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Experimental verification of the current data and methods for induced radioactivity and decay heat calculation in D–T fusion reactors

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Induced radioactivities and decay heat are of significant importance in the nuclear design of a near-term D–T fusion device from the view point of the safety consideration. In the framework of the JAERI/USDOE collaborative program on fusion neutronics, extensive experimental efforts have been devoted to verify the validity of the calculation code systems THIDA-2, REACT-2 and DKR-ICF. In the previous study, it was clearly pointed out that there were large discrepancies for several important materials between the experiment and the calculation in terms of γ-ray emission rates. This paper investigated the major sources of these large discrepancies. In addition to the previous ones, the analysis was carried out by THIDA-2 using an updated cross-section library. As a result, the following was pointed out: (1) The calculation of THIDA with the new activation cross-section library gave better agreement with experiment, especially for MnCu, W, Mo and V. As far as the higher neutron energy range above 1.0 MeV is concerned, all calculation code systems offer reasonable prediction accuracy. (2) For MnCu, W and Ta, uncertainty in the neutron spectrum was the main source for the large discrepancies because low-energy neutrons were very sensitive to the capture reaction products of ^{64}Cu, ^{187}W and ^{182}Ta.

1. Introduction

Induced radioactivity in fusion structural materials is of importance in terms of decay-heat, dose rate, and radioactive waste estimation. Extensive efforts have been addressed concerning the compilation of cross-section data and calculation code development. In order to arrive at the target accuracy for the parameters relevant to the activation, several experiments using many structural materials have been conducted in the framework of fusion neutronics studies [1–6]. They have investigated the adequacy of the cross-section data as well as the calculation codes to predict induced activities in the simulated D–T neutron environment at rather short time ranges from 10 min to several days after irradiation. In particular, the systematic experiments in the framework of the JAERI/USDOE collaborative program [4–6], have addressed serious problems: there were sever discrepancies among results obtained by different code systems, THIDA-2 [7], REAC-2 [8] and DKR-ICF [9], which are currently available and large deviations in the comparison of experiment and calculations were observed for materials like Mo, W, MnCu, Ta, V, etc. [5,6].

In this paper, we have investigated the sources of the discrepancies between calculation and experiment focusing on the adequacy of the activation cross-section data libraries. It was concluded that the main source of the uncertainty in the calculation arose mainly from inadequacy of the activation cross-section data. Also the uncertainty in the low-energy neutron flux calculation impacted on the accuracy of induced activity prediction.

2. Outline of experiments

An integral experiment of radioactivity and decay-heat was conducted at the FNS facility [10] in the framework of the JAERI/USDOE collaborative program on fusion neutronics during Phase-IIC [11] and Phase-IIIA [12]. The objectives of the experiment were to provide data for verifying radioactivity calculation codes, and to investigate the suitability of different

materials in meeting the selection criteria based on low activation and decay-heat considerations.

The Phase-IIC system consisted of an Li_2O breeder blanket with a first wall enclosed by 200 mm Li_2CO_3 with 50 mm polyethylene. A D-T point neutron source was located in the cavity of the enclosure, at 780 mm instance from the first wall of the Li_2O region. The Phase-IIIA system was featured by a new concept of a 2 m long pseudo line source with annular blanket assembly consisting of a 15 mm thick SS-304 first wall, and 400 mm thick Li_2O, 200 mm thick Li_2CO_3 and 50 mm polyethylene reflector zones. Detailed descriptions for both system were given in refs. [11] and [12].

As the first position (A) was close to the D-T neutron source in Phase-IIC, it is expected that the neutron spectrum simulates a typical one in the first wall region. The second position (B) provided simulation of a typical spectrum inside the tritium breeder blanket, Li_2O. With the line D-T neutron source configuration, the third position (C) was located at the center of the Phase-IIIA system at 5 mm depth in the Li_2O region. Neutron spectra at the three positions are shown in fig. 1.

The twenty materials used in the present study were Mg, Al, Si, Ti, V, Cr, MnCu alloy, Fe, Co, Ni, SS-316, Zn, Zr, Nb, Mo, Ag, Sn, Ta, W, and Pb. These samples were irradiated in the D-T neutron fields. After irradiation, decay γ-ray spectra were measured with Ge detectors. The gamma-ray emission rate in each sample material was deduced to be compared with the calculations. In table 1, the notations of the systems are tabulated along with the irradiation times and typical cooling times, which corresponded to the experimental conditions.

Table 1
Abbreviated notation of the cases to the experiments

Notation	System	Irradiation time	Cooling time
AS	Phase-IIC	30 m	30 m–1 hr
AL-1	Phase-IIC	10 hr	1hr–3 hr
AL-2	–	–	10 hr–3 d
BS	Phase-IIC	30 m	30 m–1 hr
BL-1	Phase-IIC	10 hr	1 hr–3 hr
BL-2	–	–	10 hr–3 d
C	Phase-IIIA	10 hr	1 hr–10 hr

3. Experimental analysis

The experimental analysis has been carried out by the currently available code systems THIDA-2, REAC-2 and DKR-ICF. Since the cross-section library of THIDA-2 has been recently updated, the experimental analysis has been carried out using the new version of library. The THIDA code system involves neutron flux calculation by DOT3.5 with the GICX40 [13] data library based on ENDF/B-VI and induced activity calculation by ACT4 with an activation cross-section

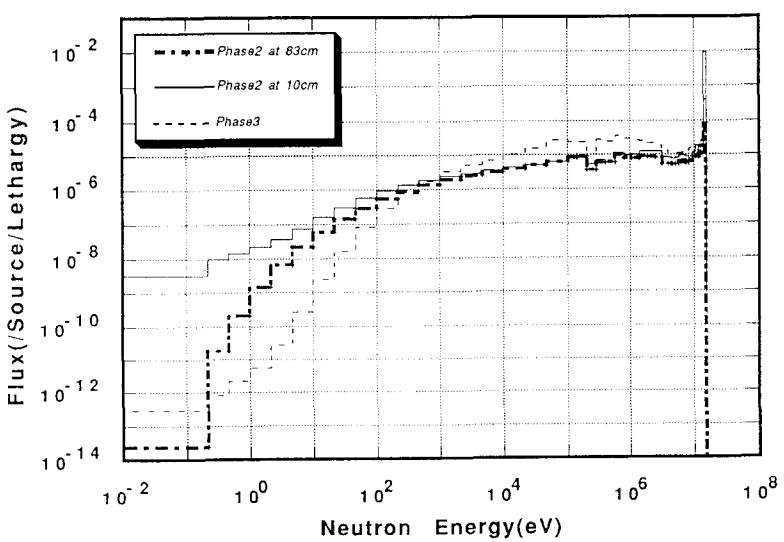

Fig. 1. Neutron spectra at position A, B and C calculated by DOT3.5 with GICX40.

library, CROSSLIB. In order to avoid uncertainties associated with the decay of activities during collecting time, THIDA computed the integrated γ-ray intensities per unit volume and an individual γ-ray energy spectrum during counting in order to avoid uncertainty in tracing all the decaying activities. The annihilation γ-ray is the case to be considered. The code systems REAC and DKR-ICF employed the MCNP for neutron transport calculation, using nuclear data libraries based on ENDF/B-V. Induced activities were generated using their own activation cross-section libraries. For REAC-2 and DKR-ICF, the ratio of computed to experimental value (C/E) was given for the integrated γ-ray emission rate per gram at the moment when γ-ray counting started.

From now, notations of THIDA-Old, THIDA-New, REAC and DKR-ICF indicate the induced activity calculations by using corresponding codes and libraries, respectively.

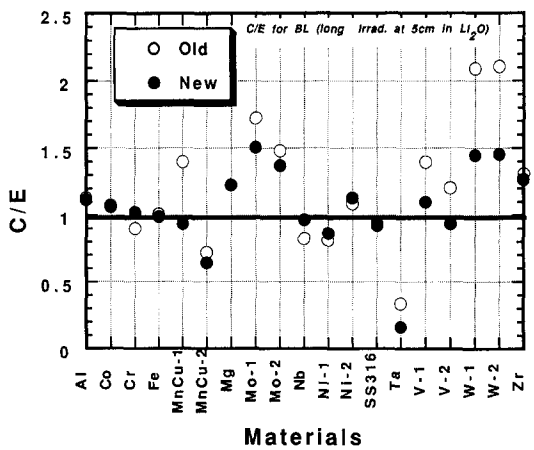

Fig. 2. The C/E values corresponding to both THIDA-Old and THIDA-New for the case of BL.

4. Discussion

4.1. Comparison of THIDA-New with THIDA-Old

In fig. 2, C/E values in the case of BL are plotted against the materials as a function of the cases. One of major topics is the improvement of the calculation accuracy in THIDA-New with respect to THIDA-Old. Figure 3 gives the direct change of the final γ-ray spectrum for W in the case AS. It was recognized that almost all γ-ray emission rates in THIDA-New were different from those in THIDA-Old.

Table 2
Experimental errors in $\pm$% for the integrated γ-ray intensities

Materials	Cases						
	AS	Al-1	AL-2	BS	BL-1	BL-2	C
Mg	–	3.2	3.2	–	3.3	3.8	–
Al	3.0	3.7	–	10	3.3	–	6.5
Si	3.5	–	–	–	–	–	–
V	2.9	4.4	4.4	8.2	5.2	6.1	–
Ti	3.2	3.7	3.8	16	4.9	–	6.4
Cr	–	3.3	3.4	–	6.1	–	–
MnCu	3.2	3.6	3.7	4.1	3.5	4.8	–
Fe	3.0	3.5	4.9	4.0	3.8	6.6	3.2
Co	3.1	3.0	3.7	4.2	3.7	4.0	–
Ni	3.4	3.3	3.6	12	3.8	5.0	6.7
Zn	–	–	–	–	–	–	5.3
Zr	3.3	–	–	10	3.7	–	5.9
Nb	–	3.7	3.7	–	3.3	3.8	4.1
Mo	3.9	4.0	3.6	6.6	3.3	3.9	5.2
Ag	–	–	–	–	–	–	4.7
Sn	–	–	–	–	–	–	5.9
Ta	–	–	–	–	–	3.1	7.8
W	3.8	3.8	4.5	4.8	3.4	4.1	5.1

Major sources for the error are due to γ-ray counting statistics, detector efficiency (2.5%) and neutron source determination (2.0%). Contributions from other uncertainties were relatively small, less than 1%.

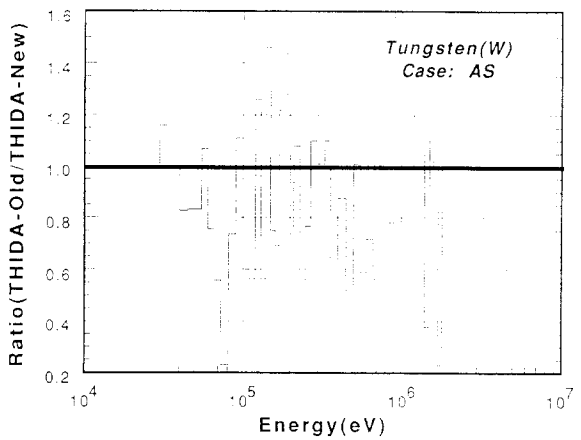

Fig. 3. The ratio of the γ-ray energy spectrum for W calculated by THIDA-Old to that by THIDA-New.

4.2. Discussion on each activity C/E

Here, we focused only on the major radioactivities in typical irradiation and γ-ray counting conditions because numbers of experimental cases were so lager to be covered. Much care was taken in the material cases which exhibited significant discrepancies among the calculations as well as experiments. Ranges of experimental errors are summarized in table 2 in order to make the discussion quantitative. Since almost all experimental errors were less than ±10% as show in table 2, it was anticipated that C/E values without errors gave a reasonably adequate basis for the discussion. In the following, material wise the studies are given.

4.2.1. Al

The activity of ^{24}Na, the product of the ^{27}Al (n, α)^{24}Na reaction, was dominant in all cases. The C/E values of all calculations were very close to 1.0 for all cases. This demonstrated the reasonable treatment in the analysis procedure of the neutron spectrum to the final γ-ray spectrum as long as the high energy part was concerned. This is a good indication for the monitoring of the system code performance because of less uncertainty in the activation cross-section and well known decay properties.

4.2.2. Mg

The C/E for magnesium looked good though there was a systematic overestimation of 18 to 37% for the case BL for all codes. This overestimation could be explained by the D–T neutron energy spectrum at the sample position and the overestimation of the cross-section of ^{24}Mg(n, p)^{24}Na in all libraries. The actual neutron peak energy at the sample position A and B were estimated 14.6 MeV from the D–T reaction kinematics. However, the cross-section was given as an averaged value for the 14 MeV region, resulting in the some overestimation. Recent experiments gave the cross-sections for this reaction to be 167 mb and 183 mb at 14.9 and 14.5 MeV, respectively, corresponding to the position of B and A. Substitution of this new cross-section value gives better agreement.

4.2.3. Si

Only one data set for the case of AS was available for silicon. The dominant activities were ^{27}Mg(50%) and ^{29}Si(50%). A very large discrepancy between calculations by THIDA-Old and -New was observed, where calculation with the New version gave much smaller values than that with Old. DKR-ICF and THIDA-Old showed reasonable agreement with experiment as shown in table 3. However, THIDA-New and REAC2 gave large underestimation and overestimation. From the investigation of the these particular reaction cross-sections given also in table 3, apparently, the cross-sections of THIDA-New are too small for both reactions. This leads to unreasonable underestimation. For the ^{29}Si(n, p)^{29}Al reaction cross-section at 14 MeV, however, values in both REAC2 and DKR-ICF seemed too high by more than a factor of two. Thus it should be noted that the agreement obtained using DKR-ICF could not be simply the evidence for the activation cross-section verification. The overestimation in REAC2 could be attributable to the existence of the other reactions which unreasonably contributed to the activity in silicon. The cross-sections in the THIDA2-New library should be corrected.

4.2.4. V

The new version of the cross-section in THIDA gave much improvement in the C/E ratios as shown in

Table 3
C/E values for silicon and cross-sections at 14.5 MeV for each library

	THIDA-Old	THIDA-New	REAC-2	DKR-ICF	FNS
C/E	0.81	0.39	2.8	1.08	–
Reaction	Cross-section (mb)				
^{30}Si(n, α)^{27}Mg	86	40	84	90	80
^{29}Si(n, p)^{29}Al	230	80	240	280	135

Table 4
C/E values for vanadium cases

Case	THIDA-Old	THIDA-New	REAC-2	DKR-ICF
AS	0.88	0.78	1.06	1.35
AL-1	1.59	1.22	1.57	3.4
BS	1.09	1.03	1.3	1.8
BL-2	1.40	1.10	1.4	3.1

table 4. This is mainly due to the improvement of the cross-section for ^{51}V(n, α)^{48}Ti being 16.2 mb which is close to the experimental data at FNS [14]: the old value at 14 MeV was 20% larger than the new one which is very consistent with the currently available experimental data at FNS. The overestimations in the REAC2 and DKR-ICF calculations by 40% and a factor of three, respectively, are also explained by too large cross-section values, 24.3 mb and 45.2 mb, respectively. It should be noted that there was no corresponding γ-ray peak to the activity of ^{47}Sc in the measurement though calculation presented prominent lines for all calculations. The cross-sections for this reaction were evaluated from reaction systematic or theoretical prediction because of lack of experimental data at 14 MeV. As far as the experimental evidence showed, the cross-section must be subjected to measurement.

4.2.5. Ti

Both THIDA calculations with the Old and New libraries gave identical results showing good agreement with experimental values. This is reflected by the identical cross-section values at 14 MeV for the main contributing reactions of ^{48}Ti(n, p)^{48}Sc and ^{47}Ti(n, p)^{47}Sc. However, REAC2 and DKR-ICF overestimated the experimental values in general by 15–86% although the cross-sections seem consistent with data in THIDA as well as the experimental value at FNS [14]. This unexplainable problem remaines to be solved in the near future. Only the case BS short cooling time less than 30 min submitted the problem of underestimation in THIDA, whereas REAC2 and DKR-ICF overestimated it by 73 and 62%, respectively. This trend can be attributed to the improper cross-section for ^{46}Ti(n, 2n)^{45}Ti. In this case, ^{45}Ti gave around 40% contribution to the total.

4.2.6. Cr

The results of THIDA-New and DKR-ICF were in good agreement with experiments. However, REAC2 showed a large overestimation by more than 50%. The improvement in C/E of the New library to that of the Old one was simply due to change in the cross-sections of ^{52}Cr(n, 2n) and ^{50}Cr(n, 2n) which produce major contributing activities of ^{51}Cr and ^{49}Cr, respectively.

4.2.7. Fe

The induced activities in iron were obviously dominated by ^{56}Mn at the short cooling time less than several hours. At one day after irradiation, ^{54}Mn becomes the only prominent contributor to the radioactivity. By the same reason for the aluminum case, C/E tends to be around 1.0.: the cross-sections for the major production reactions ^{56}Fe(n, p) and ^{54}Fe(n, p) have been well evaluated because of their importance in dosimetry application. The results for all cases are also positively supporting the validity of all code systems with the neutron transport calculations as far as the high neutron energy range above 1 MeV was concerned.

4.2.8. Co

Acceptable results in C/E values were found for cobalt. The improvement in C/E by using THIDA-New calculation indicated the properness of the cross-sections for ^{59}Co(n, α).

4.2.9. SS-316

There was generally good agreement between the experiments and all three calculations for all cases of SS-316, within 20%. As verified previously in the Li_2O assembly [1], the same level of confidence in the calculation had been provided by THIDA, resulting in excellent consistency to each other.

4.2.10. Ni

For nickel, there was general agreement between calculations and experiments for all cases. Only calculations by REAC2 and DKR-ICF for the cases of BS gave an unreasonable overestimation by a factor of two.

4.2.11. MnCu

The case MnCu suffered by very complicated trends for the C/E's as shown in fig. 4. The C/E's differ case by case (neutron spectrum, irradiation time and cooling time). Apparently, there was inconsistency among the three codes. In the case AS, THIDA with both Old and New libraries overestimated by 60% and REAC2 and DKR-ICF overestimated by more than a factor of three. This overestimation was dominantly (more than 90%) contributed by the 511 keV annihilation γ-ray

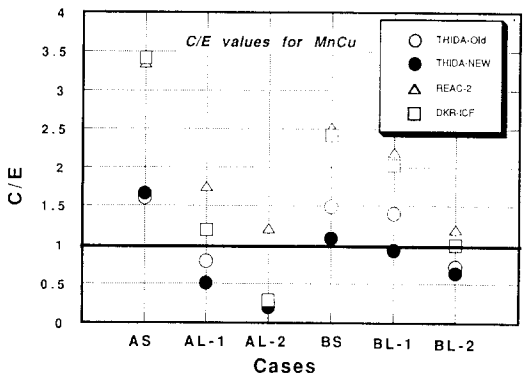

Fig. 4. The C/E values of MnCu alloy for all cases.

from ^{62}Cu, the product of ^{63}Cu(n, 2n)^{62}Cu. The cross-section values at 14 MeV for this reaction are 500, 539, 658 and 633 mb for THIDA-Old, THIDA-NEW, REAC2 and DKR-ICF, respectively. Though these values are slightly higher than the experimental value at FNS, it can not explain the overestimation. As one possible reason, the insufficient detector efficiency for this particular annihilation γ-ray which is distributed broadly due to the high energy β^+ emission around the MnCu samples. Still it is required to investigate the extremely large C/E's the for REAC and DKR.

A sudden drop in C/E was observed for the cases AL-1 and AL-2, where ^{56}Mn and ^{64}Cu dominated the γ-ray emissions. Although measurements gave 52 and 81% contributions by ^{64}Cu in AL-1 and AL-2, respectively, calculations of THIDA underestimated them by a magnitude of more than one order. Since the cross-section for ^{65}Cu(n, 2n)^{64}Cu has been well studied and data at 14 MeV in all libraries are identical, small C/E values were attributable to the underestimation in the ^{63}Cu(n, γ)^{64}Cu reaction, which is sensitive to the low-energy neutrons. In the case BS where ^{56}Mn gave 82% contribution, THIDA-NEW gave a reasonable C/E of 1.08. This result demonstrated the feasibility of the cross-section for ^{55}Mn(n, γ) for the neutron spectrum in Li_2O where the low-energy neutron flux was depressed due to the large resonance absorption at 250 keV by ^{6}Li(n, α). It may reduce the uncertainty in the low-energy neutron capture reaction of ^{55}Mn(n, γ). The C/E of 0.97 assigned by THIDA-NEW calculation in BL-1 where ^{56}Mn gave 90%, supported the adequacy in the prediction of ^{56}Mn. In the case of BL-2 at 3 days after irradiation, where the ^{64}Cu had 33% weight, C/E values tend to decrease. This is also explained by the underestimation in the ^{64}Cu in the calculation.

4.2.12. Zn

The 511 keV γ-line from ^{64}Cu, the product of ^{64}Zn(n, p)^{64}Cu, was the major contributor in the case C. THIDA-Old and -NEW tended to underestimate and overestimate the experiment by 10% and 30%, respectively. The cross-section values at 14 MeV for this reaction in the libraries of Old and NEW are 110 mb and 172 mb, respectively. The recent measurement at FNS gave 135 mb for this reaction cross-section [14]. These differences seemed to reflect the C/E trend.

4.2.13. Nb

One of the standard dosimetry reactions, ^{93}Nb(n, 2n)^{92m}Nb leaded the induced decay γ-rays. Current cross-sections around 14 MeV evaluated tend to be around 460 mb. All codes predicted reasonably the experiments in all cases. However, the C/E's for THIDA-Old systematically gave underestimation of the experiment by 10–15%. The reason was simply due to lack of the reaction of ^{93}Nb(n, α)^{90m}Y($T_{1/2}$ = 3.6 h) which contributed 15% of the total γ-ray intensity.

4.2.14. Mo

The C/E values are given in table 5. By careful checking of the γ-ray branching for the THIDA decay data, a serious mistake was found in the ^{99}Mo decay gamma branching: it gave 90% branching for the 141 keV from ^{99}Mo though that value should be negligibly small. This was the main source for the overestimations for the cases of AL, BL and C, where ^{99}Mo and ^{99m}Tc dominated the γ-ray intensities. For the cases at short cooling time, agreement seemed good between THIDA-New and experiments. However, the 54% 511 γ-ray from ^{91}Mo, product of ^{92}Mo(n, 2n), in the measurement in these cases was underestimated by a factor of two. Thus, it can be said that the agreements were the results of compensation due to the underestimation and overestimation. It is apparent that REAC2 has some trouble in the library which includes unnecessarily large cross-sections giving the main contribution.

Table 5
C/E values for molybdenum cases

Case	THIDA-Old	THIDA-New	REAC-2	DKR-ICF
AS	1.23	1.03	7.0	2.6
AL-2	1.5	1.35	2.1	1.6
BS	1.13	1.08	3.5	1.9
BL-2	1.5	1.37	3.6	1.4
C	1.7	1.5	4.3	1.6

4.2.15. Zr

The THIDA calculation tended to overestimate the measurements by 20–30%. This overestimation arose in the 909 kev γ-lines prediction. This may cause the inadequate decay chain treatment for ^{89m}Y, daughter nuclide of ^{89}Sr which is produced by the reaction of ^{92}Zr(n, α). THIDA calculation gave a comparable intensity of the 909 keV γ-line from ^{89m}Y. However, such a large contribution from ^{89m}Y is not realistic, because the half-life of ^{89}Sr is 53 d and the cross-section of ^{92}Zr(n, α) should be small, around 10 mb, in comparison with the cross-section of ^{90}Zr(n, 2n)^{89}Zr. Subtraction of the contribution from ^{89m}Y in the calculation gives excellent improvement in C/E.

4.2.16. Sn

Gamma-ray energy spectrum analysis gave no clear correspondence between calculation and measurement. This is due to miss assignment of the reaction of ^{116}Sn(n, p)^{116m}In in the THIDA-New library: THIDA dropped the correspondence to the product of ^{116m}In of 54.2 m, which was the major contributor in the measured γ-ray spectrum. On the other hand, calculation gave a very high intensity for the 158 keV line emitted by ^{111m}In, the product of ^{112}Sn(n, np). The THIDA calculation gave a large cross-section of 36.8 mb for this reaction, a value which seems too high from the reaction cross-section systematic.

4.2.17. Ag

For the Ag in case C, there was reasonable agreement between calculation and experiments as shown in fig. 5. REAC2 did not include the corresponding cross-section or decay γ-ray source.

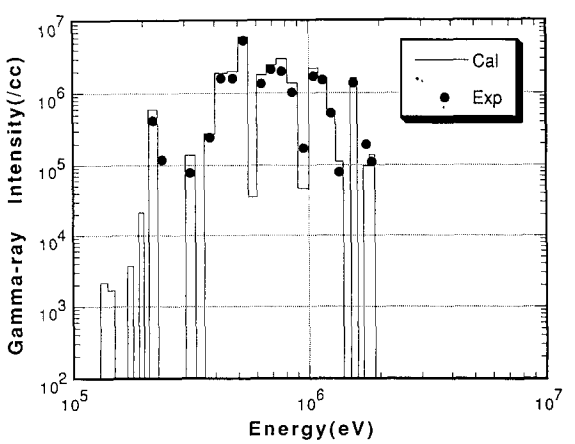

Fig. 5. Comparison of gamma-ray energy spectrum for Ag calculated by THIDA-New with the experiment.

Table 6
C/E values for Tungsten cases

Case	THIDA-Old	THIDA-New	REAC-2	DKR-ICF
AS	1.5	0.97	307	0.2
AL-2	1.6	0.91	2.6	0.03
BS	1.5	1.05	14	0.01
BL-2	2.1	1.45	2.6	0.002
C	1.12	0.97	1.3	0.005

4.2.18. W

In table 6, the C/E values are shown. There were no corresponding decay data of prominent activities of ^{187}W, ^{186}Ta and ^{183}Hf in DKR-ICF resulting in very small C/E values for all cases. On the contrary, REAC2 showed extremely high C/E's for cases with short cooling time. This was mainly due to an improperly large cross-section for ^{180}W(n, 2n)^{179}W in the library. THIDA-New presented significant improvement in the C/E ratios for all cases with respect to THIDA-Old. This decrease in C/E is attributed to the lowered cross-section value of the ^{186}W(n, γ) resonance capture at 20 eV in THIDA-New. However, the branchings for 239 and 114 keV γ-rays from ^{187}W, which are dominant contributors in the calculation, were unreasonably larger by more than a factor of 100 in the THIDA decay library. After the correction for the incorrect γ-ray branching ratio, C/E became around 0.75 for all cases. Thus it could concluded that the products by the (n, γ) reaction tended to be underestimated as shown for ^{64}Cu in MnCu cases.

4.2.19. Pb

Leading activity of ^{203}Pb was produced by the reaction of ^{204}Pb(n, 2n), though the abundance of ^{204}Pb is small, 1.4%. THIDA-NEW and DKR-ICF gave reasonable C/E's. An overestimation in REAC2 may caused by the duplication of the cross-sections for ^{204}Pb(n, 2n)^{204g}Pb and ^{204}Pb(n, 2n)^{204m}Pb.

4.2.20. Ta

Contrary to the W cases, very small C/E's in THIDA were found. This was partly due to missing branchings for 41.3, 100.1, 152.4 and 222.1 keV γ-rays from ^{182}Ta which should have 74% contribution to the total. Although correction for these data improves the C/E's, still the calculation underestimated the experiment by about 50%. This is also attributable to the uncertainty in the capture reaction calculation process as seen in cases of ^{63}Cu(n, γ) and ^{186}W(n, γ).

Finally, referring to $^{197}Au(n, \gamma)^{198}Au$ the reaction rate was systematically underestimated by 40–50% in the Phase-IIC cavity through the analysis by DOT3.5 and MORSE-DD. It was concluded that the low-energy neutron spectra in the Phase-IIA and IIB should be softer than those from the calculations [15]. Thus, the underestimation in the codes for some products by (n, γ) reactions is attributable for the neutron transport insufficiencies.

5. Summary and conclusion

The adequacy of the code systems used was assured by the integral test on the reaction products for ^{24}Na, ^{56}Mn, ^{92m}Nb, the cross-section of which were assumed well evaluated with sufficient accuracy. Also, the reasonable C/E values indicated that the neutron energy spectrum above 1 MeV could be reasonable for each code.

We have encountered serious improper data bases associated with the decay γ-ray branching ratios. This may be simply because of unexpected human error in making such a large data library. But it would be very important because many design calculations have been carried out giving the criticality of specific items of concern without noticing the incorrectness of the data. In this context, emphasis should be placed on the importance of integral experiments for verifying codes and data.

The experimental analyses for induced radioactivities in various spectra indicated inadequacy for considerable large numbers of the activation cross-section data in the currently available libraries. The present study has clearly pointed out the specific reactions which gave rather poor C/E's. Thus, we could reach a reasonable solution or acceptable levels of the adequacy in the data base as long as the threshold type reactions were taking into account. One serious problem associated with inadequate prediction in the products of (n, γ) reactions, however, is still left to be verified.

Acknowledgement

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