# TRANSMUTATION OF CESIUM-135 WITH FAST REACTORS

**Shigeo Ohki** and **Naoyuki Takaki**
O-arai Engineering Center
Japan Nuclear Cycle Development Institute (JNC)
4002, Narita-cho, O-arai-machi, Higashi-Ibaraki-gun, Ibaraki, 311-1393, Japan

## Abstract

The neutronic feasibility has been studied for $^{135}$Cs transmutation with commercial fast reactors. By irradiating Cs, the existence of stable isotope $^{133}$Cs deteriorates an effective net reduction of $^{135}$Cs owing to new creations of $^{135}$Cs by double neutron capture reactions, because Cs recovered by an element-wise separation process contains the associated isotopes $^{133}$Cs, $^{134}$Cs and $^{137}$Cs except $^{135}$Cs. The following three issues were examined to improve the transmutation efficiency: (1) selection of proper moderator material, (2) element-wise separation after the decay of $^{137}$Cs, (3) irradiation-cooling repeating method. Consequently, the transmutation efficiency was improved somewhat but not to attain break-even condition. It turns out that the $^{135}$Cs transmutation is difficult with commercial fast reactors.

## Introduction

Cesium-135 ($^{135}$Cs) is an important target nuclide for nuclear transmutation, next to iodine-129 and technetium-99 among long-lived fission products. Feasibility of practical transmutation of $^{135}$Cs in fast reactors was investigated from the viewpoint of reactor physics. One of requirements is that a reactor can transmute at least the same amount of $^{135}$Cs as its self-creation by fuel burning, which we call "break-even" condition. Referring a large-sized commercial fast reactor, we chose a transmutation manner not to deteriorate the core performance as a power reactor considerably. It was assumed that the cesium was recovered from spent fuel by the element-wise separation technology. Though the transmutation of $^{135}$Cs with element-wise separation has the problem that the associated isotope $^{133}$Cs produces a significant amount of $^{135}$Cs, the application of the isotope-wise separation technology is far from practical implementation. We made efforts to transmute $^{135}$Cs with element-wise separation by using a commercial fast reactor.

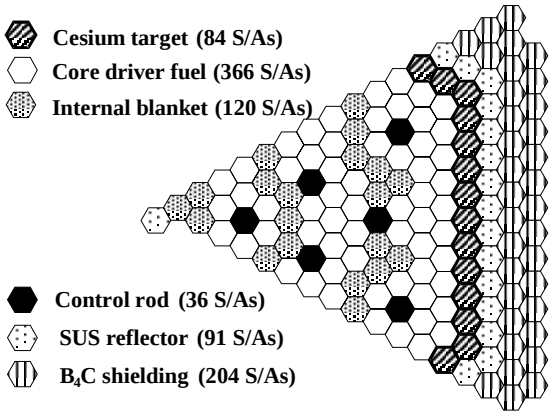
## Calculation conditions

Let us suppose the transmutation of $^{135}$Cs is carried out in neutron-moderated target subassemblies loaded on the peripheral region of an active core. In this work a 3 800 $MW_{th}$ commercial LMFBR core concept [1] was employed as a reference core. Table 1 summarises the main design parameters of the reference core and the cesium target subassembly. Loading pattern of the cesium target subassemblies is illustrated in Figure 1. The chemical form of cesium was tentatively assumed to be $Cs_2CrO_4$. Zirconium hydride ($ZrH_{1.65}$) was selected as a moderator at the beginning of the investigation. The aim of this loading method is to enhance the $^{135}$Cs neutron capture reaction rate by the moderator, with avoiding the influence on core characteristics as well as utilising the neutrons left from the core effectively.

Table 1. **Main design parameters of the reference LMFBR and the cesium target subassembly**

| Design parameters | Data |
|---|---|
| Reactor thermal power | 3 800 $MW_{th}$ |
| Operation cycle length | 18 months |
| Core concept | Radial heterogeneous |
| Fuel exchange batch (core/blanket) | 4 / 4 |
| Average fuel burn-up | 150 $GW_{th}d/t$ |
| Fuel material | $(U, Pu)O_{1.98}$ |
| Coolant | Sodium |
| Core diameter / Core height | 487 / 100 cm |
| Axial blanket thickness (upper/lower) | 30 / 30 cm |
| Cesium isotopic composition (at%) $^{133}$Cs/$^{134}$Cs/$^{135}$Cs/$^{137}$Cs | 32.3/0.6/37.4/29.7 |
| Chemical form of cesium target | $Cs_2CrO_4$ |
| Smeared density of cesium target | 50% |
| Neutron moderator | $ZrH_{1.65}$ |
| Smeared density of moderator | 90% |
| Stack length | 160 cm |
| Target exchange batch | 4 (tentative) |

Figure 1. **Loading pattern of cesium target subassemblies** (1/6th core)




Reactor core characteristics were calculated based on a two-dimensional 18-group diffusion approximation with burn-up chain model for heavy metal and cesium isotopes. Since the used constant sets are oriented for conventional fast reactor calculation, the present calculation respect to the moderated target subassembly is nothing more than a rough estimation. The cesium target subassembly was modelled by the homogeneous representation. Self-shielding effect of cesium resonance absorption in epi-thermal energy region was treated, while the aspects needed for thermal neutron calculation (upward scattering, chemical binding effect on scattering) were not taken into account sufficiently.

## Transmutation property of $^{135}$Cs

### *Isotope-wise separation case*

First of all, we are going to examine the transmutation property of $^{135}$Cs with applying isotope-wise separation, which gives us the upper limit of $^{135}$Cs transmutation when dealing with element-wise separation of Cs. Figure 2 shows the transmutation amount and rate per reactor operating cycle for $^{135}$Cs separated in isotope-wise. They are parameterised by the $ZrH_{1.65}$ moderator fraction and the number of target subassemblies. The largest transmutation amount (110 kg/cycle) appeared at moderator fraction of 10~20%. It was possible to transmute more amounts of $^{135}$Cs than that created from the core fuel (about 80 kg/cycle) by one ring of target subassemblies plus more. Note that larger loading amount resulted smaller transmutation rate per cycle. The small transmutation rate means that the core residence time of the targets should be taken long.

### *Element-wise separation case*

Transmutation property of $^{135}$Cs with element-wise separation of Cs is shown in Figure 3. The trend of the transmutation curves changed completely being compared with the isotope-wise separation case. Though the transmutation amount was maximised at moderator fraction of 10~20%, we could obtain only small value not over 20 kg/cycle. In addition, the transmutation rate did not increase with the moderator fraction. As well known, those are caused by the newly produced $^{135}$Cs from $^{133}$Cs (see the nuclide chain in Figure 4). Moderator fraction dependence of 1-group neutron capture cross-section for cesium isotopes is shown in Figure 5. It is found that the increase of the capture cross-sections for $^{133}$Cs and $^{134}$Cs by raising the moderator fraction is more notable than that of $^{135}$Cs, which is the fundamental reason for the worse transmutation property.

Figure 2. **Transmutation property of $^{135}$Cs with $ZrH_{1.65}$ moderator** (isotope-wise separation case)

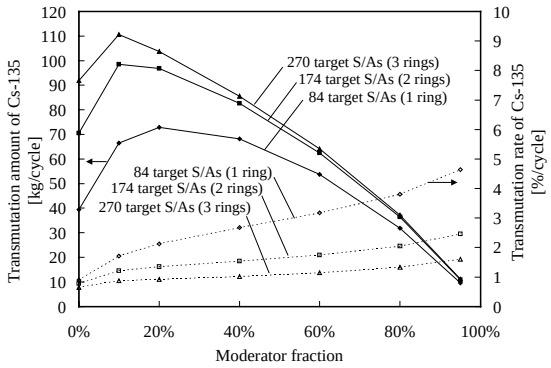
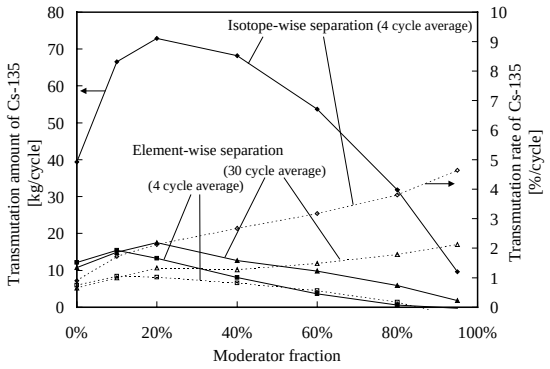
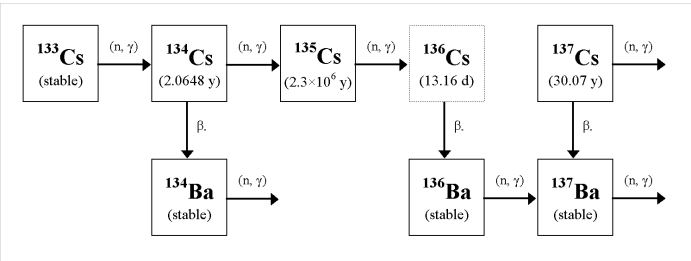



Figure 3. **Transmutation property of $^{135}$Cs with $ZrH_{1.65}$ moderator by 84 target sub assemblies** (element-wise separation case)




It is possible to achieve the break-even condition by loading a lot of cesium target subassemblies from the viewpoint of the number of excess neutron. [2] However, the transmutation rate per cycle extremely decreases with the large loading amount. If the overall transmutation rate during the target lifetime is small, the amount of $^{135}$Cs transferred into waste as a recovery loss in multi-recycling exceeds the transmuted amount in a reactor, which cannot be regarded as a meaningful transmutation manner. [3] To increase the overall transmutation rate, residence time of the target subassemblies in a reactor might be taken long until $^{133}$Cs disappears, however, the calculation result for long irradiation period (30 cycles) in Figure 3 exhibits that we can only expect a discreet improvement.

Figure 4. **Nuclear reaction chain for cesium isotopes**




Time length in parenthesis stands for the half life of natural decay. Note that the half life of $^{134}$Cs (about 2 years) is the same order as the analogical half life of its capture reaction, $\ln 2 / \left(\sigma_{(n,\gamma)}\phi\right)$.

Hereafter, we are going to find the way to obtain better transmutation property with usual residence time of target subassemblies (e.g. several cycles) in which it is easier to keep integrity of the targets. Avoiding any significant influence on core performance, is it possible to cope with the difficulty in $^{135}$Cs transmutation with element-wise separation?

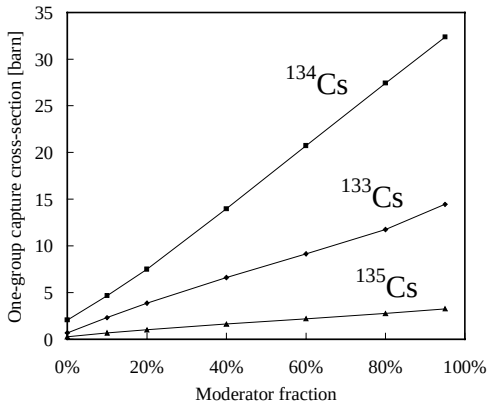
## Improvements in $^{135}$Cs transmutation with element-wise separation

In this section, methods for improving the $^{135}$Cs transmutation property with element-wise separation are described. The examined ideas are the followings:

- using weak neutron moderator such as $^{11}B_4C$, BeO, $Al_2O_3$ and SiC;

- increasing the isotopic fraction of $^{135}$Cs by delayed element-wise separation after almost all $^{137}$Cs decays to $^{137}$Ba;
- applying the irradiation-cooling repeating method.

Figure 5. **Capture cross-section for cesium isotopes as a function of moderator ($ZrH_{1.65}$) fraction**




***Weak neutron-moderator***

From Figure 5 it was found that the neutron moderation by hydride was too strong. Weaker neutron moderation would rather make the capture cross-section for $^{135}$Cs meaningful magnitude compared to those for $^{133}$Cs and $^{134}$Cs. Additionally, suppression of decrease in neutron flux level at the target region could also be expected with weak moderation. Then, the method for constituting the neutron spectrum suitable for $^{135}$Cs transmutation by using non-hydride moderator was investigated.

The results of examining the transmutation property are shown in Figure 6, where $^{11}B_4C$, BeO, $Al_2O_3$ and SiC came up for weak neutron moderator. Being compared with the case of $ZrH_{1.65}$, the weak neutron moderators have the transmutation property improved to some extent. $^{11}B_4C$ and BeO brought about better performance than $Al_2O_3$ and SiC. These were depending on the profile of neutron spectrum and the neutron flux level. The comparison of neutron spectrum for each moderator is shown in Figure 7, whilst the moderator dependence of neutron flux level and capture cross-section of cesium isotopes are illustrated in Figure 8. From these figures, we can explain the differences in transmutation property observed among various moderators as below:

- Neutron-moderation by $Al_2O_3$ or SiC was not enough.
- The adequate neutron spectrum seemed to be given by $^{11}B_4C$, BeO as well as the small fraction of $ZrH_{1.65}$, where the capture cross-sections for $^{133}$Cs and $^{134}$Cs did not increase too much.
- Higher neutron flux level was obtained by using 11B4C or BeO in comparison with the use of $ZrH_{1.65}$.

In this way, it is concluded that weak neutron moderation has an advantage to $^{135}$Cs transmutation with element-wise separation. However, the improvements were not so significant that could accomplish the break-even condition.

Figure 6. **Transmutation property of $^{135}$Cs with weak neutron moderators**
(element-wise separation case)

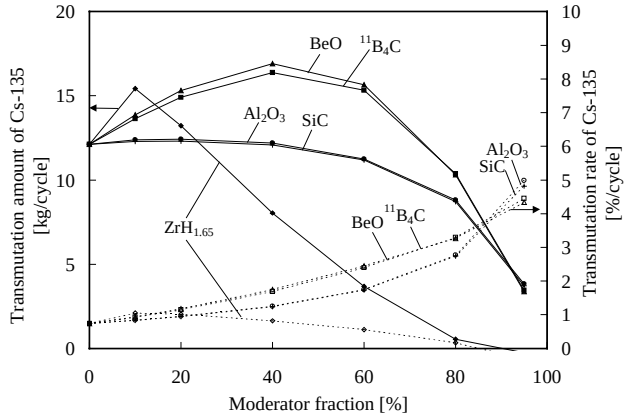
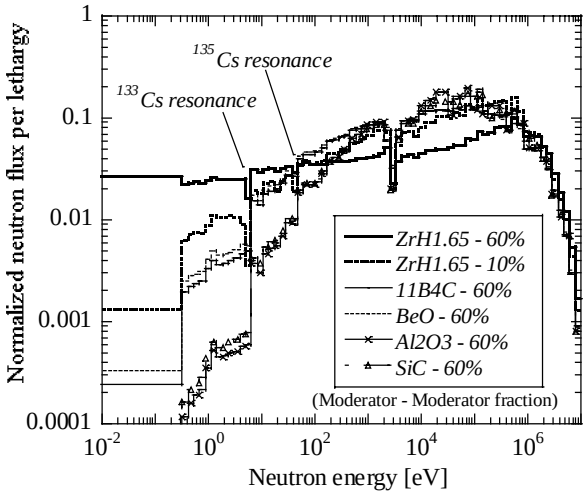



Figure 7. **Neutron spectra made by various neutron moderators**

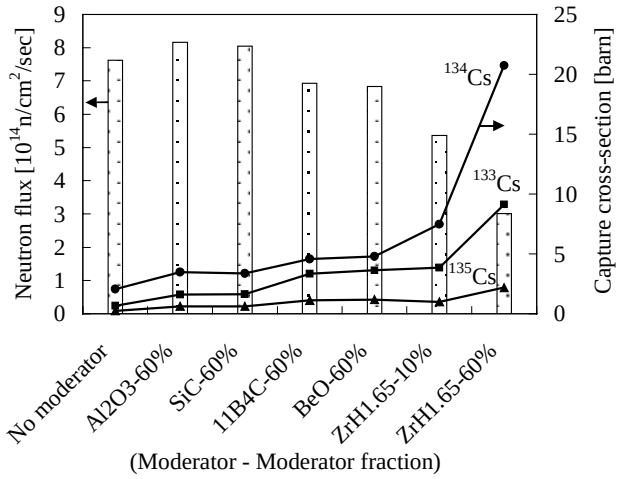



Considering the influence on the core characteristics, the non-hydride moderators have smaller effect than $ZrH_{1.65}$ (see Case-1 and Case-2 in Table 2). One may try to load cesium targets with weak neutron-moderator on active core region in order to utilise much higher neutron flux level. However it will effectively deteriorate the neutron economy of the core. Case-3 in Table 2 exhibits one example of target loading on core region. It cannot be said that the small increments in transmutation amount pay the worsening core characteristics.

***Delayed element-wise separation***

One of the problems in the $^{135}$Cs transmutation with element-wise separation is a number of isotopes existing other than $^{135}$Cs. By waiting a sufficient time (for several decades) before the separation so that most of $^{137}$Cs decay to $^{137}$Ba, it is possible to increase $^{135}$Cs inventory in target subassemblies by about 40%. Case-4 in Table 2 shows transmutation property in the case of delayed element-wise separation, in which the transmutation amount has improved by 24% compared to Case-2.

Figure 8. **Moderator dependence of neutron flux and one-group capture cross-section for cesium isotopes**




***Irradiation-cooling repeating method***

As a final measure, the irradiation-cooling repeating (ICR) method [4] was employed. It is applying the natural decay of $^{134}$Cs (half life: about 2 years) to reduce the amount of newly created $^{135}$Cs. Demerit of ICR method is that the handling of target subassemblies turns out to be extremely complicating. Case-5 in Table 2 exhibits the calculation result for ICR method, where 1 cycle irradiation and 3 cycle cooling were repeated alternately. It was found that the improvement was just the increase by 18% both for transmutation amount and rate.

Table 2. **Investigation of improvement methods for the transmutation property of $^{135}$Cs when applying element-wise separation of Cs**

| Calculation case | Ref. | Case-1 | Case-2 | Case-3 | Case-4 | Case-5 |
|---|---|---|---|---|---|---|
| Number of cesium target S/As (radial blanket region / inner blanket region) | 0 / 0 | 84 / 0 | 84 / 0 | 84 / 21 | 84 / 0 | 84 / 0 |
| Neutron moderator | – | $ZrH_{1.65}$ | $^{11}B_4C$ | $^{11}B_4C$ | $^{11}B_4C$ | $^{11}B_4C$ |
| Moderator fraction | – | 10% | 60% | 60% | 60% | 60% |
| Delayed element-wise separation | – | No | No | No | Yes | Yes |
| Irradiation-cooling repeating | – | No | No | No | No | Yes* |
| Transmutation property of $^{135}$Cs | | | | | | |
| Amount [kg/cycle] | – | 15.4 | 15.3 | 16.8 | 19.0 | 22.5 |
| Rate [%/cycle] (1 cycle = 18 months) | – | 1.0 | 2.4 | 2.1 | 2.1 | 2.5 |
| Influence on core characteristics | | | | | | |
| Pu enrichment (Pu/HM) [wt%] | 27.6 | 28.5 | 28.0 | 28.4 | 28.0 | – |
| Burnup reactivity %.k/kk'] | 3.25 | 3.60 | 3.56 | 4.12 | 3.56 | – |
| Breeding ratio | 1.25 | 1.12 | 1.13 | 1.07 | 1.13 | – |
| Radial peaking factor | 1.21 | 1.27 | 1.23 | 1.20 | 1.23 | – |
| Maximum linear heat rate [W/cm] | 398 | 439 | 408 | 404 | 412 | – |
| Average fuel burnups [$GW_{th}$d/t] | 149 | 152 | 153 | 156 | 153 | – |

* One cycle irradiation and three cycle cooling were repeated for four times.

From the above investigations for weak neutron moderator, delayed element-wise separation and the ICR method, it was turned out that each of them has an effect on the improvement of $^{135}Cs$ transmutation property. However, these improvements were absolutely inadequate for the accomplishment of the break-even condition as well as the practical residence time of target subassemblies.

## Conclusion

The $^{135}Cs$ transmutation property in a commercial fast reactor was studied in this paper. As long as we assume the application of element-wise separation for recovery of cesium, weak neutron moderator such as $^{11}B_4C$ is suitable for the transmutation rather than hydride metal, since it can suppress the newly creation of $^{135}Cs$ from $^{133}Cs$. Both the delayed element-wise separation and the irradiation-cooling repeating method contribute to an improvement of $^{135}Cs$ transmutation efficiency, which are utilising the beta decay of $^{137}Cs$ and $^{134}Cs$, respectively. However, difficulty has turned out to be obvious in achieving the break-even condition between the transmuted amount and the simultaneous creation from the core fuel. In order to accomplish the break-even condition, a large number of cesium target subassemblies have to be loaded in a reactor with small transmutation rate per cycle. Then, the residence time of the target subassemblies must be taken extremely long for a meaningful transmutation to avoid a significant recovery loss in the cesium recycling. At least from the viewpoint of reactor physics, it can be said that the transmutation of $^{135}Cs$ is difficult with commercial fast reactors. Right or wrong of $^{135}Cs$ transmutation in commercial fast reactors should be argued in consideration of lower priority compared with the other long-lived fission products like $^{129}I$ and $^{99}Tc$, as well as the difficulty in overcoming the above issues.

## REFERENCES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[4] N. Takaki and S. Ohki, *Feasibility and Challenges of LLFP Transmutation in Fast Reactor*, Proc. Int. Conf. on Future Nuclear Systems (GLOBAL2001), September 9-13, 2001, Paris, France.