PRAMANA — journal of physics

Vol. 85, No. 3
September 2015
pp. 517–523

Nuclear transmutation strategies for management of long-lived fission products

S KAILAS[1,2,*], M HEMALATHA[2] and A SAXENA[1]

[1]Nuclear Physics Division, Bhabha Atomic Research Centre, Mumbai 400 085, India
[2]UM–DAE Centre for Excellence in Basic Sciences, Mumbai 400 098, India
*Corresponding author. E-mail: kailas@cbs.ac.in

DOI: 10.1007/s12043-015-1063-z; ***e*Publication:** 27 August 2015

Abstract. Management of long-lived nuclear waste produced in a reactor is essential for long-term sustenance of nuclear energy programme. A number of strategies are being explored for the effective transmutation of long-lived nuclear waste in general, and long-lived fission products (LLFP), in particular. Some of the options available for the transmutation of LLFP are discussed.

Keywords. Nuclear transmutation; long-lived fission products; (n, γ) cross-section; EMPIRE.

PACS Nos 28.41.Kw; 25.40.Fq; 24.60.Dr

1. Introduction

It is recognized that for long-term energy security, nuclear energy is an inevitable option [1]. For a sustainable nuclear energy programme, the management of long-lived nuclear waste is very critical. Radioactive nuclei like Pu, minor actinides like Np, Am and Cm and long-lived fission products like ^{79}Se, ^{93}Zr, ^{99}Tc, ^{107}Pd, ^{126}Sn, ^{129}I and ^{135}Cs constitute the main waste burden from a power reactor. In this paper, we shall discuss the management strategies for nuclear waste in general, and long-lived fission products, in particular.

2. Management of nuclear waste

The radioactive nuclei which are produced in a power reactor and which remain in the spent fuel of the reactor form a major portion of nuclear waste. Some of the actinides and fission fragments which are long-lived (units of million years) and which reside in the nuclear fuel (reactor working on uranium as a fuel) are as follows: Actinides: ^{239}Pu (0.024), ^{237}Np (2.1), ^{242}Pu (0.40) and ^{243}Am (0.007); fission products: ^{79}Se (0.295), ^{93}Zr (1.53), ^{99}Tc (0.211), ^{107}Pd (6.5), ^{126}Sn (0.23), ^{129}I (15.7) and ^{135}Cs (2.3). The composition of a 1 ton spent nuclear fuel from a pressurized water reactor (operating at 33 GWd/t and

after 10 years of cooling) is 0.9% Pu, 0.1% minor actinides (MA) and 0.2% long-lived fission products of the total [2].

After the spent fuels are taken out of the core of the reactor, they go through a cooling cycle for short-lived activities to decay. Then Pu, MA and fission products are separated using the chemical method. In countries like India, the nuclear power programme is based on a closed fuel cycle, ie., the Pu produced in the uranium-based thermal reactor (first stage) is used as fuel in the fast reactor (second stage). This way the Pu waste is considerably reduced. Countries like India also have plans to develop Th–U-based reactor in the third stage. By adopting this route instead of the U–Pu fuel cycle, the MA production, in general, gets considerably reduced. On realizing the importance of long-lived nuclear waste management, many countries have programmes where small amounts of the MA and fission products are being incinerated or transmuted in the existing thermal and fast reactors. Dedicated nuclear waste burners and accelerator-driven systems [3,4] are also being considered for reducing the nuclear waste burden in the long term. The main focus of this paper will be on the strategies for the management of long-lived fission products.

3. Requirements for transmutation of long-lived fission products

The foremost requirement is that the half-life of long-lived fission products (LLFP) has to be brought down to a small enough value through the nuclear transmutation route so that the management of irradiated waste through safe storage in containers, etc. will be feasible. Further, this process of conversion has to be accomplished in a time-scale much smaller than the lifetime of the LLFP. This implies that both the nuclear cross-section for the proposed transmutation reaction and the intensity of the projectile have to be as high as possible. This, in turn, will help in choosing the reaction and the projectile type (along with the bombarding energy). It is also necessary that significant amount of LLFP should be incinerated in each campaign of transmutation. As isotope separation of LLFP is not practical, the choice of nuclear reaction should be such that the overall activity after the transmutation reaction should be considerably smaller than the original activity. In trying to reduce the half-life of LLFP, it should not lead to the creation of the same isotope or some others having longer half-lives through nuclear reaction induced on the other stable or short-lived isotopes of the element being transmuted. While the priority of transmutation will depend on the amount of the LLFP produced along with its activity, another factor which should be considered is the radiotoxicity of the LLFP that needs to be transmuted. For example, while the activity of ^{129}I is relatively small because of its large half-life, its biological hazard factor is large as iodine gets selectively absorbed in thyroid. Even though ^{79}Se and ^{126}Sn are not produced in relatively large amounts, the radiotoxicity of these LLFPs is high when compared to the most important LLFPs such as ^{99}Tc, ^{93}Zr, ^{129}I and ^{135}Cs which are produced in larger amounts [5].

4. Strategies for LLFP transmutation

With these requirements, one can proceed with the strategies for effective transmutation of LLFPs. Neutrons [2–7], protons [8], photons [9] (lasers [10]) can be considered as

Table 1. Neutron and proton options for Se isotopes.

Neutron-induced reaction	Proton-induced reaction
^{77}Se(n, γ)^{78}Se (stable)	^{77}Se(p, n)^{77}Br (77 h)
^{78}Se(n, γ)^{79}Se (6.5×10^4 year)	^{78}Se(p, n)^{78}Br (6.5 m)
^{79}Se(n, γ)^{80}Se (stable)	^{79}Se(p, n)^{79}Br (stable)
^{80}Se(n, γ)^{81}Se (57.3 m)	^{80}Se(p, n)^{80}Br (4.4 h)
^{82}Se(n, γ)^{83}Se (22.3 m)	^{82}Se(p, n)^{82}Br (35.3 h)

projectiles for this purpose. Neutrons, both thermal and fast, can be employed for the transmutation of LLFPs, if the neutron capture rates are high.

^{79}Se: The thermal neutron capture value is 0.33 b. However, as seen from table 1, there are problems due to neutron capture on the other isotopes present along with ^{79}Se. For example, while neutron capture of ^{79}Se will lead to stable ^{80}Se, the isotope ^{78}Se will capture neutrons to become ^{79}Se, the isotope we want to burn. The proton-induced reaction cross-sections for these isotopes are on an average around 500 mb in the energy range of 5–15 MeV. As can be seen from table 1, in proton-induced reactions, all the isotopes lead to nuclei which are stable or have short half-lives.

^{93}Zr: Even though the thermal neutron capture cross-sections for ^{93}Zr is reasonably large (1.03 b), the neutron route is not feasible for the following reason: In the case of Zr, while neutron capture of ^{93}Zr will lead to stable ^{94}Zr, along with this there will also be a production of ^{93}Zr from the neutron capture reaction on ^{92}Zr, an isotope which is produced along with ^{93}Zr in the reactor. If we choose the proton-induced reactions like (p, n), once again we end up with the production of long-lived residues in the case of ^{94}Zr and ^{92}Zr. The residual nuclei ^{94}Nb and ^{92}Nb have half-lives of 2×10^4 and 3.6×10^7 years, respectively.

^{99}Tc: The thermal neutron capture cross-section value is 4.2 b. Here there is no problem due to other isotopes as ^{99}Tc is essentially the only isotope produced. The neutron route seems to be the best.

^{107}Pd: Even though the thermal neutron capture cross-section is large (2.79 b), one must also consider the reactions that will take place on the isotopes ^{104,105,106,108,110}Pd (stable) and ^{109}Pd (short-lived) which are all produced in the reactor along with the long-lived ^{107}Pd. For example, while neutron capture can convert the long-lived ^{107}Pd to stable ^{108}Pd, the neutron capture of the stable isotope ^{106}Pd will lead to the production of long-lived ^{107}Pd. In proton-induced reactions ((p, n) reactions), the Ag isotopes produced have relatively short half-lives except ^{108}Ag which has a half-life of 418 years. The typical (p, n) cross-section values are of the order of 700 mb. In the case of ^{107}Pd, the proton option is viable.

^{126}Sn: The thermal neutron capture cross-section is very small, of the order of 0.03 b. One can employ the proton route through the (p, n) reactions. In this case, the cross-section is nearly 800 mb for protons below 15 MeV. Interestingly, all the residual nuclei produced in the (p, n) reactions on the Sn isotopes are short-lived with half-lives less than 3 months. Certainly, the proton-based transmutation should be explored.

^{129}I: The thermal neutron capture cross-section is 3.1 b. In addition to ^{129}I, the only other isotope produced is ^{127}I. Neutron capture of ^{127}I leads to the formation of ^{128}I which decays quickly. The thermal neutron capture route is a good option for the transmutation of ^{129}I.

^{135}Cs: Even though the capture cross-sections for ^{135}Cs is high (2.5 b), the co-produced isotope ^{134}Cs will again lead to the production of ^{135}Cs after neutron capture. This can be a problem. In protons, after the (p, n) reactions, the residual nuclei produced are Ba isotopes which are short-lived. However, the (p, n) cross-sections are of the order of 400 mb in the energy range 10–15 MeV.

5. Choice of transmutation route

The final choice of neutrons or protons will be influenced by the previous considerations. In a few cases, the photon-induced reactions can be considered where the neutron route is ineffective or as an alternative to the neutron route. For example, one can consider the photon-induced reactions in the transmutation of ^{99}Tc and ^{129}I, provided the beam intensities can be enhanced substantially. In table 2 the typical time-scales are given for the reduction of the LLFPs to half of their original inventory.

This is based on the capture or reaction cross-section and the flux or intensity. Certainly these factors can be optimized. In some cases, it becomes necessary to go beyond the first step of transmutation to the succeeding steps if long-lived isotopes are formed from the decay of the radioisotopes formed in the first step. In that case, we need to worry about the transmutation of these long-lived species. The amount of nuclear waste that can be transmuted will also depend on the quantity which can be placed before the neutron, proton or photon beams. In the case of protons, the amount will be decided by the effective range of protons of a given energy in the target medium. In the case of low-energy protons, the proton range in the material is small and hence the amount of material that can be transmuted in a given campaign is limited. In trying to transmute LLFPs in thermal or fast reactors, we have to keep in mind that the introduction of LLFPs should not hamper the functioning or reduce the efficiency of the performance of the reactor. While

Table 2. Thermal neutron capture cross-section and transmutation half-lives of LLFPs. Neutron flux is assumed to be 10^{14} n cm^{-2} s^{-1}.

Isotope	Capture cross-section (b)	Transmutation half-life (years)
^{79}Se	0.33	666
^{93}Zr	1.03	213
^{99}Tc	9.32	24
^{107}Pd	2.79	79
^{126}Sn	0.03	7300
^{129}I	3.12	70
^{135}Cs	2.48	89

the low-energy, high-intensity protons are useful in transmutation programmes in general, and in a few cases perhaps the only option available, this method suffers from the serious limitation of the amount of LLFPs which can be burnt. As mentioned earlier, the range of protons in the material decides the amount which can be effectively transmuted. The neutrons and photons do not have this limitation. In fact, the photon has the advantage as in this case the LLFPs are burnt outside and do not have constraints as burning inside a reactor.

6. Status of nuclear data

In general, while the nuclear cross-sections of interest for transmutation of LLFPs are available for stable isotopes, the required data for the radioactive LLFPs are rather sparse.

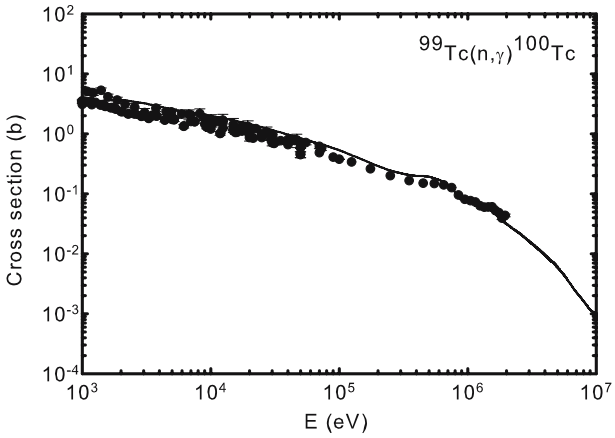

Figure 1. Calculated (n, γ) cross-section for ^{99}Tc. Experimental data [15] are indicated by filled circles.

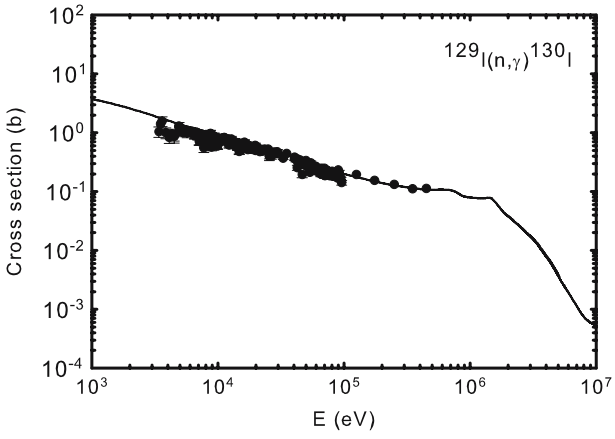

Figure 2. Calculated (n, γ) cross-section for ^{129}I. Experimental data [15] are indicated by filled circles.

Detailed comparison of the (p, n) and (n, γ) cross-section data for the stable and unstable LLFPs (where data are available) with statistical model predictions will be necessary to extend the calculations to LLFPs and energies where data are not available. In figures 1 and 2 we have shown the comparison between the data and the corresponding calculations for (n, γ) reactions on ^{99}Tc and ^{129}I. The data for thermal neutron energies are from [11,12]. The calculations have been carried out using the statistical model code EMPIRE [13]. The proton and neutron global optical model parameters are taken from Koning and Delaroche [14] available as an option in the EMPIRE code. Both the discrete and continuum levels for various nuclei have been considered. The nuclear masses are the experimental values as provided in the code. Calculations of reaction cross-sections using the EMPIRE (v 3.2) code were performed by taking into account the pre-equilibrium exciton model processes (PCROSS = 1.5). EMPIRE-specific level densities [13] (including adjustment to discrete levels) have been chosen. Cross-sections for the (n, γ) reaction have been calculated from the thermal neutron energy 1 keV–10 MeV for ^{99}Tc and ^{129}I and they are shown in figures 1 and 2. Experimental data [15], in the energy region of interest, where available, are shown by filled circles. It is seen that the calculations for both ^{99}Tc and ^{129}I agree with the data between energies 1 keV and 1 MeV.

7. Conclusion

It is clear from the discussion here that all the three options viz., the neutron, the proton and the photon routes to transmutation should be explored. Each method has its advantages and limitations. The key driving factors are reducing the overall activity finally, by reducing the half-life and the radiotoxicity. Various factors discussed in this paper will influence the final choice of methodology for transmutation. The cross-section data are available for the stable isotopes. It is desirable to measure the same for the radioactive species of long-lived fission products (LLFP). In addition, we must also consider the various options of inertial matrix to host the LLFPs for transmutation reactions. This is very important to contain the LLFPs after incineration or transmutation in a matrix or substrate so that the activity will not escape to the environment. More sustained efforts are required to address the various issues related to transmutation of LLFPs.

Acknowledgement

SK would like to acknowledge DAE for the support through the Raja Ramanna fellowship.

References

[1] *Science and culture* edited by S Kailas and R K Sinha (Indian Science News Association, 2013) Vol. 79, Nos 1–2

[2] Physics and safety of transmutation systems – a status report, NEA-6090, OCED (2006) ISBN 92-64-0182-3

[3] *Accelerator-driven system* edited by S Kailas (Indian Nuclear Society Publication, 2009)

[4] *Proc. of the Workshop on Physics of Accelerator Driven Sub-critical System for Energy and Transmutation* edited by V Kumar, P K Nema, S Kailas, S B Degwekar and S Ganesan (University of Rajasthan, Jaipur, 2006); *Pramana – J. Phys.* **68**, 141 (2007)
[5] W S Yang, Y Kim, R N Hill, T A Taiwo and H S Khalil, *Nucl. Sci. Eng.* **146**, 291 (2004)
[6] C J Jeong and W I Ko, *Energ. Convers. Manage.* **49**, 1917 (2008)
[7] W Maschek *et al*, *Energ. Convers. Manage.* **49**, 1810 (2008)
[8] A Hermanne, *J. Nucl. Sci. Tech. (suppl.)* **2**, 1202 (2002)
[9] K Imasaki *et al*, *Energy Convers. Manage.* **49**, 1922 (2008)
[10] J G Chen *et al*, *Chin. Phys. C* **32**, 677 (2008)
[11] S Nakamura *et al*, *J. Nucl. Sci. Technol.* **33**, 283 (1996)
[12] T Katoh *et al*, *J. Nucl. Sci. Technol.* **34**, 431 (1997)
[13] EMPIRE: Nuclear Reaction Model Code System for Data Evaluation, M Herman, R Capote, B V Carlson, P Oblozinsky1, M Sin, A Trkov, H Wienke and V Zerkin, *Nucl. Data Sheets* **108**, 2655 (2007)
[14] A J Koning and J P Delaroche, *Nucl. Phys. A* **713**, 231 (2003)
[15] https://www-nds.iaea.org/exfor/exfor.htm