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INTEGRAL ACTIVATION CROSS SECTION RATIOS OF $^{58}Ni(n,x)^{57}Co$, $^{60}Ni(n,p)^{60}Co$, $^{63}Cu(n,\alpha)^{60}Co$, $^{197}Au(n,3n)^{195}Au$ RELATIVE TO $^{27}Al(n,\alpha)^{24}Na$ IN THE NEUTRON SPECTRUM PRODUCED BY 23.2 MeV DEUTERONS INCIDENT ON A THICK Be METAL TARGET

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INTEGRAL ACTIVATION CROSS SECTION RATIOS OF Ni58(n,x)Co57, Ni60(n,p)Co60, Cu63(n,α)Co60, Au197(n,3n)Au195 RELATIVE TO Al27(n,α)Na24 IN THE NEUTRON SPECTRUM PRODUCED BY 23.2 MeV DEUTERONS INCIDENT ON A THICK Be METAL TARGET

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ABSTRACT

Integral activation cross section ratios of Ni58 (n,x)Co57, Ni60(n,p)Co60, Cu63(n,α)Co60 and Au197 (n,3n)Au195 have been measured relative to Al27(n,α) Na24, in the neutron spectrum produced by 23.2 MeV deuterons incident on a thick Be target.
Validation of these cross sections in a high energy neutron spectrum is the object of this work. A close examination of the spectrum of our facility (ITER1) and sensitivity functions shows that the response of these cross sections is quite significant between 10-15 MeV to 15-22 MeV.
Several evaluated cross sections libraries have been used to calculate the integral activation cross sections in the irradiation spectrum. In general the agreement between ENDF/B-VI or IRDF90 calculated values and experimental results is excellent for Ni58(n,x)Co57, Cu63(n,α)Co60 and Au197(n,3n)Au195, well within the experimental errors. However for Ni60 (n,p)Co60, excluding evaluated libraries based on too high Paulsen data (Chinese, JENDL), the nuclear model calculation evaluated library EFF-2 shows a better agreement (E/C-1 ~ 9.7%) with our experimental result than ENDF/B-VI (E/C-1 ~ 16%).

Introduction

Ni58(n,x)Co57, Ni60(n,p)Co60 and Cu63(n,α)Co60 activation cross section reactions are important nuclear data in reactor technology. On the other hand Au197(n,3n)Au195 is quite sensitive to high energy neutrons (~ 20 MeV) and may be a good high energy neutron dosimeter.

Stainless steel with different content of Ni are the constituents of fission reactors, TOKOMAK fusion reactors or accelerators spallation systems. Cu is also an important reactor material.

Co60 produced by the neutron field in the reactor structural material with several years of cooling after the reactor shut-down is the main residual activity.

These long lived activities may be useful neutron integrators in fast neutron dosimetry. As Co60 can be produced by Co59(n,γ)Co60, spectral index may be measured in the mixed spectrum neutron field, independently of the gamma detector efficiency.

Validation of some of these cross sections have been made in a Cf252 neutron spectrum, in a U235 fission spectrum and

several fast reactor spectra (En < 2 MeV). The only validation in a high energy spectrum was made in the neutron field produced by bombardment of 2 MeV deuterons in a thick Li target[1]. However the activity produced by neutrons above 17 MeV in this spectrum is small, being quite insensitive to the cross section behaviour between 15 and 22 MeV.

The object of this work is the validation of Ni58(n,x)Co57 Ni60(n,p)Co60 Cu63(n,α)Co60 and Au197(n,3n)Au195 in the neutron spectrum produced by 23.2 MeV incident deuterons on a thick Be metal target.

The neutron spectrum shape of our facility, fig. 1, must be closely examined. The main characteristic is that between 10 and 20 MeV the flux changes only a factor of seven. This makes the response (sensitivity function between 10-15 and 15-22 MeV) quite significant. See for instance the sensitivity function of Ni58(n,x)Co57,fig. 2, compared with the sensitivity function on Cf252 Spectrum.

The spectrum has been obtained by the unfolding technique with the input spectrum measured by TOF by Lone et al., and validated by measuring U238/U235, Th232/U235, Th232/U238 and U236/U235 fission ratios. The agreement with ENDF/B-6 calculated ratios is around 2 %.

I- Experimental

I-a. Irradiation and neutron flux monitoring

The activity of nickel, copper and gold foils, irradiated in two previous experiments[2,3] was measured after total decay of stronger shorter reactions, to obtain the long lived reactions of Ni58(n,np)Co57, Ni60(n,p)Co60, Cu63(n,α)Co60 and Au197(n,3n) Au195.

Experiments were made at the tandar accelerator of the CNEA, with deuterons of 23.2 MeV and the detectors were placed at 4 cm from the beam spot in the Be target.

The Ni foil irradiated in the unfolding experience TAN2 of ref. 2 was used to measure the Ni58(n,x)Co57 and Ni60(n,p)Co60 reactions. The Cu and Au foils irradiated in TAN2 and in TAN9 of ref. 3, were used to measured the Cu63(n,α)Co60 and Au197 (n,3n)Au195 reactions.

The activation stacks were irradiated together with a SSNTD stack, to assure exactly the same spectrum in both irradiations and Na24 activity distribution in Aluminum foils was used to check geometrical reproducibility and flux monitoring, as described in ref. 2 and ref. 3.

The nickel foil was 0.254 mm thick and 99.97 % pure and the copper foil 0.28mm thick and 99.999 % pure, the diameter of both foils was 0.5" . The gold foil was of 1 cm diameter and 0.07 mm thick.

I-b. Activation Measurements.

The foils activities were measured in a 20% reverse HP-Ge diode of thin Be window, against standard calibrated sources of the same energy, at 3 cm distance from the detector.

The corrections due to different counting geometry between the sample and the calibration source were experimentally obtained, by measuring the radial and axial efficiency variation of point gamma sources. The correction due to the difference in efficiency of the finite diameter foils against the point

standard gamma sources, is 0.6 % at 3 cm from the diode and 1.8 % at 1 cm, with 0.5 % error. The error due to axial difference in the mounting of the sample and the gamma calibration sources, was estimated to be less than 0.5%, from the experimental axial efficiency variation curves.

The 122.061 keV Co57 photopeak activity was measured against a Co57 calibrated source (3% error).The Ni58(n,np)Co57 activity competes with the Ni58(n,2n)Ni57 activity, Ni57 (1.503 days) has totally decayed to Co57 (271.79 days) at the measuring time. So the Co57 activity is a a measure of the (n,np) plus the (n,2n) Ni58 reactions and we refer to this activation cross section as Ni58(n,x)Co57. A -6 % calculated correction (Table 4) was applied to obtain the Ni58(n,np)Co57 cross section.

The 1173.238 and 1332.502 Co60 photopeaks from Ni60(n,p) Co60 and Cu63(n,α)Co60 reactions were measured against a Co60 calibration source (2.6 % reported uncertainty).

The 186.09 days, (10.9±0.5) %, Au195 98.83 keV gamma ray[4] from the Au197(n,3n)Au195 reaction was measured against the (3.63±0.02)%, 88.034 keV[5] photopeak of Cd109 calibrated gamma source (2.3 % error). To estimate the efficiency curve shape, the 59.537 keV Am 241 and 122.7179 keV gamma rays were also measured to determine the efficiency of the Au195 photopeak. A problem in measuring this low energy, arises from the fact that the photopeak appears above a considerable background activity and the background subtraction increases the dispersion of the measurements. The Au197(n,3n)Au195 was also measured at 1 cm from the diode. This improves greatly the dispersion but the correction due to the finite foil size relative to the point calibration source increases to 1.8%±0.5%. The reported value of table 1. is the weighed average of both measurements. Besides the gamma self absorption in the foil is large (around 25 %) and the exponential approximation may be defective. To investigate this point we measured the activity using gold absorbers of different thickness and deduced the photonic absorption coefficient with the exponential approximation and with a expression that allows for the angle sustained by a point sample to the detector. From the transmission measurements we concluded that the second expression was better and the self absorption correction coefficient, fG, is 0.733±4%. A -4.6% correction (0.5% error) was applied because the foil diameter is 1cm and the other foils diameter is 0.5", as discussed in ref. 2. In addition there is a 5% error due to the uncertainty in the percentage of the 98.8 keV Au195 photopeak that was not included in the error calculations.

Self-absorption in samples other than gold was calculated assuming exponential approximation and photonic absorption coefficients from Shirley[4] and the error was estimated as half the correction.

The saturation absolute activities per Ni58 or Ni60 isotope were corrected by gamma absorption in the Ni foil and normalized to the flux in the backing of the irradiation stack[2] as the standard reaction Al27(n,α)Na24 of the same experience (TAN2[2]).

To obtain the Cu63(n,α)Co60 and Au197(n,3n) saturation activity, the flux difference and decay time between the two irradiation runs (TAN2 and TAN9) was taken into account for normalization to TAN9 and to the irradiation stack backing[3]. Both

reactions were related to the Al27(n,α)Na24 activity of TAN9[3] to obtain the cross sections.

The relative error in the normalization factors to the back stack flux is 0.5 %.

The experimental integral cross sections and errors are summarized in Tables 1 and 2.

Table 1. Experimental Results

Reaction	Run	Rate (d/s/atom)	Ratio	<σ> (mbarn)
Al27(n,α)Na24	TAN2	1.97E-16±2.0%	1	40.54[a]
Ni58(n,x)Co57	TAN2	7.21E-16±3.7%	3.662	148.5±4.2%
Ni58(n,np)Co57	TAN2			140.3±4.2%[b]
Ni60(n,p)Co60	TAN2	2.90E-16±2.9%	1.474	59.7±3.4%
Al27(n,α)Na24	TAN9	3.85E-16±2.2%	1	40.54a
Cu63(n,α)Co60	TAN9	1.43E-16±2.7%	0.372	15.1±3.5%
Au197(n,3n)Au195	TAN9	4.62E-16±5.2%[c]	1.199	48.6±5.7%[c]

[a] <σ> Al27(n,α) calculated using unfolded spectrum ITER1 and differential data from IRDF90.
[b] The (n,2n) reaction contribution was deduced from the calculated values of table 4.
[c] 5% error in the Au195, 98.8keV, gamma ray not included.

Table 2. Source of Errors in Integral Cross Sections

error source	Ni58(n,np)	Ni60(n,p)	Cu63(n,α)	Au197(n,3n)
NON CORRELATED				
statistics	0.4 %	0.5 %	0.2 %	2.3 %
self-absorp.	2.0 %	0.3 %	0.4 %	4.0 %
flux normal.	0.5 %	0.5 %	0.5 %	0.5 %
axial position	0.5 %	0.5 %	0.5 %	0.5 %
diameter dif.	0.0 %	0.0 %	0.0 %	0.5 %
100 % CORRELATED				
Cal. sources :				
Co57	3.0 %	0.0 %	0.0 %	0.0 %
Co60	0.0 %	2.6 %	2.6 %	0.0 %
Cd109	0.0 %	0.0 %	0.0 %	2.3 %
reference reaction :				
TAN2	2.0 %	2.0 %	0.0 %	0.0 %
TAN9	0.0 %	0.0 %	2.2 %	2.2 %
sample size	0.5 %	0.5 %	0.5 %	0.5 %
Total Error	4.2 %	3.4 %	3.5 %	5.7 %

Ni58(n,np) and Ni60(n,p) integral cross section errors are about 30% correlated due to the error of the measured Na24 activity in TAN2.Ni60(n,p)Co60 and Cu63(n,α)Co60 were measured relative to the same Co60 gamma calibration source and their error correlation is greater than 50%, see Table 3.

Correlation for Cu63 (n,α) and Au197(n,3n) is 25%. If the 5% gamma percentage error is taken into account the error of

Au197(n,3n)Au195 cross section would amount to 7.5% and their correlation with Cu63(n,α)Co60 cross section would diminish to about 20%.

Table 3. Cross Sections Correlation Matrix

	Ni58(n,np)	Ni60(n,p)	Cu63(n,α)	Au197(n,α)
Ni58(n,np)Co57	1.000			
Ni60(n,p)Co60	0.292	1.000		
Cu63(n,α)Co60	0.017	0.574	1.000	
Au197(n,3n)Au195	0.010	0.013	0.252	1.000

II- Cross Sections Calculation

Several cross sections libraries have been used for Ni58(n,np)Co57, Ni58(n,2n)Ni57, Ni60(n,p)Co60 and Au197(n,3n)Au195 :
1) International Reactor Dosimetry File[6,7] IRDF90. 2) ENDF/B-VI evaluated neutron dosimetry file[8]. 3) The Chinese evaluation made by Zhao Wenrong et al[9]. 4) EFF-2, European Fusion File[10]. 5) The JENDL Dosimetry File[11]. However only for Ni60(n,p)Co60 the five libraries were available.

These evaluated cross section have been integrated with the ITER1 spectrum as reported in a previous work[2].

In our experiment we measure the Co57 activity, then it is easy to show that the cross section is the sum of Ni58(n,np)Co57 and Ni58(n,2n)Ni57. However as it is shown in Table 4, the contribution of Ni58(n,2n)Ni57 is less than 6%. Ni58(n,np) and Ni58(n,2n) integral cross sections were calculated until 20 MeV, with ENDF/B-VI and JENDL-3 . Above 20 MeV up to 30 MeV the (n,np) reaction was extrapolated following the shape of the curve quoted by Cierjacks et al.[12]. The (n,2n) reaction from 20 to 30 MeV was calculated with the normalized cross sections from Bayhurst et al.[13]. From these calculations was obtained the ratio, (n,x) / (n,np) = 1.058, to correct for the (n,2n) contribution to the experimental value of the (n,x) reaction of Table 1.

Table 4. Calculated Integral Cross Sections of Ni58 (n,np) and (n,2n) Reactions

	Ni58(n,np)Co57	Ni58(n,2n)Ni57	Ni58(n,x)Co57
ENDF/B-VI	141.7 mbarn	8.41 mbarn	150.11 mbarn
JENDL-3	140.2 mbarn	8.01 mbarn	148.21 mbarn

In table 1 are shown the integral activation cross sections obtained in our experiment relative to the standard.

The Al27(n,α)Na24 standard reaction cross section was calculated, as described in a previous work[3], with IRDF90. The value obtained is 40.54 mbarn and agrees within 1.5% with other evaluations (Table 2 of ref. 2).

III- Comparison of Experimental and Calculated Results

The experimental and calculated results are compared in Table 5.

For Ni58(n,np)Co57 the agreement between calculated and experimental is excellent (~1%) well within the experimental error (~4%), suggesting that may be a good neutron dosimeter for high energy neutrons (see sensitivity function fig. 2).

Ni60(n,p)Co60 has been object of investigation in the last years because previous evaluations were based in data from Paulsen[14]. However evaluated data based on Paulsen results give <σ> on the Cf252 spectrum higher than the experimental data result[15].

Two investigations presented at the Jülich Conference by M. Wagner et al[16], and S. Sudár et al[17], confirm that Paulsen data are definitively too high between 5 and 12 MeV. JENDL and the Chinese evaluation are based on Paulsen data. ENDF/B-VI and IRDF90 is the same evaluation and EFF-2 is a model calculation.

Then the fact that JENDL and the Chinese evaluation agrees with our results is only incidental, because they disagree for Cf252 spectrum, Table 6.

Comparison with IRDF90 and EFF-2 shows a better agreement with EFF-2, suggesting that evaluated libraries give a lower cross section above 15 MeV, as it is shown in recent experimental data between 17 and 20 MeV[18].

For <σ> of Cu63(n,α)Co60 the agreement (~1%) with IRDF90 (ENDF/B-VI) is excellent but is poor (-12%) with JENDL. In fig. 4 can be observed that these libraries differ above 10 MeV. In the Cf252 spectrum both evaluations give a reasonable agreement with the experimental value, Table 6, because the small sensitivity of Cu63(n,α)Co60 in a Cf252 neutron spectrum above 10 MeV as shown in fig. 5.

Table 5. Calculated Integral Cross Sections with Evaluated Libraries and Experimental Results

	Ni58(n,np)Co57		Ni60(n,p)Co60		Cu63(n,α)Co60		Au197(n,3n)	
LIBRARY[a]	<σ>	C/E-1	<σ>	C/E-1	<σ>	C/E-1	<σ>	C/E-1
	mbarn	%	mbarn	%	mbarn	%	mbarn	%
IRDF90[b]			50.03	-16	14.90	-1.3		
ENDF/B-VI	141.7	+1.0					46.76	-3.8
CHINESE			57.90	-3.0			46.86	-3.6
JENDL	140.2	-0.1	58.74	-1.6	13.30	-12		
EFF-2			53.91	-9.7				
EXP[c]	140.3±4.2%		59.7±3.4%		15.1±3.5%		48.6±5.7%	

[a] <σ> averaged on unfolded neutron spectrum ITER1.
[b] Same evaluation as ENDF/B-VI.
[c] Measured Integral Cross Section (Table 1).

Finally, as shown in Table 5, for Au197(n,3n)Au195 the agreement with ENDF/B-VI and the Chinese evaluation is excel-

lent (~3.7%). This reaction ia quite sensitive to neutrons around 20 MeV, fig. 5, indirectly showing that the spectrum shape in that energy region is correct.

Table 6. Cf252 Spectrum Averaged Neutron Cross Sections of Cu63(n,α)Co60 and Ni60(n,p)Co60

LIBRARY[a]	Cu63(n,α)Co60		Ni60(n,p)Co60	
	<σ>	C/E-1	<σ>	C/E-1
	mbarn	%	mbarn	%
ENDF/B-VI	0.6878	-0.28	2.530	+5.8
JENDL-3	0.6871	-0.38	3.478	+45
Chinese			4.504	+88
EFF-2			2.507	+4.8
Mannhart[b]	0.6897±1.9%		2.39±5.4%	

[a] Calculated value on Cf252 spectrum (Watt, Ew=0.359, Tw=1.175).
[b] Recommended experimental values from ref. 15.

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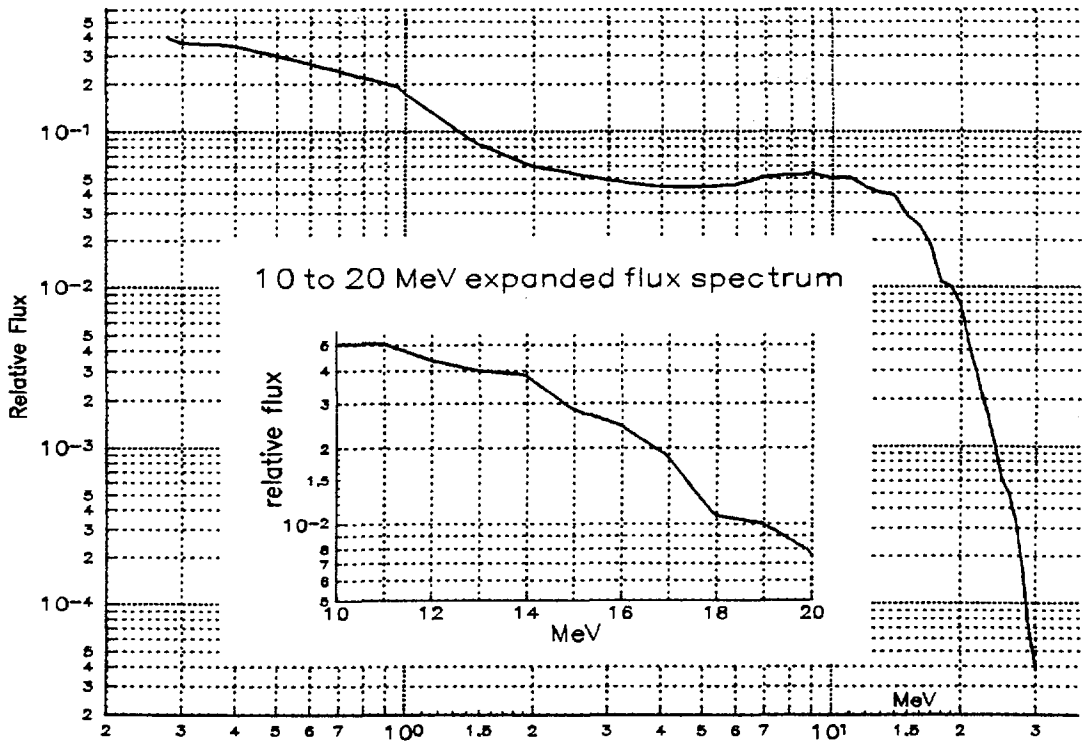

Fig. 1. Neutron Flux Spectrum on Irradiation Site, ITER1.

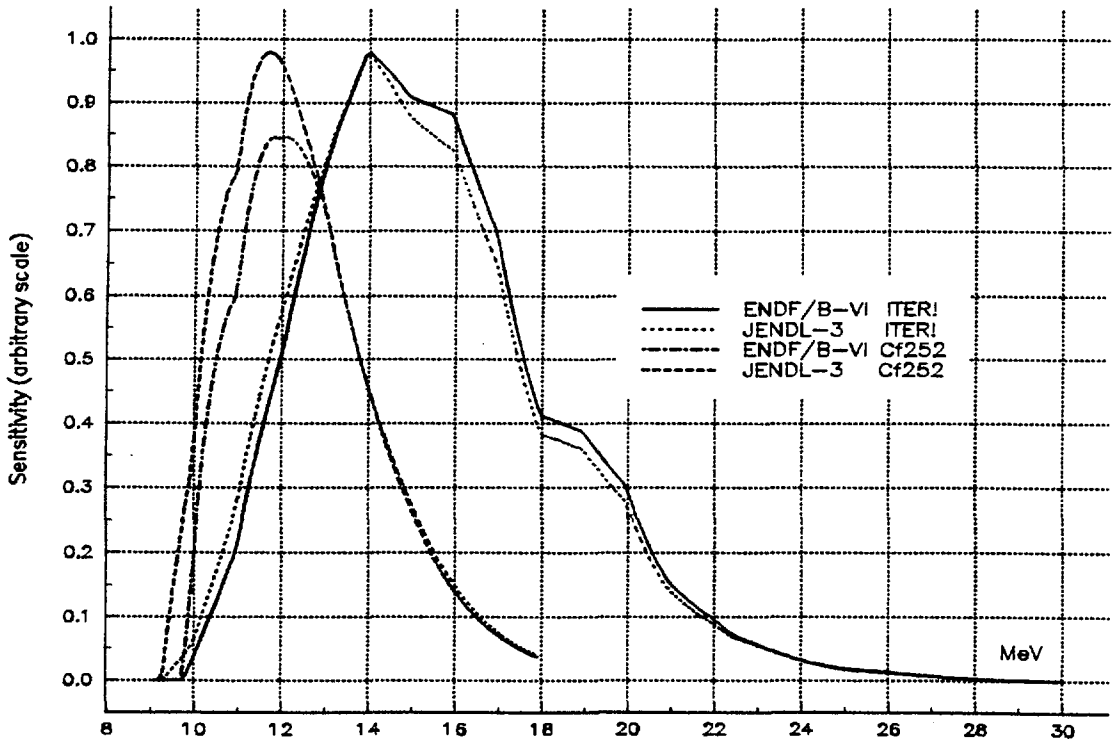

Fig. 2. Ni58(n,np)Co57 Sensivity in ITER1 and Cf252 Spectra.

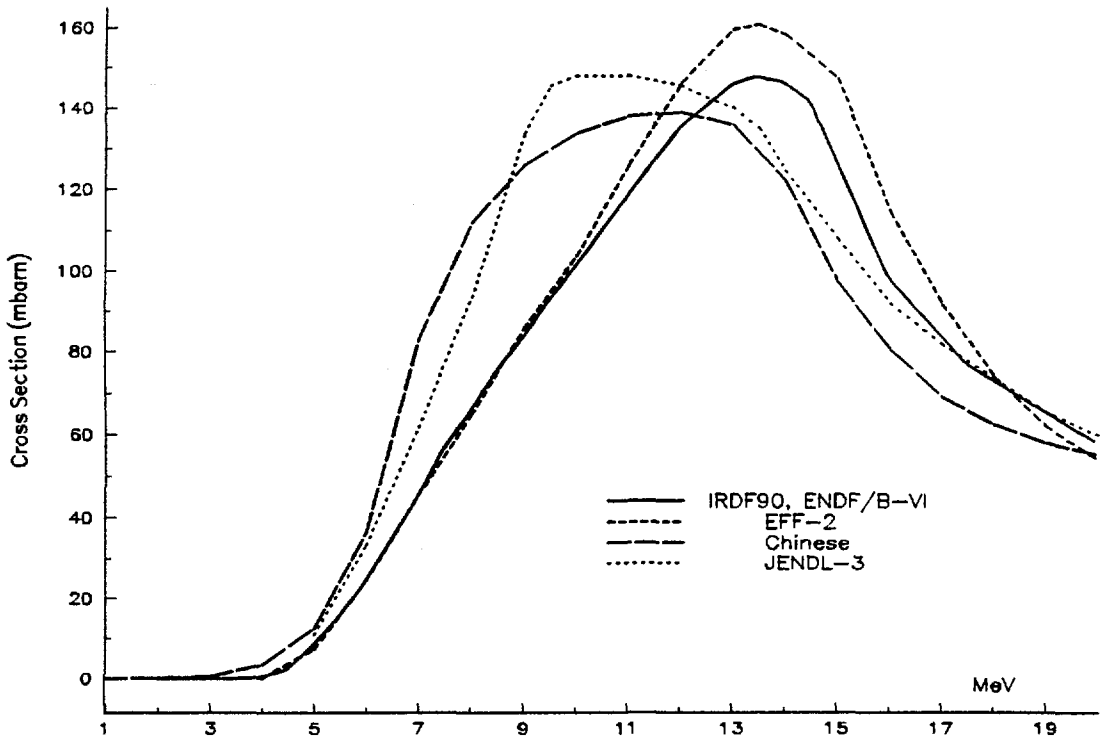

Fig. 3. Ni60(n,p)C060 Evaluated Cross Sections.

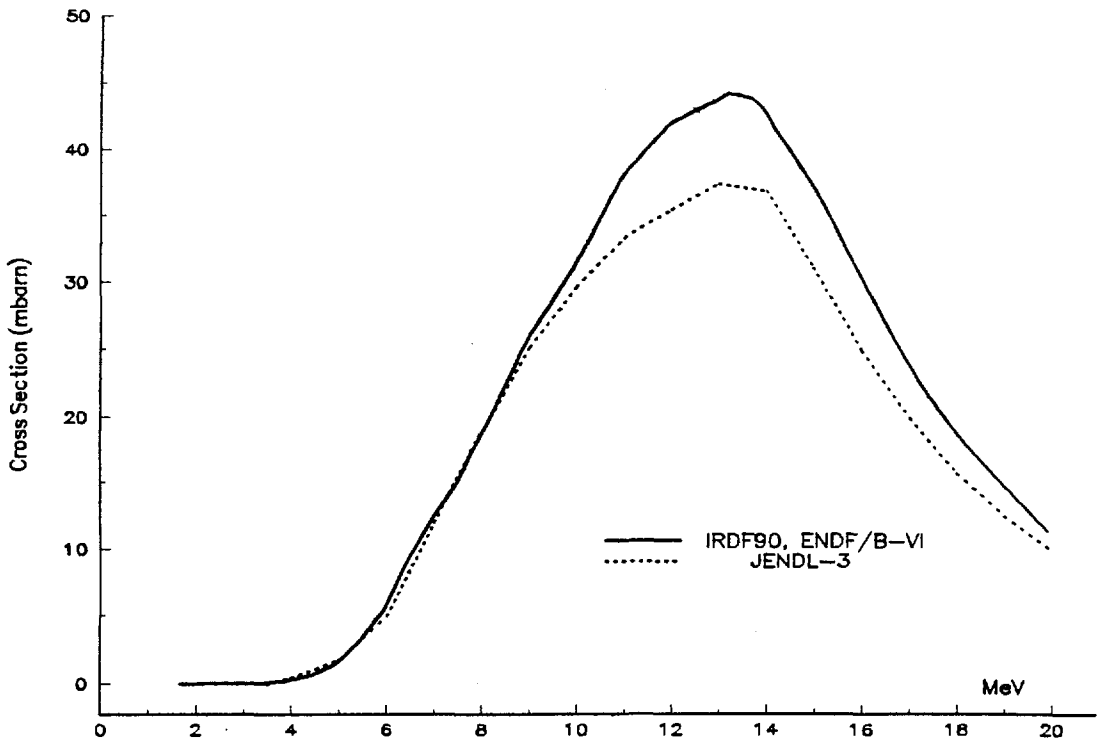

Fig. 4. Cu63(n,α)Co60 Evaluated Cross Sections.

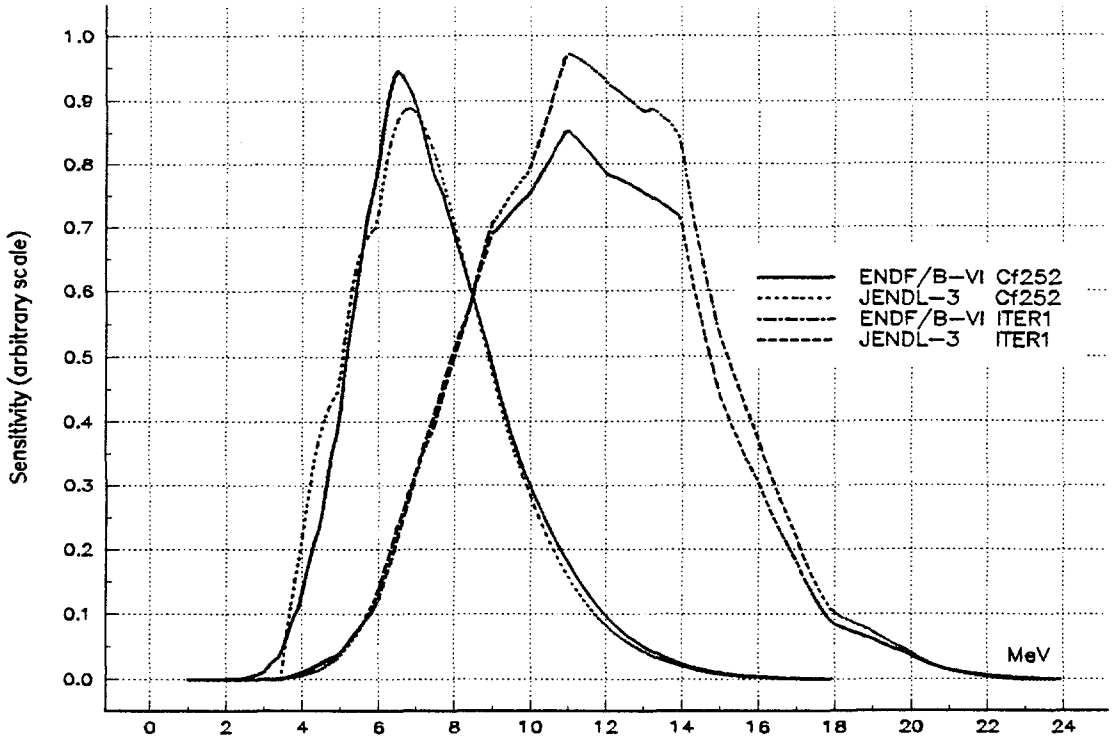

Fig. 5. Cu63(n,α)Co60 Sensitivity in ITER1 and Cf252 Spectra.

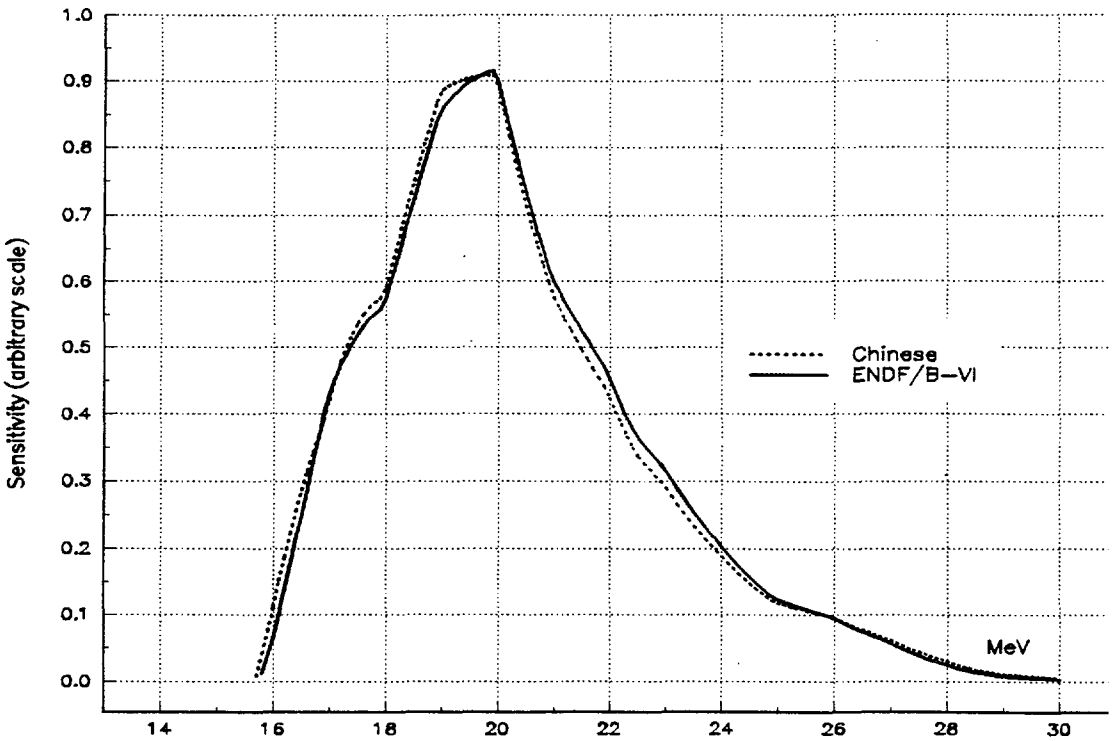

Fig. 6. Au197(n,3n)Au195 Sensitivity in ITER1 Spectra.

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