ANALYSIS OF COBALT-60 PRODUCTION EXPERIMENT IN THE FAST REACTOR PHENIX

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ABSTRACT

An effective transmutation of long-lived fission products (LLFP) in the fast reactor can be achieved by the neutron-moderated target subassembly loaded on core peripheral region. It is necessary to establish a method for neutronics calculation in such a subassembly where the spatial and energy distributions of neutron flux are considerably varied. In order to find out present uncertainty of calculation and residual problems, an analysis of the cobalt-60 production experiment in the PHENIX reactor was carried out. In this experiment the cobalt target subassembly was placed in the third row of the radial blanket and irradiated for 72.2 EFPD with full thermal power of 563 MW. All the samples were natural cobalt surrounded by calcium hydride hollow cylinder.

A continuous energy Monte Carlo code MVP was employed for neutron transport calculation together with the nuclear data JENDL-3.2. A static whole core calculation was performed to obtain the capture reaction rate of cobalt-59, where the target subassembly was modeled as built to treat its heterogeneity correctly. It is found that the calculation gave an agreement with experiment in ±10%. A slight overestimation of the spatial shielding effect was observed in the radial direction. Discrepancy of C/E values also existed in the axial direction. In order to explain these discrepancies, error estimation for both experiment and calculation was performed.

1. INTRODUCTION

The transmutation of LLFP, especially for iodine-129, is considered to be an effective means to reduce the practical long-term risk arisen from the disposed nuclear waste, because LLFP has relatively higher mobility underground. Transmutation concepts by using fast reactor have been proposed and investigated, which are making use of the excess neutrons and the smaller impact on the core characteristics than thermal reactor. These concepts are employing the neutron-moderated target subassembly loaded on core peripheral region in order to increase the transmutation efficiency with a little deterioration of core characteristics.

Calculation method for such a fast and thermal coupled system has not been verified sufficiently. It is necessary to treat the neutron flow into LLFP target region, rapid neutron thermalization as well as the target heterogeneity. There are two types of calculation method, deterministic one and Monte Carlo one. The deterministic method is favorable for design-oriented works because of its shorter calculation time, while the Monte Carlo method can obtain an exact solution except for the errors coming from nuclear data and calculation statistics. Therefore it is preferred to construct a calculation model for using deterministic codes, referring to the Monte Carlo method. The deterministic method also makes it possible to perform the perturbation analysis for physical interpretation.

This paper presents the result of experimental validation of Monte Carlo method by using the cobalt-60 production experiment[1] performed at the French prototype fast reactor "PHENIX" in the late 80's. The reaction process $^{59}Co(n,\gamma)^{60}Co$ and the utilization of neutron moderator (CaH_2) are similar to the concepts considered in the up to date LLFP transmutation study. It is the unique irradiation experiment of neutron-moderated target subassembly using a power fast reactor until now. The most accurate C/E (calculation to experiment) value was obtained by Monte Carlo code with as-built geometrical modeling. The errors for both experiment and calculation were examined in order to discuss the present calculation accuracy. These considerations are going to be the fundamental remarks on the development of calculation method using deterministic codes.

2. EXPERIMENT

The ^{60}Co production experiment analyzed here is originally called COMMODORE-4 experiment, which is the part of the series of experimental studies aiming at optimizing the production of ^{60}Co in PHENIX for use of industry. This irradiation consisted in the cobalt samples loaded in a special subassembly containing neutron-moderating material. One cobalt sample is made of natural cobalt (^{59}Co) of 27.8 gram in weight and has the cladding (diameter 8 mm, length 105.1 mm). The cobalt samples are contained in a capsule placed in the central hole left by the moderator as shown in Figure 1. The hollow disks of pure calcium hydride (CaH_2) are used as a neutron moderator. The irradiation position of the subassembly is the third row of the radial blanket region.

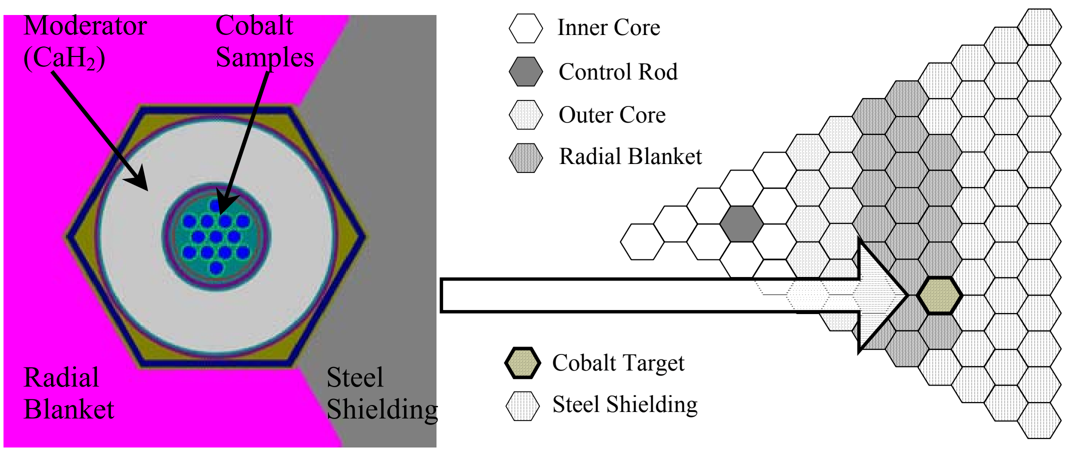

Figure 1. A schematic view of the cobalt target subassembly loaded in the PHENIX core

As illustrated in Figure 2, there are eight axial levels of cobalt sample loadings (numbered 1 to 8 from top to bottom). The stack length of the cobalt samples is about 85 cm. The level 5 is centered on the fissile zone mid-plane. Each level holds between 3 to 13 cobalt samples placed on a hexagonal grid, that is, at most there are 1 sample in the central ring, 6 samples in the intermediate ring and 6 samples occupying half of the outer ring. These different loading patterns are to be used for the examination of the radial (ring) dependence of the spatial neutron shielding effect. Totally 76 cobalt samples are contained in the subassembly.

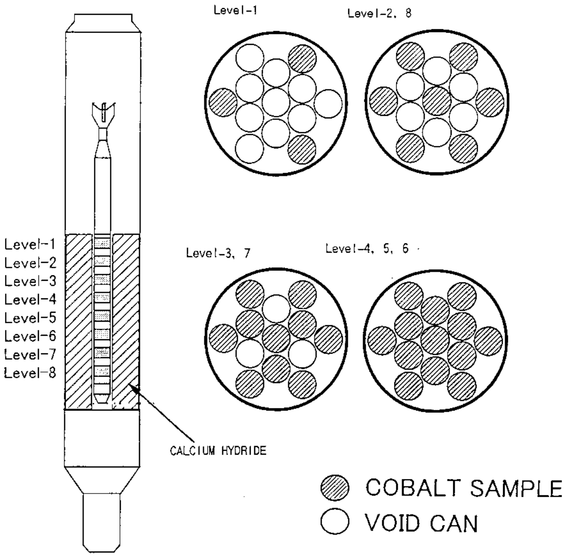

Figure 2. Loading pattern of the cobalt samples

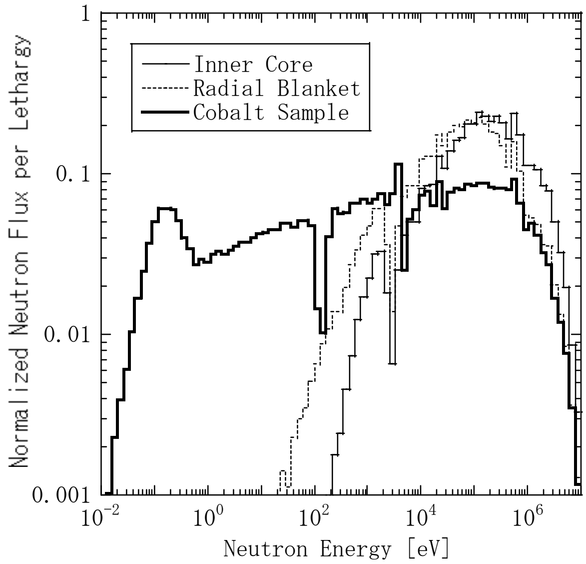

Figure 3. Neutron spectra in different regions

The cobalt target subassembly was irradiated for 72.2 EFPD with full thermal power of 563 MW. Activity of ^{60}Co was measured by gamma-ray detection after the irradiation.

Figure 3 shows the predicted neutron spectrum in the cobalt sample region comparing with those in inner core and radial blanket regions. By neutron moderation the Maxwell distribution is appearing in the cobalt sample region. We can also see the resonance absorptions of ^{59}Co around 100 eV and 10 keV of neutron energy.

3. METHOD OF CALCULATION

A continuous energy Monte Carlo code MVP[2] was employed for neutron transport calculation together with the evaluated nuclear data library JENDL-3.2[3]. A static whole core calculation was performed to obtain the capture reaction rate of ^{59}Co, where the target subassembly was modeled as built to treat its heterogeneity correctly. Ten independent MVP calculations were carried out changing the initial random number. The total number of particle history ran up to 200 million. The mean value and its standard deviation over the ten results were obtained, where the results exceeding 2σ were not included in final evaluation. As a result, the ^{59}Co capture reaction rate was accompanied by the statistic error of 1 to 3%.

Since the actual temperature distribution for the cobalt samples and the moderator are unknown, a temperature of $500\,^{\circ}C$ was approximately assumed in the present calculation. The sensitivity in changing the temperature is going to be discussed in Section 5.

The effect of chemical binding should be considered in thermal neutron physics. However, the scattering law data of hydrogen bound in calcium hydride is not available in the present data library.

Alternatively, the scattering cross-section of hydrogen bound in zirconium hydride was applied in the analysis, assuming a similarity of crystal structure. In Section 5, we are going to make a brief estimation of the ambiguity of this prescription.

The calculated neutron flux was normalized so that an energy release coincided with the reactor power. Fission induced energy and capture gamma heat for each heavy nuclide were taken into account, while the heat production due to structural materials were neglected.

The production amount of ^{60}Co was evaluated by the following expression:

$$N_{Co60}\left(t\right)=\frac{N_{Co59}\left(0\right)\sigma_c^{Co59}\phi}{\sigma_c^{Co59}\phi-\sigma_c^{Co60}\phi-\lambda_{Co60}}\left(Exp\left[-\left(\sigma_c^{Co60}\phi+\lambda_{Co60}\right)t\right]-Exp\left[-\sigma_c^{Co59}\phi t\right]\right), \tag{1}$$

where N, σ_c, ϕ, λ and t stand for nuclide concentration, capture cross-section, neutron flux, decay constant and time, respectively. The $\sigma_c\phi$ for ^{59}Co was obtained from the MVP static calculation, which lay on $1\times10^{-9} \sim 4\times10^{-10}\sec^{-1}$. We have to take the decay constant of ^{60}Co ($\lambda = 4.17\times10^{-9}\sec^{-1}$) into account because it has the same order as its production rate. While the $\sigma_c\phi$ for ^{60}Co is small ($<10^{-11}\sec^{-1}$) enough to be neglected.

4. RESULTS

Table I summarizes the measured activities and the corresponding calculation results. The measured activities are corrected to the values at the end of irradiation. These are listed in terms of every axial level and averages over each radial ring.

Table I. The measured activity and the C/E value at the end of irradiation

Level	Ring	Number of sample	Measured activity [Ci/g]	C/E value (Std.*)
1	Outer	3	5.67	1.05 (1.8%)
2	Outer	6	5.85	1.11 (1.4%)
	Central	1	5.43	1.10 (2.9%)
3	Outer	6	6.05	1.08 (0.7%)
	Intermediate	3	5.08	1.06 (1.5%)
	Central	1	4.76	1.02 (1.4%)
4	Outer	6	6.29	1.05 (1.0%)
	Intermediate	6	4.60	1.05 (1.0%)
	Central	1	3.49	1.02 (2.9%)
5	Outer	6	6.48	1.02 (0.9%)
	Intermediate	6	4.73	1.03 (0.9%)
	Central	1	3.60	0.96 (1.6%)
6	Outer	6	6.00	1.04 (0.9%)
	Intermediate	6	4.42	1.04 (1.9%)
	Central	1	3.35	0.99 (3.9%)
7	Outer	6	5.80	1.01 (1.4%)
	Intermediate	3	4.83	0.98 (1.1%)
	Central	1	4.51	0.96 (2.1%)
8	Outer	6	5.24	1.01 (1.5%)
	Central	1	4.92	0.98 (3.3%)

* Statistic error of MVP calculation

It is found that the calculation gave an agreement with the experiment in ±10%, which is quite satisfactory for global prediction of the neutron behavior in a moderated target subassembly. Decrease of C/E value in the central sample ring was observed, that is, the present calculation slightly overestimated the spatial shielding effect in the radial (ring) direction. About 10% of discrepancy in C/E values were also found in the axial direction, especially in the higher axial positions. These inconsistencies for both radial and axial directions inside the target should be solved.

5. ERROR ESTIMATION

In this section, the results of error estimation for both experiment and calculation modeling are described, so that we can make a consideration of the discrepancy observed in C/E values.

(1) Experimental error
About 3% of experimental error has been reported from the gamma-ray and sample mass measurements.

(2) Flux normalization
At least more than 2% of overall uncertainty would be expected considering the error sources coming from flux monitoring in PHENIX, energy release data, cross-section and fuel number density change by burn-up.

(3) Modeling error for thermal neutron spectrum
The thermal neutron spectrum depends on the temperature as well as the molecular structure of the moderating material. Since the present analysis was applying both an approximate temperature and the hydrogen cross-section for ZrHx as a substitution of CaHx, the effects of these treatments were examined by changing input parameters of the MVP code.

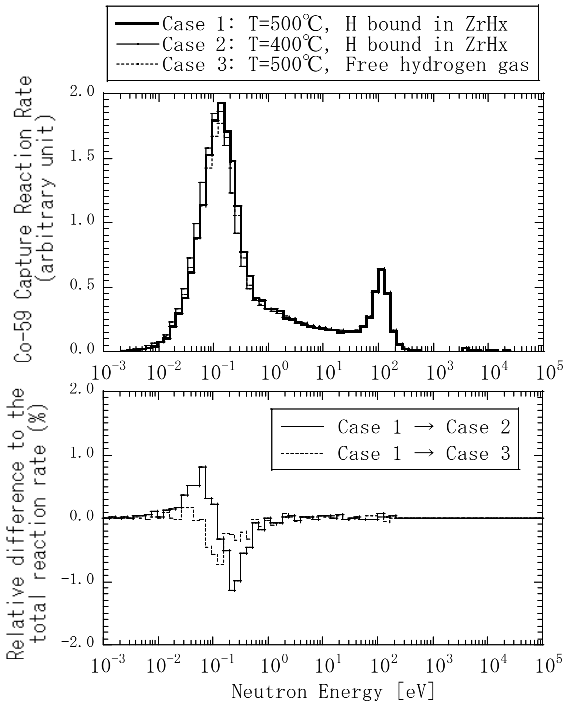

Figure 4. Effects of temperature and chemical binding of hydrogen on ^{59}Co capture reaction rate

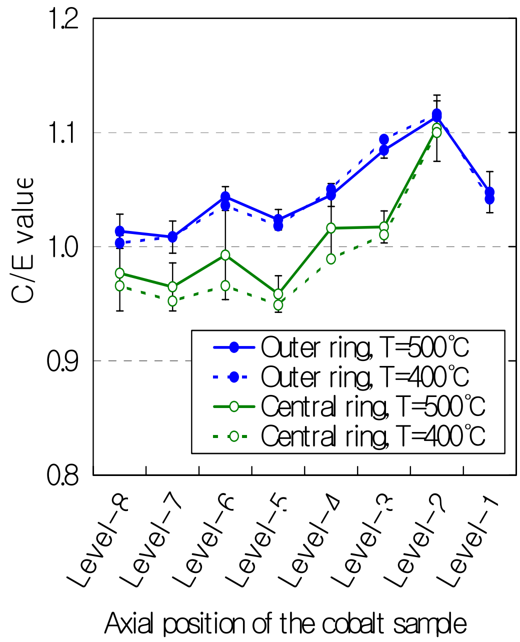

Figure 5. Temperature dependence of the C/E values in axial direction
Error bar: Statistic error of MVP calculation (1 σ)

a. Temperature dependence

The results of MVP calculation are presented in Figure 4, where the temperature of cobalt sample and moderator has been changed from $500\,^{\circ}C$ to $400\,^{\circ}C$. With the decrease of temperature, the Maxwell distribution of reaction rate was shifted towards the lower energy side, nevertheless the total reaction rate was almost unchanged. Moreover there was little difference in resonance self-shielding effect. Therefore, as shown in Figure 5, the axial dependence of C/E values can not be removed if we set a more realistic temperature distribution.

b. Effect of chemical binding

In order to deduce the modeling error of chemical binding for hydrogen, the reference calculation using hydrogen cross-section for ZrHx was compared with the case applying the cross-section of free hydrogen gas. As shown in Figure 4, about 4% of decrement in ^{59}Co capture reaction rate was observed when the free gas model was used. Though it might be possible to expect the similarity of hydrogen cross-section between CaHx and ZrHx, it had better to retain at most 4 or 5% of uncertainty.

(4) Cross-section induced uncertainty

In order to investigate the effects of nuclear data uncertainty on the ^{59}Co capture reaction rate, the cross-section sensitivity was calculated in nuclide and reaction-wise by using the SAGEP code[4] based on the generalized perturbation theory. The calculation was performed with diffusion approximation, 3-dimensional XYZ representation and the 18 energy group structure used in fast reactor analysis. Heterogeneity of the cobalt target subassembly was not taken into account.

Let us consider the quantity related to the ^{59}Co capture reaction rate:

$$R = \sum_g \sigma_{c,g}^{Co59} \phi_g \Bigg|_{target} \approx \frac{\sum_g \sigma_{c,g}^{Co59} \phi_g}{\sum_{g'} \phi_{g'}} \Bigg|_{target} \times \frac{\Phi_{target}}{\Phi_{core}} \times \Phi_{core}, \tag{2}$$

where $\Phi = \sum_g \phi_g$ is total neutron flux, and g stands for energy group. The right-hand-side of Eq.(2) is factorized into three parts. The first one is proportional to the ^{59}Co capture rate with normalized flux, which contains the direct sensitivity component (i.e. the ^{59}Co capture cross-section) and the indirect effect via neutron spectrum change. The second factor is the ratio of flux level at cobalt target to that of core central region. The last factor, the flux level in core center, is related to the flux normalization that has already been mentioned in the above subsection, and not to be analyzed in detail here. The sensitivity coefficient is defined as:

$$G_{i,g} \equiv \frac{\sigma_{i,g}}{R} \frac{dR}{d\sigma_{i,g}}, \tag{3}$$

where the suffix i represents nuclide and reaction.

The calculated sensitivity coefficients are appearing in Figures 6 and 7.

a. Direct effect

Needless to say the error accompanying ^{59}Co capture cross-section directly influences the result. Comparing the major evaluated nuclear data libraries (JEF-2.2, ENDF/B-VI, JENDL-3.2, CENDL-2), about 1 or 2% uncertainty could be suspected in thermal energy region.

b. Spectrum effect

Hydrogen is the main nuclide that can characterize the neutron spectrum in this experiment. Since the energy group structure used in sensitivity analysis was too rough in thermal and epi-thermal energy region, the sensitivity coefficient for hydrogen elastic scattering in Figure 6 was underestimated below 100 eV. However the hydrogen scattering cross-section is fairly well-known, it can be said that no significant error would induced from this nuclear data. On the other hand, except for ^{59}Co self-absorption, there is little contribution from the other nuclides such as calcium and structural materials.

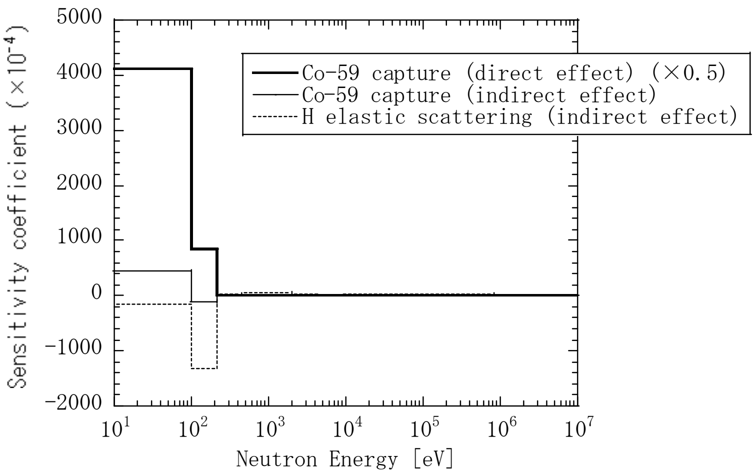

Figure 6. Sensitivity coefficients for ^{59}Co capture rate
(direct effect and indirect effect via neutron spectrum change)

c. Relative flux level at the target

Generally, the flux calculation is accompanied by large uncertainty at the third row of blanket region where the irradiation of cobalt target was performed. Main sensible nuclides and reactions are presented in Figure 7. Nuclides in blanket and shielding around the target seem to influence the neutron flux level in cobalt target, in which the effect of neutron scattering in fast energy region is remarkable. The sensitivities of ^{238}U and ^{239}Pu in lower energy region are exhibiting the effect of moderated neutron from cobalt target to the adjacent blanket fuels.

Next the uncertainty induced from nuclear data are evaluated using the calculated sensitivity coefficients and the covariance data for JENDL-3.2[5]. The covariance matrix element for the i-th nuclide and reaction $M_{ig,i'g'}$ are applied to estimate the standard deviation as:

$$\sqrt{\sum_{g,i',g'} G_{ig} M_{ig,i'g'} G_{i'g'}} . \qquad (4)$$

The results are listed in Table II. The dominant contributors are ^{238}U inelastic scattering (4.9%) and the Fe elastic scattering (3.4%). Totally 6 or 7 % of uncertainty on the flux level at the target are coming from the nuclear data.

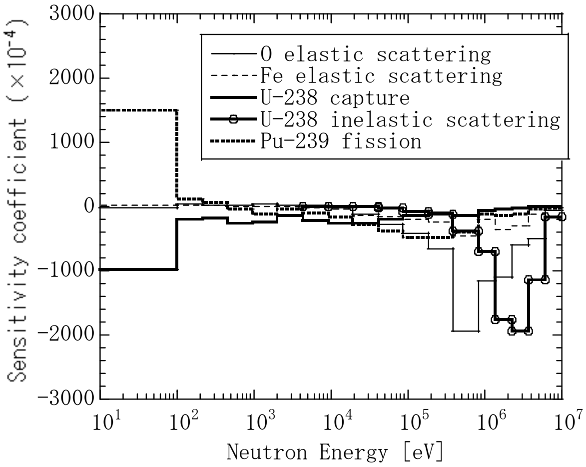

Figure 7. Sensitivity coefficients for ^{59}Co capture reaction rate (indirect effect on relative flux level at cobalt target)

Table II. Nuclear data uncertainty for ^{59}Co capture rate (indirect effect on relative flux level at cobalt target)

Nuclide and Reaction		Estimated uncertainty (1σ)
O	Elastic scattering	1.0%
Na	Elastic scattering	0.4%
Cr	Elastic scattering	1.0%
Fe	Elastic scattering	3.4%
	Inelastic scattering	1.5%
	$\bar{\mu}$	0.3%
^{235}U	Fission spectrum	0.5%
^{238}U	Capture	0.3%
	Inelastic scattering	4.9%
^{239}Pu	Fission	0.6%
	Fission spectrum	0.7%

In the above, the error factors were investigated from several points of view. Including the largest uncertainty on the neutron flux level at the target, about 10% of total error seems to exist in the present experimental analysis. However the cause of C/E dependence in ring and axial directions was not figured out clearly, which is remaining to be an issue in the future.

CONCLUSION

In order to find out the present calculation uncertainty for LLFP transmutation in a fast reactor, an analysis of the cobalt-60 production experiment in the PHENIX reactor was carried out using a continuous energy Monte Carlo code. The calculation gave an agreement with experiment in ±10%. According to the error estimation, about 10% of total uncertainty seems to exist in the present experimental analysis, where the largest uncertainty are coming from the scattering cross-section data of ^{238}U and Fe in fast energy region that can alter the neutron flux level at the cobalt target subassembly. A slight overestimation of the spatial shielding effect in the radial direction as well as the discrepancy of C/E values in the axial direction inside the target were observed, however their cause was not figured out and remains to be an issue in the future. The accuracy using the present nuclear data and Monte Carlo code is quite satisfactory for global prediction of the neutron behavior in a moderated target subassembly. The Monte Carlo method can be a reference for developing a calculation model using deterministic codes, meanwhile the residual discrepancies should be solved for more consistent prediction of LLFP transmutation.

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