7.3 Influence of Impurities in Beryllium on Tritium Breeding Ratio

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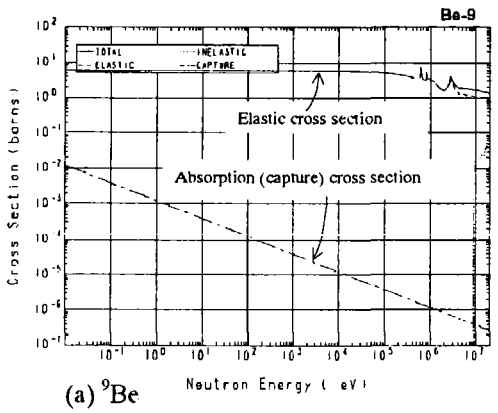
Several neutronics experiments simulating fusion blankets have been conducted with 14 MeV neutron source to assess the reliability of nuclear analysis codes. However, the analyses have not always presented good agreements so far between calculated and measured tritium production rates. One of thereasons was considered as impurities in beryllium which has negligibly small neutron absorption cross section in low energy range. Chemical compositions of beryllium were analyzed by Inductively Coupled Plasma (ICP) method, and a pulsed neutron decay experiment discovered that the macroscopic neutron absorption cross section for beryllium medium may be about 30% larger than the value calculated by the data specified by manufacturing company. The influence of the impurities on the calculations was studied on the basis of the fusion DEMO-reactor blanket design. As a result of the study, it was made clear that the impurities affect the local tritium production rates when the size of beryllium medium is more than 20-30 mean free paths (30-40 cm) in thickness. In case of some blanket designs that meet the above condition, the effect on tritium breeding ratio may become as large as about 4%.

1. INTRODUCTION

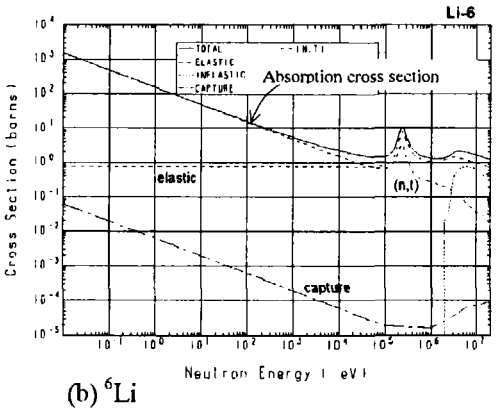
In fusion reactors based on the D-T reactions, the tritium breeding ratio (TBR) produced in blankets must be large enough to compensate the consumed amount of tritium in the burning plasma to sustain nuclear fusion reactions, and the uncertainty of TBR must be as small as possible so that the blankets are efficiently designed. A fusion blanket consists of structural material, tritium breeder, coolant and neutron multiplier. Beryllium is used for neutron multiplication in the blanket. Several neutronics experiments have been performed [1-7] at FNS (Fusion Neutronics Source) of JAERI to evaluate the reliability of nuclear analysis codes and nuclear data to be used for the blanket design. The facility produces 14 MeV neutrons through deuteron and tritium (D-T) nuclear reactions. In the results of integral experiments and analyses for the assembly consisting of beryllium layers, low activation ferritic steel F82H and lithium titanate (Li_2TiO_3) ceramics, the tritium production rates calculated with the Monte Carlo method were about 20 % larger than the measured values [1-7]. One of the causes of the differences was presumed to be impurities in beryllium. A chemical analysis based on the Inductively Coupled Plasma (ICP) method was employed to identify the elements and estimate the concentrations of the impurities. In addition, a pulsed neutron decay experiment was conducted to measure the macroscopic absorption cross section for the beryllium medium containing impurities. As a result of the study, some of the minor impurity elements turned out to have large contribution to the cross section. The influence of the impurities on the tritium production rate was analyzed for the fusion DEMO-reactor blanket design [8] by three-dimensional neutron transport calculations [9]. The analysis provided the condition in which the local tritium production rates were affected by the impurities and the knowledge how much the total tritium breeding ratio may be affected.

2. NEUTRONICS PROPERTY OF BERYLLIUM IMPURITIES

Beryllium is a material used in the design of fusion reactors with high priority. The characteristics are expected to meet the performances of a neutron multiplier in the tritium-breeding blanket. Neutron multipliers are required to compensate the obvious neutron losses in the blanket to attain the net tritium production enough large for a self-sustaining fusion reactor. In addition, beryllium is an excellent neutron moderator with an extremely low thermal neutron absorption cross section, as it is shown in Figure 1 (a) [10]. On the other hand, lithium-6 has an very large absorption cross section mainly represented by $^{6}Li(n,\alpha)^{3}H$ reaction as it is shown in Figure 1 (b) [10]. The reaction is used for tritium production, however, when lithium-6 exists in beryllium it becomes a very influential impurity as well as ^{10}B having large possible effect on neutronics calculation. The effect decreases the number of thermal neutrons, which will result in the lower tritium production rates.



(a) ^{9}Be



(b) ^{6}Li

Figure 1. Neutron cross section of ^{9}Be and ^{6}Li.

2.1. Chemical composition of structural beryllium grades

The impurity concentration depends on the purity grade of the beryllium. Manufacturer information concerning the element composition of several grades for commercially available beryllium is shown in Table 1. Structural grades S-65C, S-200F and S-200E (Brush Wellman Inc.) are being considered for use in the ITER [11]. In typical specifications, major impurity elements are characterized by a high concentration level of about 1000 ppm. Minor impurities are commonly not specified in detail. Instead, the total impurity level, of about 400 ppm, is shown in the lowest row of Table 1.

Table 1
Specifications of chemical composition for structural beryllium grades.

Chemical composition	S-65C	S-200F	S-200E
Be, min %	99.0	98.5	98.0
BeO, max %	1.0	1.5	2.0
Al, max ppm	600	1000	1600
C, max ppm	1000	1500	1500
Fe, max ppm	800	1300	1800
Mg, max ppm	600	800	800
Si, max ppm	600	600	800
Others, max ppm	400	400	400

Very limited information concerning the composition of minor impurities is available. Various studies [12-14] have shown that impurities can contain elements such as: the Li, B, Cl, Cr, Mn, Co, Cd, Dy, Th, U and others. According to the study [12] conducted with the use of neutron activation and gamma spectrometry, a uranium concentration as large as about 20 ppm was found in relatively pure beryllium.

2.2. Impurity effect on tritium production

Some of the elements remaining in beryllium even after the purification process tend to have a significant effect on the absorption cross section of thermal neutrons, in spite of a low impurity level, because of the large absorption cross sections of the

elements. Significances of the effect for representative elements were compared defining the value R, that is, the impure-beryllium to pure-beryllium ratio of the total macroscopic absorption cross sections. Results for impurities having a high absorption cross section are presented in Table 2.

Table 2
The effect of minor impurities on the ratio of macroscopic absorption cross section of impure to pure beryllium.

Composition			R
Be, %	Supposed impurity		
	Element	Level,ppm	
100	---	0	1.00
99.99	B	5	1.43
99.99	Gd	5	2.78
99.99	Cd	5	1.15
99.99	Li	5	1.06
99.99	Cl	100	1.11
99.99	Co	100	1.08
99.99	Mn	100	1.03
99.99	All the above	320	3.63

As indicated in this table, even a small amount of impurities, less than 10 ppm, can remarkably increase the total absorption cross section in beryllium.

As an example of influence of the parasitic absorption in the beryllium moderator on tritium production, an estimation of the tritium production rate on lithium-6, TPR-6, was performed for the beryllium assembly, irradiated by D-T neutrons. The parasitic absorption was simulated by boron with a level of 5 ppm, and a concentration corresponding to a 40% increase of the absorption cross section. The ratio of TPR-6 in the impure beryllium to pure beryllium is presented in Figure 2. The parasitic absorption decreases the TPR-6 by about 5% on the surface of assembly. The decrease of the TPR-6 becomes more prominent as the location grows deeper, due to the moderation and thermalization of neutrons, as a result of the increasing absorption cross section, which follows the 1/v-law. Nevertheless, the impurity effect depends on the blanket design, volumetric ratio of lithium and beryllium, and enrichment of lithium.

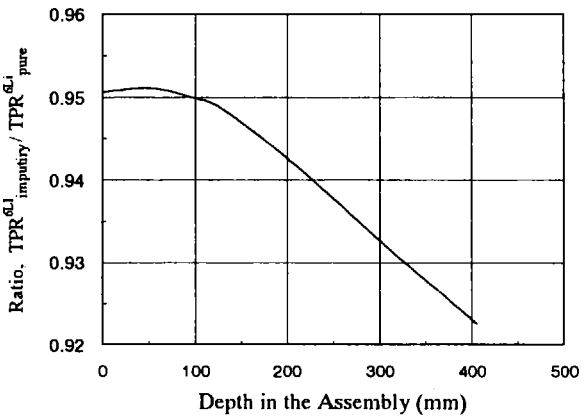


Figure 2. Effect of impurity as a function of depth in beryllium assembly.

3. IMPURITY ANALYSES

Since impurities have large effect on the macroscopic neutron absorption cross section of the beryllium material, the concentrations of impurities in the beryllium medium that had been used in the FNS experiment were investigated with Inductively Coupled Plasma (ICP) Spectrometry and the macroscopic cross section was estimated with neutronics experiment. The methods and the results are described in this section.

3.1. Inductively Coupled Plasma method (ICP)

Inductively Coupled Plasma-Atomic Emission Spectrometry (ICP-AES) is an optical analysis method useful to know small amounts of impurities. Quantitatively analyzed by the method were such elements as Mg(110ppm), Al(570ppm), Mn(96ppm), Fe(1300ppm) and Ni(250ppm). On the other hand, Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) is more powerful method that enables to analyze 2-3 orders of magnitude smaller bits of impurities than ICP-AES. The method identified B(<3ppm), Cd(<1ppm), Li(<1ppm), Th(1.5ppm) and U(82ppm). Results of the analysis are summarized in Table 3. Some of the elements were detected, though their quantities were not measured absolutely. Those are indicated by symbol "S".

Table 3
Chemical composition of the S-200F standard grade beryllium measured with the ICP-MS method.

Element	Content	Element	Content
Be, %	97.9±0.8	Ni, ppm	250±30
Li, ppm	< 1	Zr	S
B, ppm	< 3	Nb	S
Mg, ppm	110±5	Mo	S
Al, ppm	570±50	Cd, ppm	< 1
Sc	S	Dy	S
Ti	S	Ta	S
V	S	W	S
Cr	S	Hg	S
Mn, ppm	96±5	Pb	S
Fe, ppm	1300±70	Th, ppm	1.5±0.1
Co	S	U, ppm	82±3

Results obtained from the spectroscopic study can introduce some uncertainties to calculations of the tritium production rate due to the sensitivity level of the technique, therefore not being able to meet the benchmark requirements for the integral experiment. Besides, additional uncertainties can exist as well, due to the non-uniform impurity distribution, since only one sample of the beryllium material was analyzed. In such circumstances, for qualification and validation purposes, measurements of the integral effect seem very useful.

3.2. Pulsed Neutron Decay Experiment

The impurity effect results in smaller tritium production rates mainly owing to the $^{6}Li(n,\alpha)$ reactions. Therefore, it is important to estimate the impurities that reduce the thermal neutron flux, that is, estimate the macroscopic thermal neutron absorption cross section for beryllium medium. The pulsed neutron technique [15], which had been developed for precise determination of the neutron diffusion parameters in the moderator media, was adopted as the most effective measure to estimate it.

The overall setup of the experimental equipment is shown in Figure 3. It consists of a pulsed D-T neutron source (FNS facility in JAERI), a beryllium assembly ranging from 30 to 60 cm in size, a BF_3 neutron detector, and a time analyzer. The 80-degree beam line in the first target room of the FNS facility was used for the present experiment. The room provided good experimental conditions to minimize the neutron background due to the large size of the room (15×15×12 m) and the grating floor.

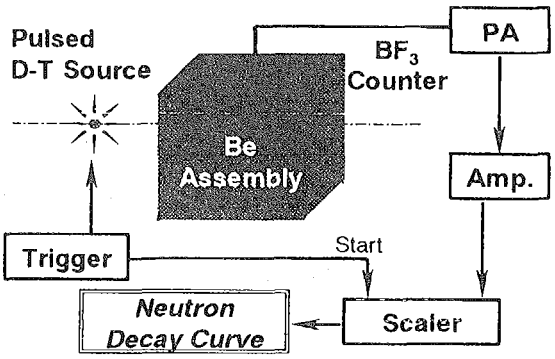


Figure 3. Pulsed neutron decay experiment system.

The pulsed neutron technique consists of time behavior observations of the neutron density, following the injection of a burst of neutrons into the moderator. When a bunch of fast neutrons is fed into a lump of beryllium, energy degradation and neutron thermalization are caused. The slowing-down time to thermal velocities in beryllium is about 100 μsec. When the neutrons approach thermal equilibrium with the moderator, they continue to leak slowly out of the assembly or may be absorbed. The thermal neutron flux decreases exponentially, and the impurities affect the decay constant. The decay rate of the neutron flux was measured with BF_3 neutron detector for twelve different sizes of Be assemblies. The decay constants are expressed by a parabolic function of the buckling factors. When the data for beryllium assemblies of various sizes have been obtained, the decay constant for infinite beryllium medium, that is a product of neutron velocity and macroscopic cross section, can be obtained.

The effective absorption cross section of structural beryllium (S-200F) for thermal neutrons was approximately 30% higher than that calculated on the basis of the data specified by the manufacturing company. The value corresponds to 2.9×10^{17} ^{6}Li atoms/cm^3 (2.6ppm), if the impurity was

supposed to be ^{6}Li only. For the pure beryllium assembly, such effect will decrease the tritium production rate by at least 5 %. In the blanket of the fusion reactor, the effect will depend on the blanket design, volumetric ratio of lithium and beryllium, and enrichment of lithium.

4. INFLUENCE OF INPURITIES ON FUSION BLANKET DESIGN

In the preceding sections, it has been confirmed that impurities in beryllium medium have some influences on the macroscopic neutron absorption cross section of the medium to be used in neutronics calculations. A tritium production rate (TPR) is specified as a reaction rate producing tritium per lithium nucleus and a 14-MeV-neutron generated by a D-T reaction. On the hand, a tritium breeding ratio (TBR) is obtained by integrating TPR for all of lithium nuclei over the breeding blankets in a reactor. In the present work, TPR is calculated only by $^{6}Li(n,\alpha)^{3}H$ reaction, because it usually produces more than 99% of total TPR and suffers the influence of impurities. In order to maximize TBR, some of the blanket designs consist of homogeneous material of neutron- multiplying beryllium and tritium breeding lithium. Another design may be composed of relatively broad beryllium zone and lithium area. Here in this section, the conditions in which the influence of impurities becomes large were studied.

4.1 Calculation model and method

The design of fusion DEMO-reactor [8] was employed to study the influence of impurities. The calculation model is shown in Figure 4. Though the configuration of the reactor is torus, the structure from the plasma centerline was cylindrically modeled to simplify the matter. Inner surface of the first wall is 160 cm in radius, and the outer surface of the blanket back plate is 235 cm from the plasma centerline. The vacuum vessel is 1 cm apart from the back plate and 24 cm in thickness. Breeding material is lithium titanate (Li_2TiO_3). Although ^{6}Li-enriched lithium in 90% is supposed in the DEMO-reactor design, natural lithium was employed in the present study. The blanket was radially divided into seven layers ranging from 5.5 cm to 19 cm in thickness, and in each layer, a breeding region 0.7 cm to 7 cm in thickness was provided. In the present analysis, the water region simulating coolant was removed and replaced by beryllium. In the design of fusion DEMO-reactor, the multiplier is supposed to be pure beryllium.

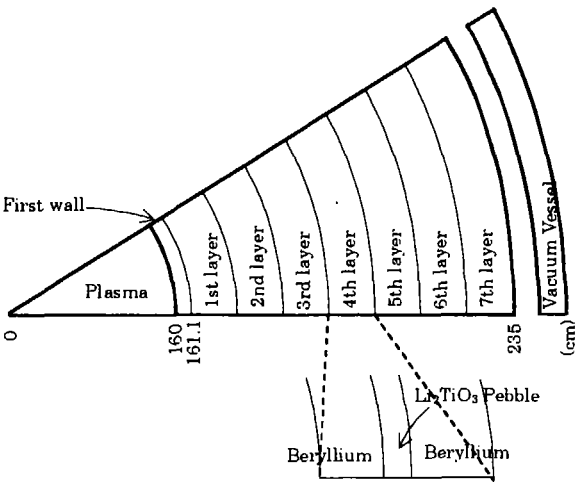


Figure 4. Calculation geometry of fusion DEMO-reactor blanket.

For the study of the impurities influence, the region between 161.1 cm and 235 cm behind th e first wall was surveyed for the following cases;

Case 1: Beryllium only through 7 layers,
Case 2: Lithium titanate only for the 1st layer with beryllium for the other layers,
Case 3: Lithium titanate for the 1st, 3rd and 7th layer with beryllium for the other layers,
Case 4: Lithium titanate in the breeding regions for 7 layers filling beryllium in the remaining area of the blanket.

The values of TPRs were calculated in breeding regions of 7 layers even though some of them might be filled with beryllium according to the calculation case. For each case of material condition in the blanket region, two calculations were conducted. One is done with pure beryllium, and the other is with beryllium containing impurities. The data obtained by ICP spectrometry were employed as the impurity concentrations. Although the geometry was two-dimensional, calculations were performed by a three-dimensional Monte Carlo code MCNP-4C [9] with continuous energy nuclear data JENDL-3.2 [10].

4.2 Feature of TPR distribution

The ^{6}Li TPR distributions in the blanket region are shown in Figure 5. The ordinate is 10^{-24} times micro- scopic reaction rates per unit D-T reaction, and the abscissa is the distance in cm from the plasma centerline.

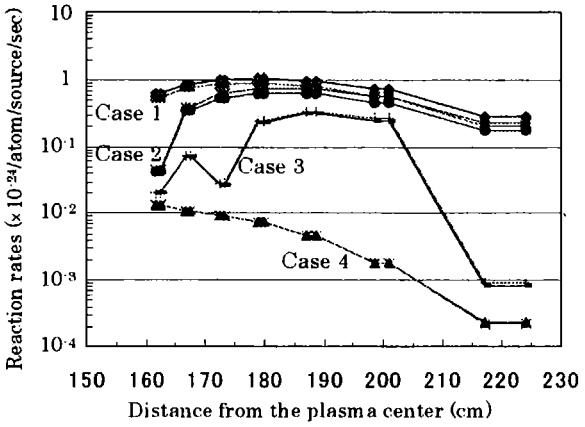


Figure 5. TPR distributions in fusion DEMO-reactor blanket.

In case 1 in which 7 layers are fully filled by beryllium only, the TPRs are nearly as large as 1×10^{-24}. In the other cases when lithium titanate exists only in some of the 7 layers, the TPR in the layer of lithium titanate becomes about 2 orders of magnitude smaller than those in case 1. As for the layers where beryllium region exists, the value of TPR depends on the distance to the lithium titanate layer. When the location is far from the lithium titanate layers, TPR becomes as large as that in case 1. However, when it is close to a lithium titanate layer, TPR is relatively small. The feature is observed in cases 2 through 4. In case 2, TPRs in 4th through 7th layers far from the lithium-filled 1st layer are roughly as large as those in case 1. In case 3, although TPRs in 4th through 6th layers are large, the value in the 2nd layer lying between lithium titanate layers is relatively small. In case 4, TPRs are generally small because all 7 layers were filled with lithium titanate.

4.3 Effect of impurities on TPR

When impurities existed in beryllium material, TPRs becomes smaller than those obtained with pure beryllium. The ratios of TPRs calculated with impurities-containing beryllium to those with pure beryllium are shown in Figure 6 as a function of the distance from the plasma centerline.

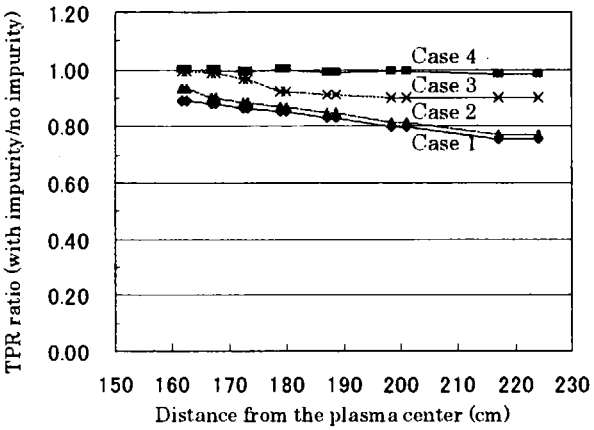


Figure 6. Effect of impurities on fusion DEMO-reactor blanket.

When 7 layers are filled with lithium titanate, the difference of TPRs caused by impurities is within 2 %, whereas, the difference becomes as large as 10-25 % when beryllium only exists through whole region. In the other cases when lithium titanate layers are sparsely provided, the difference grows larger as the location becomes farther from the nearest lithium titanate layer. This feature can be explained by the understanding that the impurities have an influence mainly upon low-energy neutron behavior. When the space is large enough for neutrons to moderate down to thermal or epithermal neutrons, absorption of neutrons by impurity becomes noticeable. On the other hand, when the space is not so large, not a few neutrons are absorbed by ^{6}Li constituting lithium titanate on the way of slowing-down.

The scattering cross section of beryllium is almost constant at about 6 barns below 10 keV. The mean free path (mfp: the inverse of macroscopic cross section) of neutron scattering in beryllium is about 1.3 cm. According to the result in Figure 6, the influence of impurities becomes prominent when the size of beryllium grows larger than 20-30 mfps (30-40 cm in thickness). Some consideration is recommended in case of the design for low energy neutrons. The example may be the design of thermal neutron flux monitors to be installed somewhere near beryllium region.

As it was shown in Figure 1 (b), neutron absorption cross section of ^{6}Li is very large. The feature observed in Figure 6 is due to the extremely large absorption cross section of ^{6}Li. Neutron spectra in the

lithium region of the 1st and the beryllium region of the 5th layers of the case 3 are compared in Figure 7. It is clearly shown that neutrons less than 1 eV are very few in the 1st layer comprising ^{6}Li.

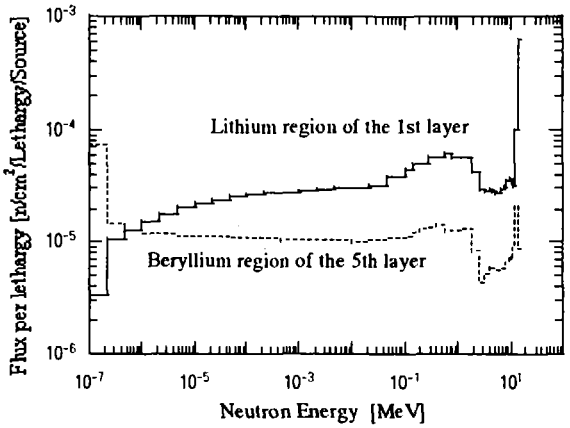


Figure 7. Neutron spectra in fusion DEMO-reactor blanket.

4.4 Effect of impurities on TBR

Tritium breeding ratio (TBR) is calculated by integrating macroscopic TPR (product of microscopic TPR and atomic number density of Li) all over the breeding region. The TBRs calculated with pure beryllium neutron multiplier in 4 cases are summarized in Table 4. Water coolant was neglected in the calculations.

Table 4
Summary of ^{6}Li TBRs with pure beryllium neutron multiplier.

Case \ Layer	1st	2nd	3rd	4th-6th	7th	Sum
1	-	-	-	-	-	-
2	1.17	-	-	-	-	1.17
3	0.53	-	0.98	-	0.31	1.82
4	0.34	0.33	0.33	0.83	0.08	1.90

In case 1, TBRs are null for all layers as lithium does not exist. Since lithium was filled only for the 1st layer in case 2, total TBR is not very large for the case. However, the total TBR is considerably large in case 3 even though lithium was sparsely provided only for three layers. The total TBR is nearly as large as that in case 4 when lithium was filled in all 7 layers. The choice which of case 3 or case 4 is proper depends on what is required in the design. The influence of impurities on TBRs in the cases are shown in Table 5.

Table 5
Influence of impurities in beryllium upon ^{6}Li TBRs.

Case \ Layer	1st	2nd	3rd	4th-6th	7th	Sum
1	-	-	-	-	-	-
2	0.93	-	-	-	-	0.93
3	0.99	-	0.96	-	0.90	0.96
4	1.00	1.00	0.99	0.99	0.98	0.99

When lithium is filled only in the 1st layer, the influence of impurities reaches 7 %. As the number of lithium-filled layers grows larger, the influence becomes smaller. It is meaningful that the influence differs by 3 % between case 3 and case 4, although TBRs for the cases were nearly the same amount.

5. CONCLUSION

Beryllium has an extremely small neutron absorption cross section in low energy range. Accordingly, when some impurities exist, the macroscopic absorption cross section of beryllium material may become considerably larger than that of pure beryllium. The amount of impurities was analyzed by Inductively Coupled Plasma (ICP) spectrometry, and minor elements in beryllium blocks used for integral experiments in JAERI were identified. The result of ICP spectrometry was well consistent with the data obtained by pulsed neutron decay experiment. The effective thermal neutron absorption cross section of the grade S-200F structural beryllium was approximately 30% higher than that calculated on the basis of the impurities data provided by the manufacture. The quantity of the impurities explains about 5 % of the difference between measured TPRs and calculated values in the clean beryllium experiment, however, the influence on TPRs in the integral experiments for lithium titanate and beryllium assembly may be smaller. As for the influence upon design calculations, local TPR may be miscalculated by more than 20 % at maximum after the deep neutron transportation through beryllium layer. Low energy neutron flux needs to be carefully treated in case that

beryllium layer is more than 20-30 mfps in thickness. The TBR, integrated TPR over whole breeding region, is not seriously affected by the impurities so long as the breeding material and beryllium are mixed as small lumps, however, some effect needs to be examined when they are used separately in large volume. Independently of the impurity problem, the cross section of beryllium itself seems to have unreliability. More reliable nuclear data, especially of (n,2n) reaction for beryllium, is of a great concern for neutronics designers of fusion reactors.

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