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LUCAS HEIGHTS

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ABSTRACT

The feasibility of transmuting the hazardous long-lived radioactive isotopes present in reactor waste to stable or short-lived isotopes is examined. Even to transmute only the most hazardous fission product isotopes, caesium-137 and strontium-90, by protons, electrons or gamma rays requires more electrical energy than the reactor can produce. Neutron transmutation of these fission products would require high neutron fluxes such as might be obtainable from a controlled thermonuclear reactor or a spallation target. The development of both systems is many years in the future, and the spallation system would use almost all of the reactor power output.

(continued)

The neutron transmutation of the transuranic component of the waste is feasible by recycling in nuclear reactors. The transmutation occurs by neutron absorption and fission, so that the waste is converted to a smaller quantity of less hazardous fission product and the fission energy is utilised.

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ACTINIDES; ELECTRONS; FISSION PRODUCTS; GAMMA RADIATION; NEUTRONS; RADIOACTIVE WASTE DISPOSAL; RADIOACTIVE WASTES; THERMONUCLEAR REACTORS; TRANSMUTATION; TRANSURANIUM ELEMENTS; SPALLATION

CONTENTS

1. INTRODUCTION

The disposal of the long-lived radioactive waste produced in the nuclear fuel cycle is a problem that must be considered in any plan to introduce nuclear power. The high level waste from the fuel reprocessing plant contains most of the activity from the whole cycle and is the most hazardous component. The reactor causes little radioactive pollution during operation because most of the waste is contained wholly within the fuel elements.

The spent fuel from a reactor is normally stored for up to 100 days to allow the shorter-lived fission products to decay and hence reduce radiation damage to the reprocessing medium. The fuel is then reprocessed to obtain the plutonium and/or the residual enriched uranium. The high level waste from the reprocessing plant is a nitric acid solution containing fission products and small quantities of uranium, plutonium and other heavy elements.

The liquid high level waste is currently stored in large tanks that have cooling systems and leak detectors. The USA estimates that by 1980, with 150,000 MW(e) installed, the annual production of high level liquid waste will be 3,700 m^3y^{-1} (Rubin 1972). Methods of solidifying this waste into the form of calcine cake, microcrystalline ceramic, glass or powder, are being investigated to reduce the risk of leakage and to reduce the volume of the waste. Solidification will reduce the volume of high level waste per annum from 25 m^3 per 1,000 MW(e) to about 2 m^3 per 1,000 MW(e).

This report will examine the possibilities of transmuting the problem wastes into less hazardous isotopes to obviate the need for the long term storage. First, there is a need to look at the nature of the fission process and the properties of the wastes produced.

2. FISSION PRODUCTS

Every fission event produces about 200 MeV of thermal energy and two fission fragments. About 6.7 per cent of the energy is obtained from the subsequent beta and gamma emissions as the fission fragments decay to the more stable fission products. The rate of production of fission products in a reactor is directly related to the fission rate; a fission rate of 3×10^{10} fission s^{-1} produces 1 watt of thermal power and a 500 MW(e) station (with a thermal power of 1,500 MW(th)) fissions about 1,500 g of uranium-235 per day, yielding about 1,500 g of fission products per day.

The fission product inventory within a fuel element depends on the irradiation time, the cooling time, the total burnup, the fuel composition, and the neutron flux and energy spectrum. Neutron rich fission fragments are the preferred products of fission and each mass chain steadily decays towards

a stable isotope by beta and gamma emission. At the same time, all of the isotopes in the reactor are subject to neutron absorption. In this way, activation products like europium-154 ($t_{½}$ = 16 years) and caesium-134 ($t_{½}$ = 2.05 years) are produced from the direct fission products europium-153 and caesium-133. In this report, all such products will be considered as part of the fission product waste.

The properties of the long-lived radioactive fission products, including daughter nuclides and activation nuclides, are listed in Table 1. These nuclides account for only 0.15 per cent of the total activity one day after fission, but they account for more than 99.9 per cent of the residual activity after 10 years. Naturally occurring isotopes with half lives between 10^{10} and 10^{15} years have not been included in the table. One other significant omission is tritium ($t_{½}$ = 12.3 years) produced by neutron absorption in heavy water or lithium used in some reactors and by ternary fission. At the present time, most of the tritium and the krypton-85 is released to the atmosphere, but methods are being developed to remove the krypton-85.

The isotopes strontium-90 and caesium-137 stand out as most significant; not only are their half lives relatively long, but they also produce energetic beta and gamma emissions. The isotope strontium-90 and its daughter nuclides emit 71 per cent of the total fission product beta energy at 10 years, and caesium-137 emits 24 per cent of the beta energy and 75 per cent of the gamma energy (Wolkenhauer 1972).

The importance of these two isotopes is confirmed by considering the relative hazard of the fission product isotopes. The derived water concentration limits (DWC) for each isotope are shown in Table 1 (IAEA 1973). The continuous ingestion of water containing the isotope at this concentration over a 40-hour week would produce the level of risk that is considered acceptable for the occupational exposure of radiation workers. The relative hazard of the isotope can be considered to be the quantity of water needed to dilute the fission product isotope to the concentration limit, *i.e.* the hazard H(t) is

$$H(t) = Ci(t)/DWC \quad ,$$

where Ci(t) is the activity in curies as a function of time.

Figure 1 shows the hazard associated with the fission products produced by the fission of 1 kg of uranium-235. Strontium-90 dominates the fission product hazard from 10 years to 600 years. Beyond 600 years a group of very long-lived isotopes, including iodine-129, control the hazard. It has been

estimated that the hazard from these very long-lived isotopes could be controlled by isotopic dilution (Bell & Dillon 1971).

The fission products produced in a fast reactor vary somewhat from those shown in Table 1. From a hazard point of view, the most important difference is that the yield of strontium-90 from uranium-238 or plutonium-239 fission by neutrons with a fission energy spectrum is only about half that for thermal neutron fission of uranium-235. Nevertheless, the strontium-90 isotope remains the principal hazard to be considered for waste disposal.

3. TRANSURANIC ELEMENTS

As well as the fission products, the reactor produces numerous transuranic actinide isotopes by neutron capture. Whereas the great majority of the fission products have half lives less than 30 years, many of the actinides have half lives of thousands of years.

The aim of fuel reprocessing is to remove all of the uranium and plutonium from the waste. Currently, the extraction process is about 99.5 per cent efficient, leaving 0.5 per cent of the uranium and plutonium in the waste stream. In addition there are significant amounts of americium, curium and neptunium produced in the reactor.

Table 2 lists the most important actinides in the waste from a light water reactor (LWR). The concentrations were calculated for the Diablo Canyon Reference LWR operating with a burnup of 33,000 MWd t^{-1}, a power density of 30 MW t^{-1} and a neutron flux of 2.9×10^{13} neutron $cm^{-2}s^{-1}$ (ORNL 1970). The uranium and plutonium recovery process has been assumed to be 99.5 per cent efficient, so that the quantities in the table correspond to 0.5 per cent of the uranium and plutonium content of the discharged fuel.

The waste from a fast breeder reactor (FBR) contains more actinide activity than that from conventional reactors, with appreciably larger quantities of americium and curium (Table 3). The total actinide activity of the fast breeder reactor fuel is about five times greater than that of the light water reactor after 1,000 years.

The actinide waste nuclides undergo complex decay chains involving alpha and beta emissions, and some of the intermediate nuclides have long half lives, *e.g.* neptunium-237 with $t_{½} = 2 \times 10^6$ years. The neptunium-237 component builds up after 1,000 years due to the alpha decay of americium-241 ($t_{½}$ = 458 years) and curium-245 ($t_{½}$ = 9,300 years).

Clairborne (1972) calculated the production of actinides in a pressurised water reactor (PWR) and included nuclides up to californium-254 and einsteinium-253. The principal actinide waste hazard was controlled by

americium and curium up to 10^4 years while the long-lived neptunium dominated at longer times. The remaining actinides - actinium, thorium, protactinium, berkelium, californium and einsteinium - make a negligible contribution to the waste hazard.

The total relative hazard of the actinides in a PWR has been indicated in Figure 1 and shows that beyond 400 years the actinide waste hazard is much greater than the fission product waste. While geological stability can be reasonably assured over the 1,000 years sufficient for the bulk of the fission products to decay, time scales of greater than 10^5 years must be considered for the actinides.

4. TRANSMUTATION

The long term hazard of the fission product and actinide wastes makes the possible transmutation of the hazardous nuclides particularly attractive. Figure 2 shows two transmutation schemes and their relation to the reactor fuel cycle. In the first scheme the reactor waste is further processed to separate the transuranic actinides. These actinides are then recycled through the reactor where they are burned up by neutron absorption and fission. A more complex transmutation system uses some of the reactor electrical power output to drive a particle accelerator. The accelerator beam is then used to transmute selected hazardous isotopes from the high level waste.

Any transmutation scheme must satisfy the following basic requirements to be feasible:

- The required energy should be a small fraction of the fission energy obtained when the waste was formed.
- The products of the transmutation scheme should be less hazardous than the waste being transmuted.
- The rate of removal of radioactive waste should be significantly greater than the rate of natural decay.
- The rate of transmutation should be high enough to transmute a significant fraction of the waste produced by reactors.

Considerations of the energy balance show that it is not feasible to transmute all of the high level waste. The original fission produces two fission product nuclei and 200 MeV of thermal energy. The thermal efficiency of light water reactors is only about 32 per cent, but high temperature gas cooled reactors (HTGCR) and fast breeder reactors should attain 42 per cent efficiency. Taking the higher efficiency, 84 MeV of electrical energy is obtained per fission, or 42 MeV per fission product nuclide.

Initial consideration will be restricted to transmuting the more hazardous waste, strontium-90, caesium-137 and the actinides. The reactor generates 700 MeV of electrical energy for each strontium-90 or caesium-137 nucleus produced. The reactor system is basically a source of electrical energy, so any transmutation process must use only a small fraction of the energy generated.

The isotopic distribution of the problem nuclides needs to be considered. Three strontium isotopes are produced as fission products: strontium-88, -89 and -90 with yields of 3.69 per cent, 4.77 per cent and 5.89 per cent respectively. However, strontium-89 decays with a 52-day half life to the stable yttrium-89 which can be readily separated. The stable strontium-88 has a small thermal neutron capture cross section, σ_c = 0.006 b, compared to strontium-90, σ_c = 0.8 b. Hence the chemical separation of strontium after a suitable delay is sufficient for thermal neutron transmutation.

Three isotopes of caesium occur as direct fission products: caesium-133, -135 and -137 with yields of 6.7 per cent, 6.4 per cent and 6.3 per cent respectively, and caesium-134 occurs as an activation product. Caesium-133 is stable, caesium-135 has a half life of 3 x 10^6 years, and they both have high thermal neutron cross sections of σ_c = 29.5 b and 9 b respectively. In comparison, the problem isotope caesium-137 has a thermal cross section of 0.11 b. Hence caesium-137 would need to be isotopically enriched to obtain reasonable economy in any thermal neutron transmutation scheme.

The rate of transmutation is proportional to the amount of material present and so it is not possible to completely transmute a sample. The transmuted material must be separated periodically from the untransmuted material to maintain the efficiency of transmutation.

Unfortunately, any extra processing of the radioactive waste increases the low level radioactive release to the environment. Although the total amount of hazardous material might be reduced by transmutation, the reduction would occur in the material held in engineered storages. The radioactive waste in the environment would increase. A compromise must be made between the possible future accidental release from a storage and the present release from extra processing.

The aim of the transmutation scheme is to convert the hazardous waste to less hazardous materials. Most simple nuclear transmutation interactions, (n,γ), (p,n), (n,2n) *etc.*, with strontium-90 will produce an isotope that will soon decay to a stable nuclide. The longest-lived intermediate isotope is yttrium-91 with a half life of 59 days. The only exceptions are rubidium-90

which will decay back to strontium-90 and the mass 93 chain which decays to zirconium-93 with a half life of 9×10^5 years. Neither the rubidium-90 or the mass 93 chain are likely products of a transmutation scheme.

For caesium-137 the same is true; most products of simple transmutation interactions will soon decay to stable isotopes. The exceptions are xenon-137, which decays to caesium-137, the mass 135 chain which decays to caesium-135 (half life 2×10^6 years) and the mass 134 chain which decays to caesium-134 (half life 2.1 years).

5. NEUTRON TRANSMUTATION OF FISSION PRODUCTS

The neutron transmutation of fission products is the most promising because neutrons can be produced in large numbers and they are unaffected by the nuclear coulomb barrier. However, the very presence of large quantities of an isotope in the reactor waste means that its neutron absorption cross section must be small; if not, the isotope would be transmuted in the reactor. High neutron fluxes can be obtained from high flux reactors, controlled thermonuclear reactors, or spallation reactors.

5.1 Burner Reactors

The burning of the isotopes strontium-90 and caesium-137 in a fission reactor has been considered by Steinberg, Wotzak & Manowitz (1964) and Clairborne (1972). Ideally, we would want a reactor in which the rate of transmutation was greater than the rate of production of the strontium-90 and caesium-137. The yield of these isotopes is 0.12 atoms per fission, so that even to obtain a transmutation rate twice the production rate would require 0.25 neutrons per fission from the chain reaction. These neutrons would otherwise be used for fission, or for breeding or converting more fuel from fertile material. The use of these neutrons for transmutation would have a significant effect on the neutron economy of the reactor. The small neutron capture cross sections mean that a very high neutron flux is required to transmute the isotopes at a rate significantly faster than radioactive decay. A neutron flux of 10^{15} neutron $cm^{-2}s^{-1}$ (corresponding to a current high flux reactor) would only transmute half of the strontium-90 in 27 years and half of the caesium-137 in 200 years. Although future liquid metal fast breeder reactors could have fluxes up to 3×10^{15} neutron $cm^{-2}s^{-1}$, the lower neutron capture cross sections at the higher neutron energies mean that the transmutation rate is similar to that of thermal reactors. Clairborne has considered strontium-90 recycle in a conventional PWR and found only a 5.5 per cent reduction in the strontium-90 inventory at an incremental cost of 0.01 cent per kWh(e). This reduction is too small to reduce significantly

the strontium-90 hazard. There is also the disadvantage of the large quantity of radioactive nuclides being actively handled, processed and effectively stored in the reactor.

5.2 Controlled Thermonuclear Reactor

As an alternative to the burner reactor, Wolkenhauer (1972) has suggested the use of a controlled thermonuclear reactor (CTR) as a source of high energy neutrons. The CTR could provide the high flux levels necessary for transmutation.

Wolkenhauer suggested that the deuterium-tritium CTR with a tritium breeding ratio of 1.2 could provide a 14 MeV neutron flux of 5×10^{15} neutron $cm^{-2}s^{-1}$. The (n,2n) reaction generally has a maximum cross section at about 15 MeV. By analysing the transmutation chains for strontium-90 and caesium-137 and the competition for neutrons between the parent and daughter nuclei, Wolkenhauer was able to show the transmutation process could be a net producer of neutrons by virtue of the (n,2n) reaction. This would make the transmutation process relatively cheap.

Wolkenhauer estimated that the caesium-137 and strontium-90 from a nuclear economy could be transmuted in CTR plants having 8 per cent of the total generating capacity. Of course, the CTR plants are many years in the future and there must be doubts about the desirability of increasing the hazard by incorporating the highly radioactive wastes in an operational device. Isotopically pure materials were assumed in the analysis. Isotopic enrichment could not be justified so the analysis needs to be extended to elemental loadings.

Finally, the economic value of the 14 MeV neutrons needs to be considered. If electrical energy can be sold for \$0.006 kWh^{-1}, then thermal energy is worth \$0.002 kWh^{-1} and the 14 MeV neutron is worth about $\$1.25 \times 10^{-21}$ for just its thermal energy. Probably this thermal worth would be obtained as well as the transmutation. However, if uranium-233 costs \$13 g^{-1}, then the neutrons captured in thorium-232 are worth $\$5 \times 10^{-21}$ (Fraser, Hoffman & Tunnicliffe 1973). Because of these economics, it is likely that the 14 MeV neutrons would be used for fuel breeding rather than for fission product transmutation.

5.3 Spallation Reactor

A spallation reactor could produce a sufficiently high neutron flux for transmutation. When 1 GeV protons are incident on a uranium target, about 39 neutrons per incident proton are produced. As lighter target nuclei are used, the neutron production decreases so that 1 GeV protons on lead only

give about 20 neutrons per incident proton. The neutron flux could subsequently be increased by multiplication in a subcritical uranium target if necessary.

The beam energy required to produce each neutron is 25 MeV for a uranium target or 50 MeV for a lead target. Theoretically, accelerator efficiencies of 50 per cent are possible for the conversion of a.c. power to beam power. Assuming no neutron losses in the target, structural materials or daughter nuclei, the minimum electrical energy required for each transmutation could be 50 MeV for a uranium target or 100 MeV for a lead target. Even the idealistic energy is probably too large a fraction of the 700 MeV electrical output from the reactor for each nucleus of strontium-90 or caesium-137. In macroscopic quantities the strontium-90 transmutation rate becomes 1.13 kg y^{-1} per MW beam power, compared with a production rate of 8.2 kg y^{-1} in a 500 MW(e) reactor (HTGCR).

Gregory & Steinberg (1967) proposed a transmutation system comprising a 1,300 MW(e) nuclear reactor driving an accelerator to produce a 650 MW proton beam with energies in the region 1 to 10 GeV. Unfortunately, their fission yields were too small and they ignored the isotopic concentrations in the waste. However, they did look at the problems of heat removal in the target, flux depression in the target and process recycling schemes.

The Canadian Intense Neutron Generator (ING) project considered, in some depth, the problems of constructing a 65 MW beam power proton accelerator and spallation target (Bartholomew & Tunnicliffe 1966). They concluded that uranium had two disadvantages as a spallation target; firstly, the high fission cross section resulted in a large heat production in the target and secondly, the high neutron absorption cross sections resulted in an undesirable flux depression. However, the ING choice of a lead-bismuth eutectic target produces only 20 neutrons per MW beam power.

Many isotopes are produced as spallation products in the target. The ING target was estimated to produce 79 g per year of strontium-90, about equal to that from a 10 MW(th) fission reactor (Church 1967). The quantities of such waste need to be well known before a spallation transmutation scheme can be considered.

Although the energy requirements for transmutation by spallation might be reasonable, the required accelerator systems are still considerably ahead of the state of the art. The Los Alamos Meson Physics Facility (LAMPF) currently has the highest proton beam power of 0.8 MW at 800 MeV. Eight such

accelerators would be needed to transmute the strontium-90 from each 500 MW(e) reactor. Obviously the capital cost and effort would be enormous for a whole reactor economy using the current technology.

6. PROTON TRANSMUTATION

A proton accelerator can be used to cause transmutation directly by proton interactions. The efficiency of proton transmutation can be estimated by considering the competition between ionisation losses and nuclear interaction. The cross section for nuclear interaction above the coulomb barrier was assumed to be equal to the geometric nuclear cross section,and the range-energy relations were obtained from Barkas & Berger (1964).

The beam energy expended per nuclear interaction decreases as the incident proton energy increases (Figure 3). At low energies, the increased ionisation losses greatly reduce the proton range and no nuclear interactions can occur when the proton energy becomes less than the nuclear coulomb barrier. The nuclear interaction cross section includes inelastic and compound elastic scattering as well as transmutation processes.

Figure 3 shows the results of assuming either a maximum of one nuclear interaction per proton, or allowing successive nuclear interactions with no energy loss. For only one interaction, there is a minimum of 670 MeV expended per nuclear interaction. This minimum occurs for incident protons with 450 MeV energy. However, the multiple interaction assumption is probably more realistic in that it makes some allowance for secondary processes. In this case, the energy expended continues to decrease to an asymptotic value of about 200 MeV per interaction above 4,000 MeV.

Assuming an idealised accelerator efficiency of 50 per cent for converting a.c. to beam power, the transmutation process would require 400 MeV per transmutation. This is too large a fraction of the 700 MeV electrical energy produced in the reactor per nucleus of strontium-90 and caesium-137; hence, direct proton transmutation is not feasible for disposing the fission products. The protons could be better utilised producing neutrons by spallation as discussed in Section 5.3 .

7. GAMMA RAY AND ELECTRON TRANSMUTATION OF FISSION PRODUCTS

Gamma rays can cause transmutation provided the gamma ray energy is above the nucleon binding energy (6 to 8 MeV). The thresholds for photoneutron emission vary from 8 MeV for lead to 10 MeV for cobalt (Stephens 1960). Photoproton emission is less probable than photoneutron emission owing to the coulomb barrier.

The photonuclear cross section has a broad, large peak, the 'giant

resonance' at 14 MeV for heavy nuclei and 22 MeV for light nuclei. The peak cross sections are about 0.5 b, and the width varies from ~ 3 MeV for spherical nuclei to ~ 7 MeV for others. The principal reactions that occur in the resonance are (γ,n) and $(\gamma,2n)$ which could transmute the troublesome fission products. The integrated dipole photoabsorption cross section (Segrè 1964) is:

$$\int \sigma\ (E_\gamma)\ dE_\gamma = \begin{cases} 1.3 \text{ MeV b for strontium-90} \\ 1.9 \text{ MeV b for caesium-137} \end{cases} .$$

The main competing process for the gamma rays is pair production, with cross sections of 3.9 b $atom^{-1}$ for strontium-90 and 7.7 b $atom^{-1}$ for caesium-137 for 15 MeV gamma rays (Davisson 1965). The pair production cross sections increase with increasing energy.

Gamma rays can be produced from an accelerator source of electrons or from neutron capture in a suitable isotope. Neutron capture is not suitable for transmutation because most of the gamma rays are below the photonuclear threshold. The gamma rays from an accelerator source of electrons are produced by stopping the electrons in a target, preferably of a high atomic number. Above 7 MeV for lead or 22 MeV for copper, radiation loss is the main cause of energy loss for the electrons. The resulting bremsstrahlung radiation has a broad spectrum with a maximum photon energy equal to the incident electron energy. To a first order approximation, the photon energy spectrum has an equal energy density from zero energy up to this maximum energy.

By combining the equi-energy distribution of gamma rays with the integrated dipole cross section and allowing for the pair production, the probability of photoabsorption in the giant resonance per incident electron is:

$$P_\gamma = \begin{cases} 0.0222 \text{ for strontium-90} \\ 0.0167 \text{ for caesium-137} \end{cases} .$$

The total electron energy input for each gamma induced transmutation is then 900 MeV for strontium-90 and 1,200 MeV for caesium-137. Even without putting in accelerator efficiencies and the inevitable losses, the energy required for the transmutation is greater than the electrical energy obtained from the reactor.

Electrons can cause transmutation of the nucleus by direct electron-nuclear interaction, but the electron cross sections are only about α (= 1/137)

times the gamma ray cross section for the same energy (Segrè 1964). Hence, the cross section for neutron emission is only about 5 mb at 20 MeV. This small cross section is far too small to be useful for transmutation. The probability of interaction is only 0.00003 $(g\ cm^{-2})^{-1}$ for strontium-90 and 0.00002 $(g\ cm^{-2})^{-1}$ for caesium-137. The extrapolated range of 20 MeV electrons is about 10 $g\ cm^{-2}$, giving a total beam power of about 80 GeV per electron transmutation.

8. ACTINIDE TRANSMUTATION BY NEUTRONS

The actinides are more suitable candidates for transmutation than the fission products because they pose a greater long term hazard and are not suitable for dilution by a stable isotope. Also their neutron cross sections are larger and their transmutation ultimately results in a fission. Hence, the neutron transmutation of the actinides produces fission product wastes plus the fission energy. The process is justified because the fission products are less hazardous than the parent transuranic actinides by 250 years. The transuranic actinide fission products are essentially the same as the uranium fission products.

Calculations of the effects of recycling the actinides through a PWR reactor have shown appreciable hazard reduction with only a small effect on the reactor neutron economy (Clairborne 1972). For an actinide extraction efficiency of 99.5 per cent from the waste, the equilibrium hazard reduction, after about 20 three-year reactor cycles, varies from a factor of 3.6 at 1,000 years to a factor of 42 at 10^6 years. With an extraction efficiency of 99.9 per cent, the equilibrium hazard reduction increases to a factor of 17 at 1,000 years and a factor of 206 at 10^6 years. The hazard is approximately proportional to the amount of neptunium, americium and curium sent to the waste after separation. The recycling of the actinides and use of a 99.9 per cent efficient extraction process reduces the long term actinide hazard to the same order as the fission product hazard after 1,000 years.

The recycling of the actinides produces a buildup of the heavier elements such as curium-245 and -246, and a change in the isotopic distribution of the waste. Hence, the hazard of the recycled waste has a different decay curve to the unrecycled waste. This is shown in the differing hazard reduction factor for 1,000 years and 10^6 years quoted above. The buildup of the heavier actinides causes a serious increase in the neutron emission rate by spontaneous fission and (α,n) reactions. This could cause difficulties in fuel fabrication.

The recycled actinides have a negative reactivity effect on the reactor.

In a PWR, recycling its own actinides, the average reactivity decrease is about 0.8 per cent. This decrease could be counteracted by only about a 2 per cent increase in fissile material (Clairborne 1972).

The recycling has two disadvantages; first, it increases the radiation problems associated with chemical processing and fuel fabrication and second, there is an increased possibility of a release into the environment due to the handling of the actinides. Any extra processing results in an increase in the volume of contaminated waste because the process chemicals required are discharged into the waste stream along with significant contamination.

Perhaps the biggest difficulty in the way of actinide recycling is the need for a chemical method that will separate the americium and curium but leave the chemically similar rare earth fission products. The californium and berkelium will separate with the americium and curium. If neutron emission is a problem, it might be necessary to have additional processing to separate the californium.

Some of these difficulties could perhaps be avoided by having special burnable poison rods constructed from the actinides. During the lifetime of the reactor the reactivity effect of the rods could become less negative due to the buildup of more fissionable isotopes.

Fast breeder reactors will be more suitable for recycling the actinides because the average fission to capture ratio for these nuclei is higher at fast neutron energies than at thermal neutron energies.

9. CONCLUSION

The fission product hazard is dominated by the isotopes strontium-90 and caesium-137 with half lives of 30 years and 29 years. The energy required for accelerator transmutation of just these two isotopes is summarised in Table 4. The electrical power listed in the table assumes a 50 per cent efficiency for conversion of a.c. power to beam power. Only neutrons produced by a spallation target can be produced with a high enough energy efficiency.

The various transmutation schemes are compared in Table 5. The cost of accelerator systems is very high since, even for the spallation system, 8 MW of beam power would be needed to transmute the strontium-90 from each 500 MW(e) reactor. This compares with 0.8 MW beam power of the 'state of the art' accelerator, LAMPF. None of the methods for transmuting fission products are feasible at the present time, but the Controlled Thermonuclear Reactor might be possible in the future. It is likely that the benefits of transmutation would be less attractive than the gain from using the neutrons to breed

more fuel. This is even without considering the disadvantages of introducing the fission waste into an operating thermonuclear reactor.

The transuranic actinides are a greater long term hazard than the fission product nuclides, because of their much longer half life and the lack of any natural stable isotopes. The transmutation of the actinides in current reactors is feasible and it has only a small effect on the reactor economics. Such a recycling will depend on the development of adequate chemical separation and decontamination processes.

Transmutation is one of many disposal methods currently under study, including engineered storages, deep holes, nuclear cavities, salt deposits, space vehicles, marine deep trenches, and Antartic ice. For the present, the waste is likely to be kept in recoverable storage until much more is known about the different processes and possible associated dispersion mechanisms. The rapid increase in the use of radioactive isotopes might make the fission waste a worthwhile resource in the future.

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TABLE 1

LONG-LIVED RADIOACTIVE FISSION PRODUCTS PRODUCED BY THE THERMAL FISSION OF 1 kg OF URANIUM-235

Isotope	Yield (1) (%)	$t_{½}$ (years)	t = 1 day		t = 10 years	Decay		DWC (4) (Ci m^{-3})
			Mass (g)	curies	curies	max β MeV	max γ MeV	
^{79}Se	0.055	6.5×10^{4}	0.185	0.013	0.013	0.16	0	9×10^{-5} (5)
^{85}Kr	1.30	10.73	4.70	1,844	968	0.67	0.51	10^{-5} (air)
^{90}Sr	5.89	29	22.56	3,092	2,435	0.55	0	1×10^{-5}
^{90}Y	(2)	0.0073	-	-	2,435	2.3	-	-
^{93}Zr	6.34	1.5×10^{6}	25.09	0.064	0.064	0.060	0	0.02
^{99}Tc	6.14	2.1×10^{5}	25.87	0.439	0.439	0.29	0	5×10^{-3}
^{106}Ru	3.03	1.01	13.67	45,000	47.8	0.04	0	3×10^{-4}
^{106}Rh	(2)	(30 s)	-	-	47.8	3.5	1.5	
^{107}Pd	0.17	7×10^{6}	0.77	0.00040	0.00040	0.04	0	9×10^{-5} (5)
^{125}Sb	0.030	2.7	0.16	165	13.0	0.61	0.66	3×10^{-3}
$^{125}Te^{*}$	(2)	0.16	-	-	2.6	0.10	0.11	-
^{129}I	1.00	1.6×10^{7}	5.49	0.00096	0.00096	0.15	0.04	10^{-5}
^{134}Cs	(3)	2.05	5.25	6,960	252	0.66	1.4	3×10^{-4}
^{135}Cs	6.45	2.3×10^{6}	37.05	0.043	0.043	0.21	0	3×10^{-3}
^{137}Cs	6.27	30.0	36.55	3,181	2,520	0.66	0.66	4×10^{-5}
^{144}Ce	5.42	0.78	33.21	106,000	14.6	0.31	0.13	3×10^{-4}
^{144}Pr	(2)	(17 min)	-	-	14.6	2.99	2.2	-
^{147}Pm	2.17	2.62	13.57	12,600	895	0.22	0	6×10^{-3}
^{151}Sm	0.41	87	2.63	67.1	62.3	0.076	0.022	0.01
^{154}Eu	(3)	16	1.44	209	136.0	1.85	1.3	6×10^{-4}
^{155}Eu	0.29	1.8	1.94	2,470	53.7	0.25	0.10	6×10^{-3}
TOTALS			230	1.8×10^{5}	9,898			

(1) Yield mass chain (Crouch 1973)

(2) Daughter of long-lived nuclide

(3) Activation product - mass and activity for a LWR (ORNL 1970)

(4) Derived water concentration limits for occupational exposure

(5) Default value.

TABLE 2

ACTINIDE WASTE FROM A LIGHT WATER REACTOR PER 1,000 MWd(th)

(*i.e.* for 1 kg of fissioned fuel - ORNL 1970)

Nuclide	$t_{½}$ (years)	Waste (g)	Activity (Ci)	
			10 years	1,000 years
^{235}U	7×10^{8}	1.21	2.58×10^{-8}	2.64×10^{-8}
^{236}U	2.4×10^{7}	0.618	3.91×10^{-5}	4.67×10^{-5}
^{238}U	4.5×10^{9}	143	4.76×10^{-5}	4.76×10^{-5}
^{237}Np	2.1×10^{6}	23.1	0.016	0.017
^{238}Pu	86.4	0.025	3.09	3.82×10^{-3}
^{239}Pu	2.4×10^{4}	0.815	0.050	0.063
^{240}Pu	6,580	0.327	0.136	0.249
^{241}Pu	13.2	0.154	10.4	1.2×10^{-22}
^{242}Pu	3.8×10^{5}	0.053	2.07×10^{-4}	2.15×10^{-4}
^{241}Am	458	1.61	5.33	1.26
^{243}Am	7.6×10^{3}	2.74	0.117	0.482
^{242}Cm	0.449	0.177	0.096	1.05×10^{-3}
^{244}Cm	17.6	0.094	52.1	1.8×10^{-15}

Note: Assumes 0.5% of uranium and plutonium lost to waste

TABLE 3

ACTINIDE WASTE FROM A LIQUID METAL FAST BREEDER REACTOR PER 1,000 MWd

(ORNL 1970)

Nuclide	$t_{½}$ (years)	Waste (g)	Activity (Ci)	
			10 years	1,000 years
^{235}U	7×10^{8}	0.215	4.7×10^{-9}	1.0×10^{-8}
^{236}U	2.4×10^{7}	0.0057	5.5×10^{-7}	2.1×10^{-5}
^{238}U	4.5×10^{9}	133	4.4×10^{-5}	4.4×10^{-5}
^{237}Np	2.1×10^{6}	3.82	2.8×10^{-3}	0.011
^{238}Pu	86.4	0.101	11.0	0.057
^{239}Pu	2.4×10^{4}	8.83	0.536	0.561
^{240}Pu	6,580	2.92	0.676	0.673
^{241}Pu	13.2	0.797	53.6	6.4×10^{-22}
^{242}Pu	3.8×10^{5}	0.494	1.9×10^{-3}	2.1×10^{-3}
^{241}Am	458	14.7	47.9	11.0
^{243}Am	7.6×10^{3}	7.72	1.50	1.37
^{242}Cm	0.449	0.60	2.07	0.022
^{244}Cm	17.6	0.564	25.7	8.7×10^{-16}

TABLE 4

ENERGY FOR ACCELERATOR TRANSMUTATION

	Beam Power	Electrical
Proton	200 MeV	400 MeV
Gamma	1 GeV	2 GeV
Electron	80 GeV	160 GeV
Neutron (U target)	25 MeV	50 MeV
(Pb target)	50 MeV	100 MeV

Reactor electrical power: 700 MeV per

strontium-90 and caesium-137

TABLE 5

COMPARISON OF TRANSMUTATION SCHEMES

	Energy Balance	Rate	Cost	Status
FISSION PRODUCTS:				
Accelerator				
Proton	?	-	high	future
Electron	neg	-	high	future
Gamma	neg	-	high	future
Neutron	pos	-	high	future
Burner reactor	pos	too slow	-	present
CTR	pos	OK	-	future
ACTINIDE RECYCLE	pos	OK	-	present

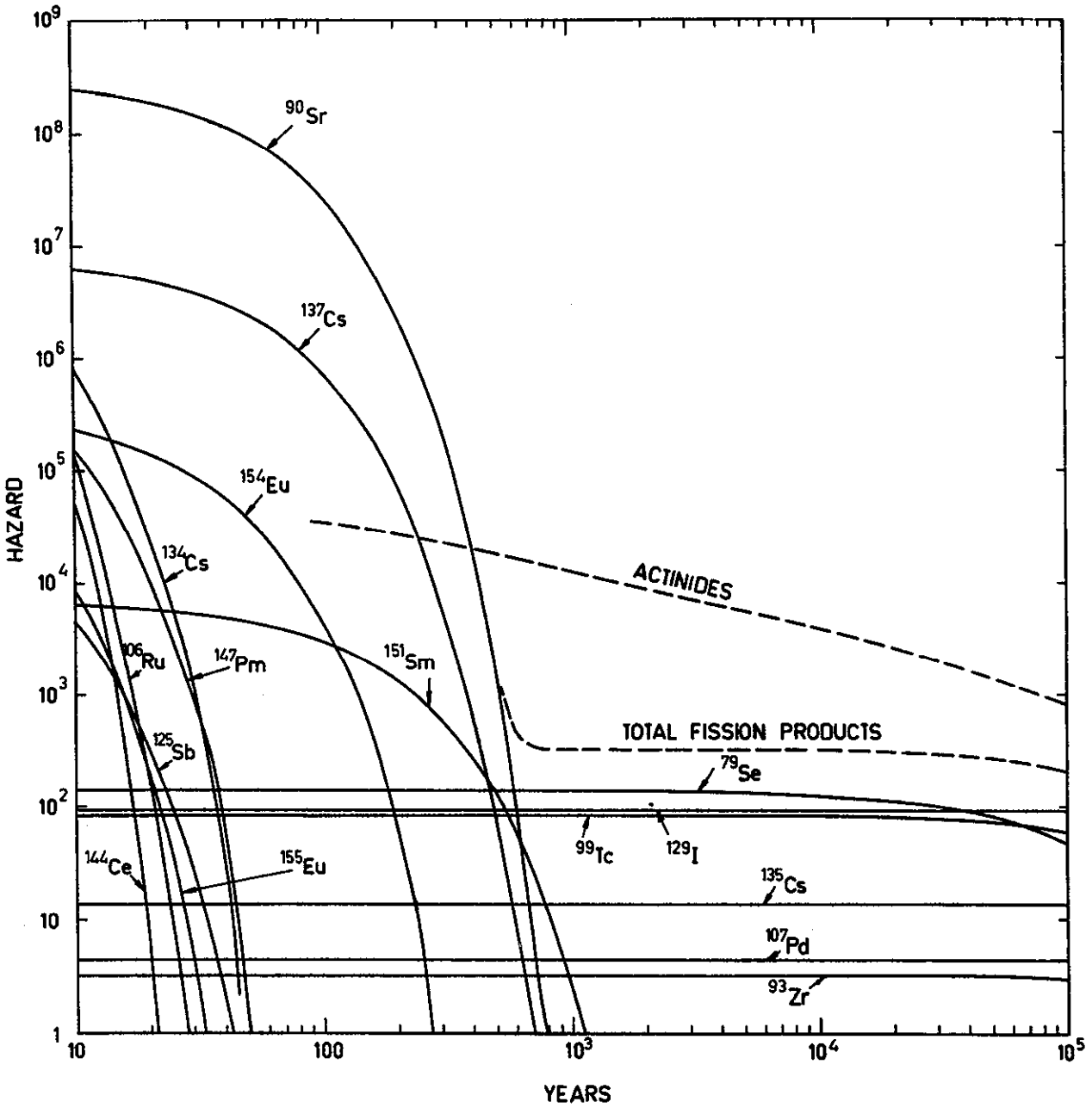

FIGURE 1. RELATIVE HAZARD OF THE FISSION PRODUCT ISOTOPES

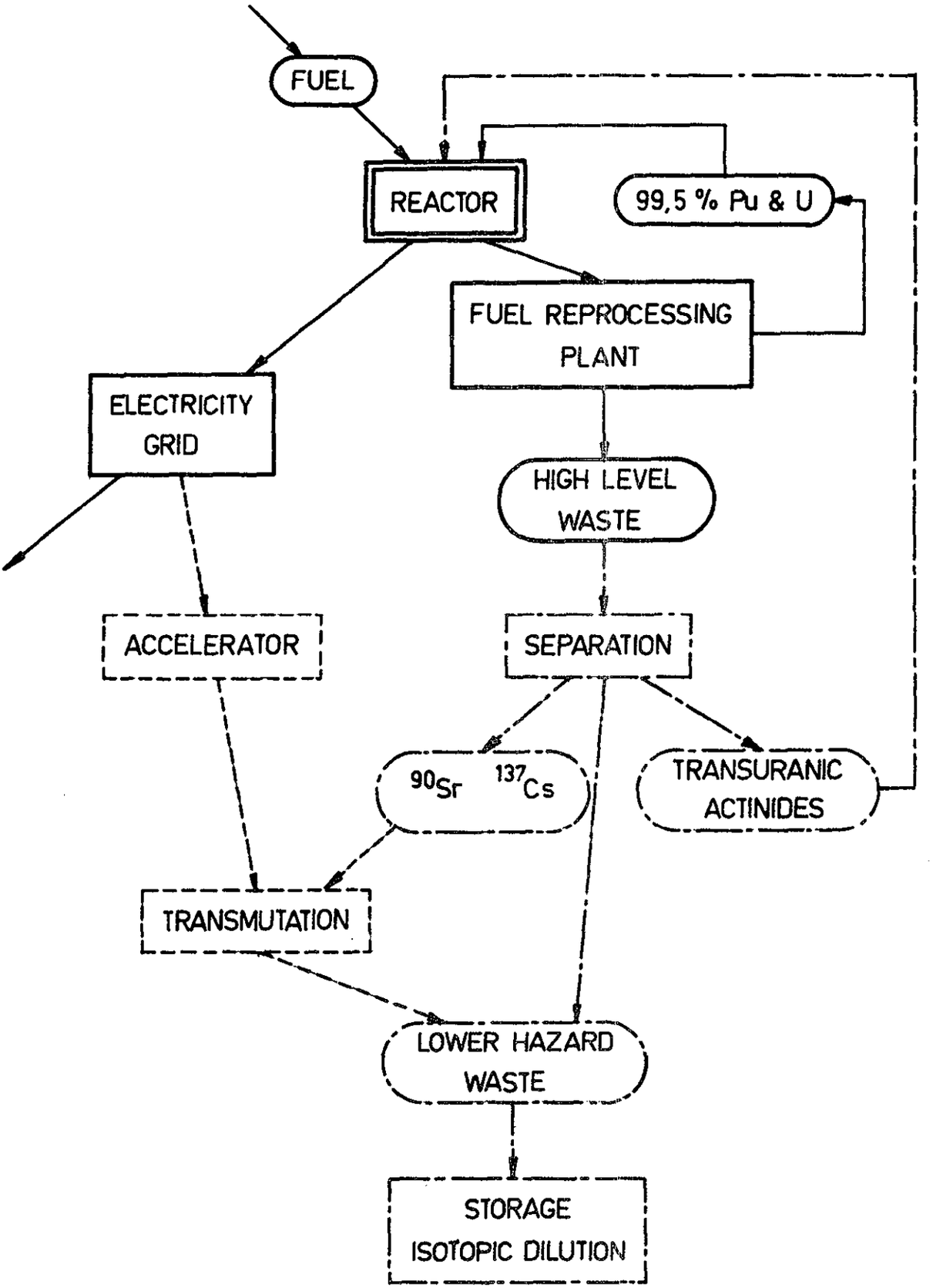

FIGURE 2. THE REACTOR FUEL CYCLE WITH ACTINIDE RECYCLE AND ACCELERATOR TRANSMUTATION

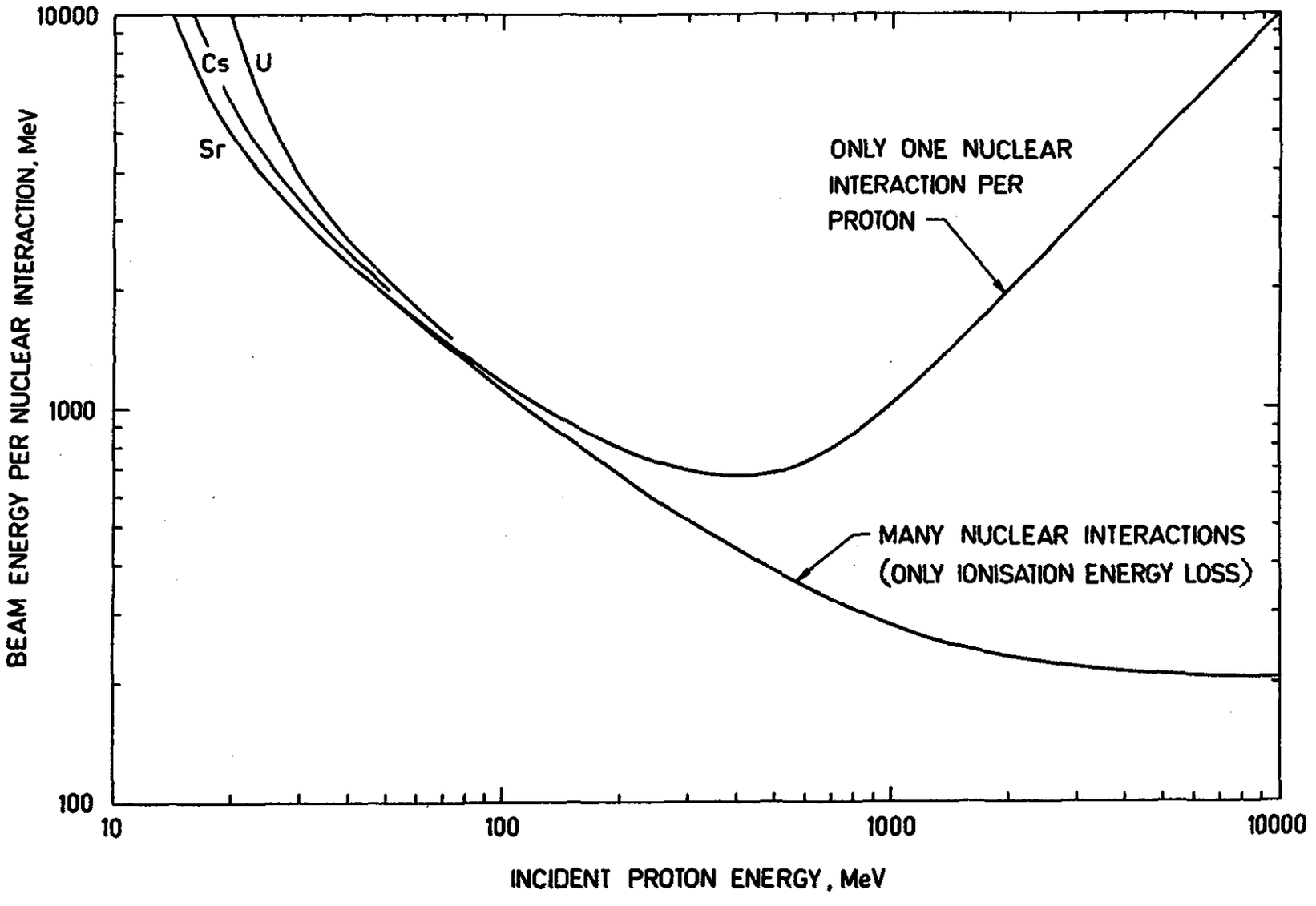

FIGURE 3. ENERGY REQUIREMENTS FOR PROTON TRANSMUTATION

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