Integral Activation Experiment of Fusion Reactor Materials with d-Li Neutrons Up to 55 MeV

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An integral activation experiment of fusion reactor materials with a deuteron-lithium neutron source was performed. Since the maximum energy of neutrons produced was 55 MeV, the experiment with associated analysis was one of the first attempts for extending the energy range beyond 20 MeV. The following keywords represent the present study: d-Li neutrons, 55 MeV, dosimetry, SAND-II, spectrum adjustment, LA-150, MCNP, McDeLi, IFMIF, fusion reactor materials, integral activation experiment, low-activation, F82H, vanadium-alloy, IEAF, ALARA, and sequential charged particle reaction.

1. Introduction

An integral activation experiment of fusion reactor materials with a deuteron-lithium (d-Li) neutron source was performed at Forschungszentrum Karlsruhe (FZK), Germany, for the sub-task “Fusion Neutronics” under the IEA Collaboration [1, 2]. Since the maximum energy of neutrons produced was 55 MeV, the experiment with associated analysis was one of the first attempts for extending the energy range beyond 20 MeV that has been the upper limit energy for fusion applications. Accordingly, several codes and data developed recently for higher energy applications were used for practical analysis, and several important experimental facts were found. This report summarizes the experiment focusing on important experimental findings and code & data newly introduced.

2. Neutron Field Characterization

2.1. Experiment

A lithium target shown in Fig. 1 was used for the experiment. Metallic lithium was contained in a stainless

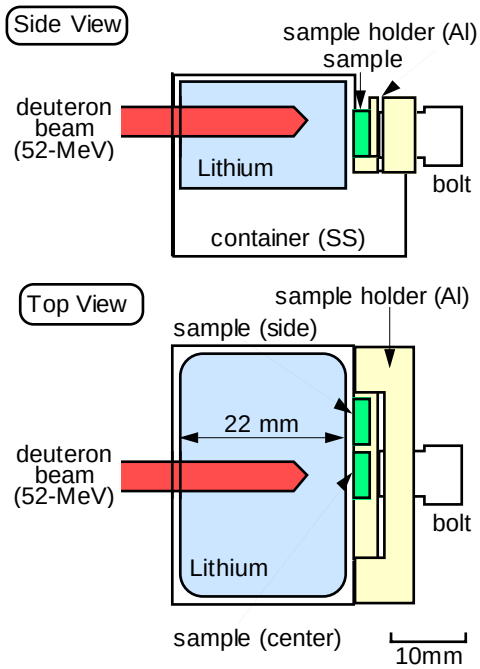

Fig. 1 The lithium-target and sample arrangement.

steel case. A deuteron beam of 52 MeV of typically 3 μA was impinged into the target to produced neutrons. An approximate energy of deuterons entering into the lithium regionwas 40 MeV due to energy loss in the stainless steel wall of 1 mm. The lithium thickness of 22 mm was enough to stop the deuteron beam. Samples were placed at the center and side positions on the downstream side surface of the target. The d-Li neutron source was a simulation of a neutron environment for the International Fusion Materials Irradiation Facility (IFMIF). Before the integral activation experiment, a neutron field characterization was performed by means of multiple-foil activation technique to define neutron flux spectra at the sample positions precisely.

Twenty-seven threshold reactions listed in Table 1 were selected as activation detectors. In addition to the low threshold reactions which have been used for fusion neutronics studies, such as the (n,n'), (n,p), (n,α) and (n,2n) reactions, the (n,3n) ~ (n,6n) reactions having high threshold energies were selected to characterize the high energy neutron flux up to 55 MeV. The most important point for selecting the high threshold reactions was availability of experimental cross section data beyond 20 MeV which were measured with quasi-monoenergetic p-Li neutron sources [3-6]. All the activation foil samples were pure metal in a uniform size of 5 x 5 mm^2. Thicknesses of bismuth samples were 2 mm while those of the other samples ranged from 0.01 mm to 0.1 mm.

The samples were irradiated by the d-Li neutrons for 20 min, 2 hours or 12 hours. After the irradiations, gamma-rays emitted from the samples were measured with a high purity germanium (HP-Ge) detector. Radioactivity of reaction product for each sample was then determined from the measured gamma-ray peak count, detection efficiency, decay constant, etc. in a unit of reaction per atom per coulomb. A typical overall experimental uncertainty of the measured reaction rates was 5 %.

According to the experience in higher energy dosimetry, features of each dosimetry reaction when it was used practically were revealed. The most remarkable finding is suitability of the ^{169}Tm(n,xn) (x=2, 3 and 4) reactions as dosimeters. Thulium is an element consisting of a single isotope, and the three (n,xn) reactions can be measured very easily in a single and short measurement. Since the only one drawback of the reactions is lack of experimental cross section data above 30 MeV, measurements of the ^{169}Tm(n,xn) reaction cross sections above 30 MeV with quasi-monoenergetic p-Li neutron sources are strongly desired.

2.2. Calculation of Initial Guess Spectra

The initial guess spectrum to be adjusted was calculated by the Monte Carlo neutron transport

Table 1 Dosimetry reactions used for the neutron field characterization experiment.

Reaction	E_{th}[MeV]	Half-Life	Reaction	E_{th}[MeV]	Half-Life
^{27}Al(n,p)^{27}Mg	1.9	9.46 m	^{93}Nb(n,2n)^{92m}Nb	8.9	10.15 d
^{27}Al(n,α)^{24}Na	3.2	14.96 h	^{115}In(n,n')^{115m}In	0.3	4.49 h
^{55}Mn(n,2n)^{54}Mn	10.4	312.1 d	^{169}Tm(n,2n)^{168}Tm	8.1	93.1 d
Fe(n,x)^{54}Mn	0.0	312.1 d	^{169}Tm(n,3n)^{167}Tm	15.0	9.25 d
Fe(n,x)^{56}Mn	3.0	2.58 h	^{169}Tm(n,4n)^{166}Tm	23.7	7.70 h
^{59}Co(n,2n)$^{58m+g}$Co	10.6	70.82 d	^{197}Au(n,2n)^{196m}Au	8.7	9.7 h
^{59}Co(n,3n)^{57}Co	19.4	271.9 d	^{197}Au(n,2n)$^{196m+g}$Au	8.1	6.18 d
^{59}Co(n,p)^{59}Fe	0.8	44.5 d	^{197}Au(n,3n)^{195}Au	14.8	186.1 d
^{59}Co(n,α)^{56}Mn	0.0	2.58 h	^{197}Au(n,4n)^{194}Au	23.2	38.0 h
Ni(n,x)^{58}Co	0.0	70.82 d	^{209}Bi(n,3n)^{207}Bi	14.4	31.6 y
Ni(n,x)^{57}Co	8.3	271.8 d	^{209}Bi(n,4n)^{206}Bi	22.6	6.24 d
Ni(n,x)^{57}Ni	12.4	35.60 h	^{209}Bi(n,5n)^{205}Bi	29.6	15.31 d
^{89}Y(n,2n)^{88}Y	11.6	106.7 d	^{209}Bi(n,6n)^{204}Bi	38.0	11.22 h
^{89}Y(n,3n)$^{87m+g}$Y	21.1	79.8 h			

code MCNP-4B [7] combined with the Monte Carlo Deuteron Lithium (McDeLi) source model [8]. In the calculation, the experimental configuration consisting of the lithium target, container, sample, copper base, cooling water system and surrounding cyclotron parts was modeled. Track length estimators (cell detectors) of 5 x 5 x 1 mm^3 were used for the tally. The Los Alamos LA-150 library [9] up to 150 MeV processed into the ACE type format for MCNP was used as transport cross sections. Since no neutron transport cross section data for lithium was available in the energy range above 20 MeV, cross section data for beryllium up to 100 MeV in the 100XS library [10] was used instead.

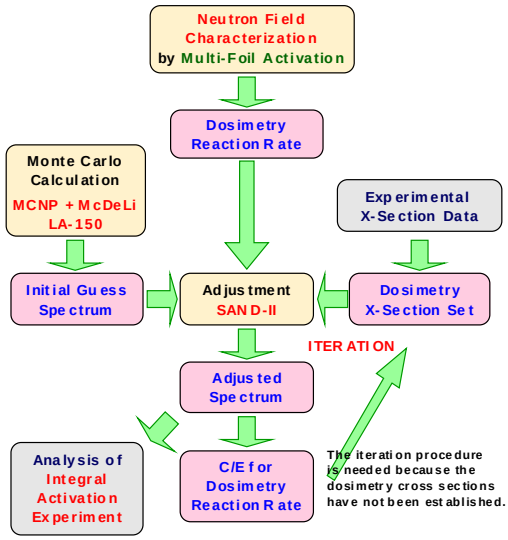

Fig. 2 Flow of the spectrum adjustment.

2.3. Spectrum Adjustment

Figure 2 illustrates a flow of a spectrum adjustment procedure presently adopted. The adjustment required a set of well known dosimetry cross sections. However, such a cross section set extending to 55 MeV was not available especially in the energy region above 20 MeV. Hence, a cross section set was tentatively produced by considering experimental cross section data [3-6], a cross section set extending to 24 MeV determined previously [11], the Intermediate Energy Activation File (IEAF) [12] up to 150 MeV evaluated at Institute of Physics and Power Engineering, Obninsk, Russia, and so on.

The initial guess spectra were adjusted by the SAND-II code [13] with the dosimetry cross section set so as to be consistent with the measured reaction rates. The SAND-II code provided ratios of calculated reaction rates with the adjusted spectrum and the cross section data to the experimental reaction rates (C/E). These C/E values distributed around 1.0 while some of them deviated more than 10 % apart from 1.0. Since the most probable reason for the deviation was lay in uncertainty in the cross section data, cross sections for the largely deviated reactions were revised within a range where consistency with

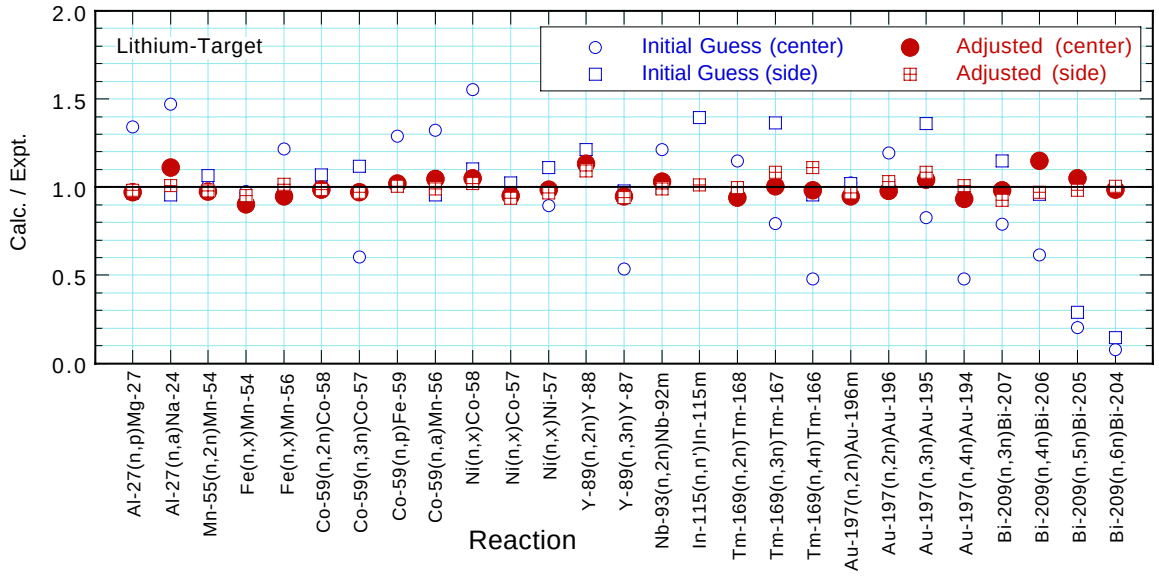

Fig. 3 The C/E values for the dosimetry reactions before and after the adjustment.

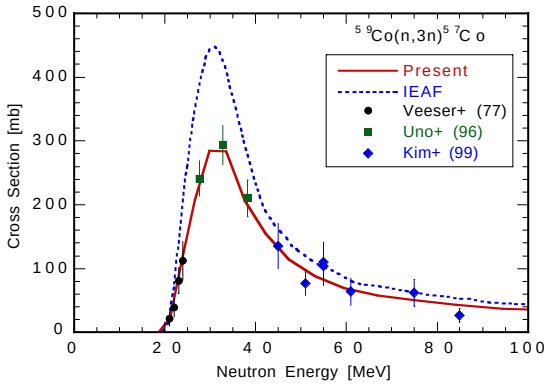

Fig. 4 The ^{59}Co(n,3n)^{57}Co reaction cross sections.

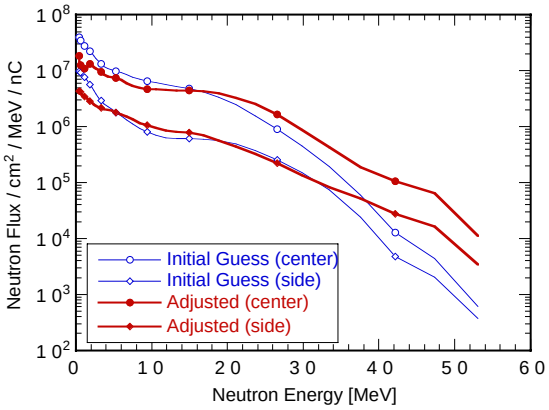

Fig. 5 The initial guess and adjusted neutron flux spectra for the d-Li neutron field.

the experimental cross section data was kept. The initial guess spectra were then adjusted again with the revised cross section data. The procedure of the adjustment and the revision of cross section data was repeated five times to attain convergence of the C/E ratios approximately in a range of 0.9 ~ 1.1 as shown in Fig. 3. An example of the dosimetry cross sections presently adopted is shown in Fig. 4, and more detailed discussion on the dosimetry cross section is given elsewhere [14].

Figure 5 compares the initial guess and adjusted neutron flux spectra at the center and side sample positions. Discrepancies between the initial and adjusted spectra can be explained by deficiencies in the calculation of initial guess spectra: omission of neutron production in the stainless steel case and the use of the beryllium cross section data for lithium. Nevertheless, the initial guess and adjusted neutron flux above 10 MeV agree within 10 % with each other for both the center and side positions. This good agreement gives experimental validation for the MCNP/McDeLi codes which are used for IFMIF neutronics designs.

3. Integral Activation Experiment

3.1. Experiment

Four samples of fusion reactor structural materials, i.e., pure-vanadium, vanadium-alloy (Ti: 4%, Cr: 4%, V: balance), SS-316LN of ITER-grade (Cr: 17.5%, Ni: 12.3%, Mo: 2.5%, Mn: 1.8%, Si: 0.4%, Fe: balance) and low activation ferritic steel F82H (Cr: 7.7%, W: 1.95%, Mn: 0.16%, V: 0.16%, Si: 0.11%, Fe: balance), were irradiated at the center position in the d-Li neutron field for 1 hour. After the irradiation, radioactivities induced in these samples were measured by the HP-Ge detector at several cooling times ranging from 1 hour to 5 months.

3.2. Low-Activation Property

Low-activation properties for the sample materials irradiated in the IFMIF-like neutron field were demonstrated. After 5 months since the irradiation, the total activity of F82H steel, 1.6 x 10^4 Bq/g, was considerably less than that of SS-316LN, 5.1 x 10^4 Bq/g, mainly due to the reduction of nickel and molybdenum content in F82H. Low-activation property was further enhanced in vanadium-based materials: the total activities after 5 months cooling were 2.9 x 10^3 Bq/g and 1.1 x 10^3 Bq/g for vanadium-alloy and pure-vanadium samples, respectively, which were less than the total activity of F82H steel by about one order.

3.3. Sequential Charged Particle Reaction

Figure 6 shows a gamma-ray spectrum for the pure-vanadium sample measured after 1 month cooling. Although vanadium does not produce ^{51}Cr via the ordinary neutron induced reactions, the strongest radioactivity observed is ^{51}Cr. Contents of impurity elements that can produce ^{51}Cr, such as chromium and manganese, are too low to explain the ^{51}Cr production. The reaction mechanism to produce ^{51}Cr in vanadium is the sequential charged particle reaction (SCPR) which is illustlated in Fig. 7. A charged particle, a proton in this case, is produced by the V(n,xp) reaction, then the proton induces the ^{51}V(p,n)^{51}Cr reaction sequentially. The SCPR process should be considered when we estimate radioactive inventories in low-activation materials irradiated in higher energy neutron fields.

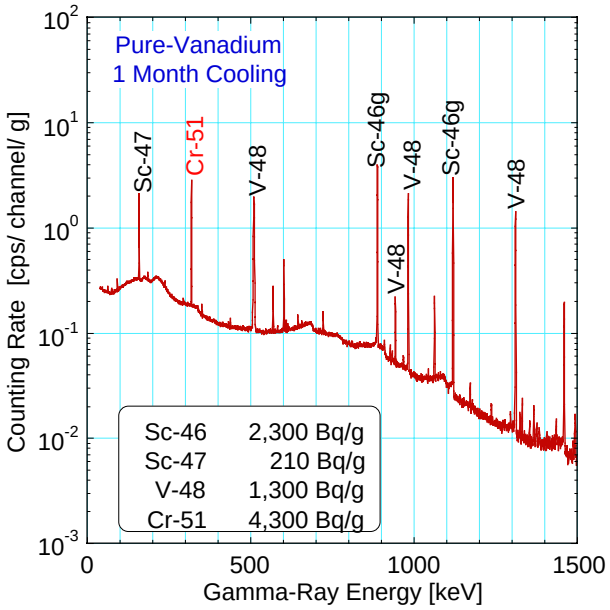

Fig. 6 Gamma-ray spectrum for the pure-vanadium sample measured after 1 month cooling.

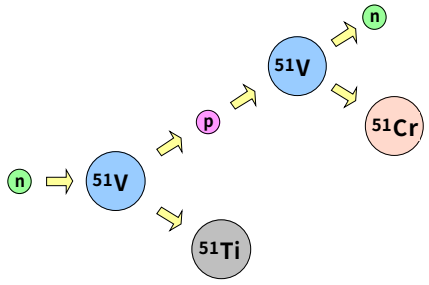

Fig. 7 Sequential charged particle reaction.

3.4. Validation of IEAF and ALARA

An experimental analysis was performed with IEAF and ALARA. The IEAF [12] is practically the only existing activation cross section library which includes activation cross section data for higher energy

Table 2 The C/E ratios of radioactivities intensely observed (> 1% to the total activity).

Nuclide	Half-Life	SS-316LN	F82H Steel	Vanadium-Alloy	Pure-Vanadium
Sc-43	3.9 hr			0.024± 30%	
Sc-44	3.9 hr			1.50 ± 20%	
Sc-46	84 d			0.91	0.77
Sc-47	3.4 d			7.73	8.67
Sc-48	44 d			0.98	1.05
V-48	16 d			1.11	1.14
Cr-49	42 min			0.75 ± 15%	
Cr-51	28 d	1.06	1.08	1.00	*0.0004*
Mn-52	5.6 d	3.28	3.78		
Mn-54	312 d	1.05	1.06		
Mn-56	2.6 hr	1.11	1.12		
Co-56	77 d	3.60	*0.02*		
Co-57	272 d	1.02			
Co-58	71 d	1.13			
Ni-57	36 hr	1.25 ± 20%			
Nb-92m	10 d				1.32 ± 30%
Mo-99	66 hr	1.26			
Tc-99m	6.0 hr	1.26			
W-187	24 hr		0.54		

Estimated experimental uncertainty ± 10% except where indicated.

neutrons beyond 20 MeV. The IEAF adopts cross section data in EAF-97 [15] in the energy range below 20 MeV. The ALARA, "Analytic and Laplacian Adaptive Radioactivity Analysis", is a radioactivity inventory calculation code developed at FZK [16].

Table 2 summarizes ratios of C/E values for radioactivities which are observed intensely (> 1% to the total activity). Tow very low C/E values, 0.0004 for ^{51}Cr in pure-vanadium and 0.02 for ^{56}Co in F82H, can be attributed to omission of the SCPR process in the calculation. The ^{56}Co is produced mainly by the ^{56}Fe(p,n)^{56}Co reaction where protons are provided by neutron induced proton emission reactions. The very large C/E values of ~ 8 for ^{47}Sc are due to a problem in the ^{51}V(n,nα)^{47}Sc cross section in EAF-97 which has been pointed out previously [17]. Except for these particular cases, the calculated values agrees with the experimental data. Accordingly, the activation cross sections in IEAF were validated experimentally by the integral activation experiment that dealt with neutrons up to 55 MeV.

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