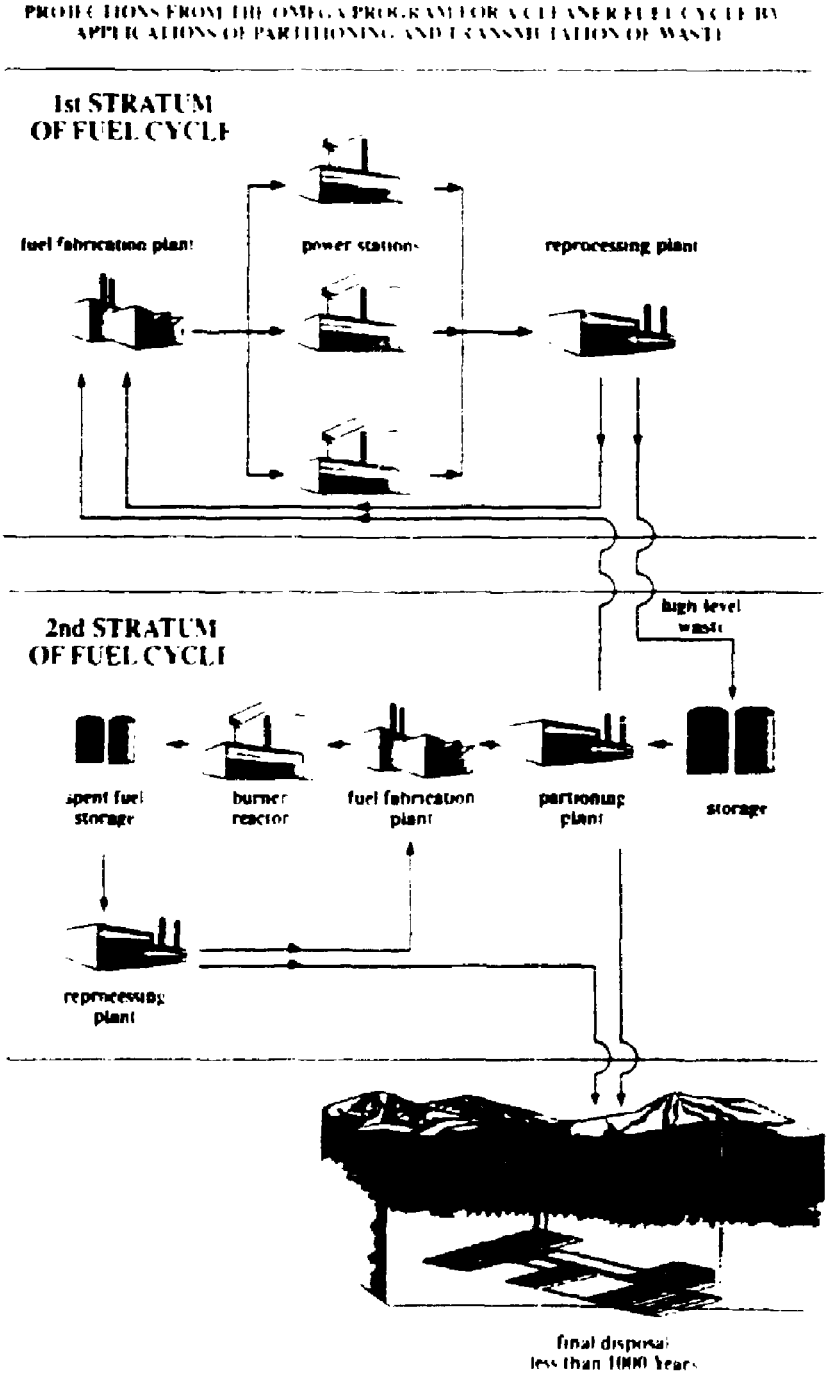


Radon gas and mining risks

Globally the dose risks due to natural radon gas in dwellings and from potassium in food exceed by far the present dose risks from stored high level nuclear waste. Radon is a decay product of the natural uranium in the soil, which leaks to the surface while it decays in a few days. It is revealing to realise that the radon dose-rate is about 50% of the total radiation dose-rate to which mankind is submitted, and that this dose-rate would disappear in

the very unlikely case that all natural uranium on earth would have been removed or well covered. This paradox has been phrased as: "uranium: don't leave it in the ground". In the mean time however, the uncovered uranium mill tailings and the emanations thereof are radiological risk factors of the nuclear fuel cycle, which cannot be neglected. Although the radio-toxicity of ore is more than a factor thousand less than that of the spent fuel (see fig. 1), the volume of mining waste is much too high to consider vitrification followed by deep storage in some geologically stable repositories. Therefore one should limit mining by applying a much more efficient incineration to the uranium and to the heavier actinides. "Limit the need for mining" as opposite to the above-mentioned paradox, might better be rephrased as "uranium: fission it entirely or leave it deep in the ground".

Figure 2:



Fuel-conservation will not only reduce need for mining, but it can also reduce long-lived radio-toxicity of high level waste. Combining fuel utilization with actinide-transmutation would be a procedure to limit the production of long-lived toxic waste. These techniques are to be integrated in nuclear fuel cycles in order to improve economy and to limit the transport.

This figure is based on information presented in reference 2.

Risks from mobile components

Just like the radon gas leaks out of uranium waste, there also are other geo-chemically mobile components, which may leak out of deep repositories towards the surface. In contrast with the case of radon, this is not a matter of days, but of millennia. Nevertheless a few long-lived (up to millions of years) waste elements give a high contribution to the long-term dose-rate. Special candidates for transmutation are therefore fission products such as technetium (Tc-99), iodine (I-129), and cesium (Cs-135), as well as actinides such as neptunium (Np-237) and its precursors. Table 1 (refs. 3,4) shows, for a hypothetical set of unperturbed granite repositories the global surface dose-rate, due to leakage of the stored amount of nuclear waste from the total nuclear production over the next 300 years (relative to natural dose-rates).

TABLE 1: RELATIVE SURFACE DOSE RATE DUE TO LEAKAGE OF SPENT STORED LWR FUEL FROM A 2000 GW(e) REACTOR PARK OPERATING FOR 300 YEARS (such a park would generate most of the global electric power)

Period :	One million years		Hundred million years	
Storage :	Vitrified	Direct storage	Vitrified	Direct storage
Nuclides :				
Tc-99	100 %	98 %	17 %	46 %
I -129	-	2 %	-	1 %
Cs-135	-	-	10 %	24 %
U -235	-	-	-	6 %
U -238	-	-	-	14 %
Np-237	-	-	71 %	5 %
Pu-239	-	-	-	4 %
Am-243	-	-	3 %	-
Relative dose rate	1 ppm	10 %	60 ppm	0.2 %

The last line in table 1 indicates the dose rate relative to the natural background. In the case of direct storage there is an expected average individual dose-rate less than 10 % of present natural dose rates. This is not marginal as it would give a total dose to the world population of 5000 mega man Sievert over a full period of one million years. Proper vitrification or removal of Tc would reduce the collective dose rate to 0.05 man Sievert per year, and would lead to marginal individual dose rates. Contributions of U-235 and U-238 show that vitrification is beneficial even for uranium ore which never has seen a reactor! It could be concluded from this table that the surface dose-rates due to leakage, would be negligible if one could vitrify or transmute mobile components. Nevertheless a reduction of the source term before storage should be advocated also for less mobile waste on basis of safeguarding and to avoid the possibility of leakage due to human intrusion.

Recycling plutonium, uranium and minor actinides

Not only commercial waste but also the decommissioning of weapons could yield considerable amounts of plutonium. Most of this ought to be transmuted and used for energy production, as it is hardly possible to guarantee safeguarding for very long periods (one wants to avoid visions of future mines for plutonium). Moreover recycling of plutonium with uranium in MOX (mixed oxide) fuel, could well be applied to gain electricity without having to mine uranium. Use of commercially produced MOX fuel is considered in Europe and Japan.

Recycling studies invariably lead to the conclusion that long-lived minor actinides circulate for quite a long time in the MOX uranium-plutonium cycle before the total production of minor actinides will diminish. Neutron interactions with uranium lead to neptunium, and the capture of neutrons in plutonium will lead to americium, which is a long-lived precursor of neptunium in the waste. The amount of actinide waste can be limited by irradiation in a high neutron flux. A possible objective could be to achieve zero-growth of minor actinides during constant energy production. Because there is no cladding yet nor a fuel which can stand the required irradiation dose, remanufacturing after reprocessing will be necessary. Long cooling periods of the spent fuel are required to limit the heat and the radiation during fuel handling. This however again leads to extra formation of long-lived waste by beta-decay, and the shorter cooling periods are the better it would be. In this view it is important to limit actinide storage to unavoidable traces within the fission product waste and to start recycling right away. In current commercial practice however one hesitates to handle hot spent MOX fuel, as severe conditions of radiation and heat make it hard to operate a fuel cycle with a high decontamination factor.

Burners, which allow continuous reprocessing of minor actinides and fission products, are being considered as an option for the future. Various types of fuel for these burners are mostly studied for minor actinides. Some samples of minor actinides have already been irradiated in the reactors in several OECD countries. Processes to transmute minor actinides need special target techniques; these are studied e.g at CEA and TUI Karlsruhe and other laboratories within the OECD. In the USA a generic system called Integral Fast Reactor is developed, in which pyro-metallurgical reprocessing techniques can be applied to metallic fuel of a fast reactor. This burner reactor is integrated in the fuel cycle, just as was shown for the OMEGA project in fig. 2.

Possibilities to include transmutation of fission product waste in common fuel cycles are not available, and special burner scenarios should be studied. Fission products might be transformed into less harmful or even valuable materials by neutrons, and some groups are

working on this solution. Such work is proceeding within the OMEGA project in Japan and the ATW project in Los Alamos. Demonstration tests e.g. at the Petten High Flux Reactor and the PHENIX reactor (in a moderated irradiation facility) are prepared by the Dutch and the French organisations ECN and CEA respectively.

Not only reactors but also accelerators could be the source of neutrons. In Japan and in the USA some accelerator scenarios are being studied. Beams of accelerated protons can be used to create a neutron source in a spallation target, and capture of neutrons could convert actinide waste into fuel. Accelerator Transmutation of Waste (ATW) is proposed by Los Alamos. In the USA quite some interest is related to cleanup of military waste. Also special techniques are being developed to avoid the build-up of minor actinides, during transmutation. For a very far future hybrid reactor-accelerator systems are now being considered in Japan and in the USA. By continuously feeding accelerator neutrons to sub-critical molten salt reactors, reprocessing of fuel and feeding-extraction of waste could be applied (ref.5) and thorium might be used as fuel. If this concept is feasible a very clean, safe, and almost inexhaustible nuclear fuel cycle can be foreseen. This is one of a number of interesting concepts whose engineering and economical feasibility needs more work to be properly demonstrated and for which international collaboration is worthwhile.

International cooperation

Short term benefits of transmutation of nuclear waste are marginal but international networks create visions on a really long-term period for future-energy production schemes. A NEA network for the exchange of information between OECD countries was established. A CEC network is set up to coordinate studies of targets and fuels by TUI and KfK (Karlsruhe) by CEA and EdF in France and by ECN-Petten in the Netherlands. Activities in the USA are concerned with the earlier mentioned generic systems IFR and ATW. Outside OECD, especially the CIS shows interest in fast reactors to transmute minor actinides (such as curium).

Economics

The costs of all transmutation seem high; figure 2 for example suggests that for the minor actinides a doubling of the present fuel cycle would be needed. One could try to proceed in such a way that each step pays for itself, for example by exploiting precious metals in the waste or by more economic use of the energy which is stored in actinides. During incineration of fission products no energy is liberated and much care has to be taken to limit the cost of waste transmutation to values which are lower than for example the price of the original nuclear fuel.

Conclusions

Nuclear waste disposal in geologically stable repositories is considered to be safe and effective, and the assumptions, which lead to the very long term predictions seem to be satisfied. As possibilities to perturb the repositories, can never be entirely excluded, it could be an attractive option to reduce the toxicity of waste by supplementing the uranium-plutonium cycle with minor actinide burning cycles. In this option the amount of mining waste is limited at the same time because uranium is used economically. If requests for the reduction of long-lived actinide waste would result in much higher costs for nuclear energy, the innovative thorium-uranium cycle might become competitive. It is of vital interest that efforts are now being internationalized in networks to make a proper use of the experience from past civil and military programs. Visions for almost pollution-free energy production could arise if well prepared minds are concentrated on this issue.

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