NUCLEAR TRANSMUTATION FOR ⁸⁵Kr BY ITS ACCELERATION UP TO 100 MeV/u

T. Itahashi

Research Center for Nuclear Physics, Osaka University, Mihogaoka, Ibaraki, Osaka 567, Japan

Abstract

To eliminate or decrease radiotoxity for radioactive rare gas element, produced in nuclear power or reprocessing plants, such as ⁸⁵Kr a transmutation scheme using accelerators is proposed. The acceleration up to an energy of a few 100 MeV/u would be useful for such nuclei to react and transmutate into another less toxic nuclei. Accelerator complex is discussed on the transmutation efficiency.

Introduction

There are several options for disposition of high-level wastes produced in nuclear fusion power or fuel reprocessing plants. In a nuclear technological field, many techniques have been developed to safely place these wastes in sealed geologic formations where the waste can decay to safe levels of radioactivity^{1,2,3)}.

As a non-retrievable option, full elimination of actinides by injection into space or by transmutation in accelerators or reactors have been proposed.³) If the cost performance could be much improved, the transmutation option would be strongly supported by an environmental safeguard.

Up to now, the transmutation with accelerators or reactors has been discussed on the methods such that the accelerated particles or neutrons react to the long-lived nuclei in the wastes and transmutate an element or nuclide into another less toxic one. Further, recent accelerator development has made possible the acceleration of toxic radionuclide with being selected in nuclear charge or mass, and has made possible the transmutation, annihilation of these nuclide by the high energy reaction.

In this study, we propose the transmutation scheme by using the accelerator complex such as, injector linear accelerator and super conducting cyclotron.

Gaseous wastes

There are fundamental researches and vast technologies in a disposition of the nuclear wastes, and there are many useful references for the treatment or management of radio active wastes.²⁾ These technologies which have been developed so far, require isolated or sealed places to safely keep solidified nuclear wastes. Regardless of an on-site storage or on an away from reactor storage, the disposition of radio active wastes is a crucial problem for the public and the environment.

In table 1~3, the gaseous or liquid wastes from nuclear power plants (Dresden-1) and the liquid disposal from NFS fuel reprocessing plants are listed.⁴⁾ Among the disposal, a technical treatment for inert gas has been seriously behind other than that developed for other chemical disposal. Thus, from the environmental point of view, ⁸⁵Kr ($T_{1/2}$ =10.7y) would make global radio active pollution due to its longer half-life without accumulating into the earth's surface.

Fig. 1 shows the variation of ⁸⁵Kr concentration in environmental air in the northern hemisphere produced by explosions of the nuclear weapons and by commissioning of nuclear power plants.



Fig. 1. Concentration of ⁸⁵Kr in the northern hemisphere.

If a 3000 MWe atomic power plant is operated in one year, ⁸⁵Kr which should be treated, would be 5×10^5 Ci in total; if we assumed 0.3% for the fission yield of ⁸⁵Kr, the disintegration rate (A) can be estimated as

$$A = N\lambda = \frac{0.693}{(10.76)(3.16 \times 10^7)} \times 9 \times 10^{24}$$

$$= 1.8 \times 10^{16} d/s$$
,

where N is the total produced number of 85 Kr per year, and λ is the decay constant.

Further, a nuclear fuel reprocessing plant having a scale of 5ton/d would produce 5×10^4 Ci of 85 Kr per day; this number would be extremely larger amount than that of nuclear power plants.

Estimation of a transmutation rate

Since high-energy heavy ion beams became available, the projectile fragmentation reaction has been studied extensively.⁵) It was then realized that through that process, the high energy projectile (of radio active or not) could be transmuted or annihilated into other elements. There are useful results on measurements of the cross sections for several radio active proectiles on the beryllium, carbon and aluminum targets. From these measurements, it is noted that these interaction cross sections would be more than 0.5 barns, though they depend on the target-projectile combination.⁶)

Therefore, if 85 Kr nuclei are accelerated up to its energy of 100 MeV/u, these nuclei would transmute into another less toxic or more useful one by nuclear reaction. If we assume the transmutation cross section of ⁸⁵Kr at 100 MeV/u as $\sigma \ge 0.5$ barn and the target number (n) as $n=10^{25}$ atoms/cm², we get the reaction rate for the projectile as nearly as 100%.

Accelerator scheme

Acceleration scheme of 13 N and several proposals in isol projects suggest that accelerator complex composed of heavy ion linac for low energy section and super conducting cyclotron for further acceleration section would be most promising scheme to transmutate 85 Kr efficiently.^{7,8}) In order to select a final accelerator among the various choices which can afford the particles up to the required energy, the cost performance to operate such machine, should be taken into account as an important factor. According to this, we designed transmutation system as follows, which is schematically shown in Fig. 2.

From atomic power and reprocessing plants, inert gas separated from other radiotoxic gas, is stored in a reservoir and introduced into an ECR ionizer by an amount to hold the ECR plasma. Some parts of introduced gas go back in neutral state to reservoir again by closed evacuating system. Outgoing ions with relatively lower charge state are accelerated with heavy ion linear accelerators, for instance, RFQ or Wideröe type and Alvarez type. After this, we should install the charge stripper and charge forcuser. Final accelerator would be superconducting cyclotron which has K=1000 equipped with an internal target for the transmutation by projectile fragmentation reaction. By using a K=1000 machine, the charge state of the accelerated particle would be 27, since we require the projectile energy of more than 100 MeV/u.

Each efficiency in various sections of accelerators and ion source could be estimated by using various data of existing machines or isol facilities as follows:

i) Ion source efficiency (ϵ_{ion}) of 85 Kr¹⁺ at ECR source would be

 $\varepsilon_{ion} = 0.65$

if we assume the same values as Xe¹⁺ ion formation.⁹⁾

ii) Stripping efficiency for 85 Kr²⁷⁺ or fraction of equilibrium charge state (ε_{frac}) at the energy of a few MeV/u with

single stripper is estimated¹⁰⁾ as

$$\varepsilon_{frac} = F(\bar{q}) = 0.2.$$

- iii) Acceleration efficiency (ε_{acc}) from injector linac to super conducting cyclotron, although considerable amount of the beam would be lost at injection to the 1st orbit of the cyclotron, would be
 - $\varepsilon_{acc} = 0.15$

where it doesn't include the extraction efficiency.

iv) Reaction rate (ε_{react}), as we already mentioned, would be

 $\varepsilon_{reac} = 1.0.$

In fact, it is probably possible to increase substantially the total transmutation rate by recycling neutral gas and ions non-accelerated or accelerated up to a certain energy.

By this procedure, the effective or artificial decay rate of 85 Kr would be much faster than the natural one; the simple estimation by using the above efficiencies for various stages, gives the transmutation rates $10^5 \sim 10^6$ times faster than the natural decay rate of 85 Kr.

There are, however, many inevitable problems produced by the acceleration of radio-active ions: i) in total system, any leak into environment should not be allowed; ii) energetic particles lost in due course of the acceleration or transportation should be recollected and recycled; iii) radio active damage due to the radiation induced by the high energy particles should be protected.

Scope

To realize the full elimination of high level wastes, the proposed scheme should be justified by using the several facilities. Nuclear science laboratories such as RIKEN, NSCL and GANIL are most promising machines to test and to get useful



Fig. 2. Accelerator scheme.

data concerning the technical problems or real transmutation efficiency by using stable isotopes.¹¹⁾

The cost to build these acceleration systems would be about 1 billion yen^{11} , which should be compared to the cost of building for 1000 MWe power plant.

References

- 1) The Journal of Nuclear Engineers and Scientists or Nuclear Technology.
- 2) Management of radioactive wastes from US atomic energy commision's fuel reprocessing operations, Experience and Planning.
- 3) Van Nostrand's Scientific Encyclopedia (6th edition).
- 4) Radioactive Materials ed. by the Chemical Society of Japan, Maruzen.
- 5) A.S. Goldharber and H.H. Heckmann, Ann. Rev. Nucl. Sci. <u>28</u> 161 (1978).
- 6) I. Tanihata, et al., Phys. Rev. Lett. 160B, 380 (1985).
- M. Arnold et al., Proc. of Intnl. Symp. on Heavy Ion Physics and Nuclear Astrophysical Problems, pp.287-293. ed. by S. Kubono et al., Tokyo, 1988.
- 8) T. Nomura, ibid. pp.295-309.
- 9) V. Bechtold, H. Dohrmann and S.A. Sheikh, Proc. of the 7th Workshop on ECR ion sources, Jülich 1986 p.248.
- 10) Numatron., p.83, INS NUMA-5 ed. by Y. Hirao, 1977.
- 11) 11th Int. Conf. on Cyclotrons and their Applications, Tokyo, Oct. 1986, see List of Cyclotrons.

Table 1. Gaseous wastes from Dresden	1.
--------------------------------------	----

		discharg	jing (μCi/s)
Nuclides	Ides Half-life	measured	estimated
⁸⁹ Kr	3.2m		140
137Xe	3.8m		320
¹⁸ N	10m		450
¹³⁵ ^m Xe	15m		370
¹³⁸ Xe	17m	3 000	2 800
87Kr	1.3h	790	1 170
^{83 m} Kr	1.9h		220
⁸⁸ Kr	2.8h	740	1 250
^{85m} Kr	4.4h	400	370
185Xe	9.2 h	1 250	1 310
¹⁸⁸ Xe	2.3d	14	14
183Xe	5.3d	510	380
⁸⁵ Kr	10.7y	0.14	0.60

Table 2. Liquid wastes from Dresden 1.

Nuclides	Half-life	(%)
^{se} Co	71.3 d	35.4
⁶⁰ Co	5.26 y	17.7
89Sr	52.7 d	16.4
90Sr	27.7 у	2.0
181 I	8.05 d	2.0
ынСв	2.05 y	5.5
¹⁸⁷ Cs	30 y	14.4
140Ba—140La	12.8 d	5.6

Table 3. Liquid disposal from NSF fuel reprocessing plant.

Nuclides	Activity(µCi/m)
Groos a	1.2×10 ⁻⁶
Gross 3	1.2×10 ⁻³
зН	4.5×10 ⁻²
106Ru	8.7×10 ⁻⁴
137Cs	7.2×10 ⁻⁵
¹³⁴ Cs	1.9×10 ⁻⁵
⁹⁰ Sr	8.1×10 ⁻⁵
144Ce	4.0×10 ⁻⁶
⁹⁵ Zr	1.4×10^{-7}
[∞] Co	1.1×10 ⁻⁶
125Sb	7.7×10 ⁻⁶
⁵⁴ Mn	3.7×10^{-7}
239Pu	1.5×10 ⁻⁶
²³⁹ Pu	2.9×10 ⁻⁸
241Pu	1.8×10 ⁻⁶
*32U	4.2×10^{-7}
234 U	1.9×10 ⁻⁷
:38U	2.2×10 ⁻⁵