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ACTIVATION CROSS SECTIONS FOR THE GENERATION OF LONG-LIVED RADIONUCLIDES OF IMPORTANCE IN FUSION REACTOR TECHNOLOGY

Proceedings of an IAEA Consultants' Meeting held at
Argonne National Laboratory, Argonne, Illinois, USA
11 - 12 September 1989

Edited by Wang DaHai

January 1990

IAEA NUCLEAR DATA SECTION, WAGRAMERSTRASSE 5, A-1400 VIENNA

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Foreword

Along with the development of fusion reactor research, more activation cross-section data for the generation of long-lived radionuclides are needed for estimation of radioactive waste and selection of materials leading to low-level activities. At its 16th Meeting, the International Nuclear Data Committee (INDC) recommended to establish a Coordinated Research Programme (CRP) to measure and evaluate selected important activation cross sections leading to the production of long-lived radionuclides.

Following these recommendations, the IAEA Nuclear Data Section has established a new CRP concentrating on the cross sections for the reactions suggested by the 16th INDC meeting.

This Consultants' Meeting was actually the first of the Research Coordination Meetings of the CRP. The main objectives of the meeting were to review the first results under the CRP and the status of long-lived activation cross section data, exchange experience in measurement and evaluation, and to fix the future working programme for the CRP.

The proceedings contain the progress reports of the CRP and the contributed papers presented at the meeting as well as the summary of the conclusions and recommendations of the meeting.

The Scientific Secretary of the meeting wishes to express his appreciation to the Argonne National Laboratory, especially Dr. D.L. Smith, for their kind assistance in the organization and preparation of the meeting. His particular thanks and gratitude go to Prof. Dr. H. Vonach for acting as Chairman and preparing the summary of the conclusions and recommendations of the meeting.

Contents

IAEA Consultants' Meeting on Activation Cross Sections for the generation of long-lived Radionuclides for Fusion Reactor Technology

Argonne National Laboratory
Argonne, Illinois, U.S.A.

Agenda

Monday, 11 September 1989

08:00 Bus leaves Holiday Inn for Argonne National Laboratory

08.30 1. Opening of Meeting

Adoption of Agenda

08:50 2. Progress reports by several participants to the Coordinated Research Programme (CRP) entitled "Activation Cross Sections for the Generation of Long-Lived Radionuclides" on their work

- H. Vonach (Austria)
- Lu Hanlin (China)
- S.M. Qaim (FRG)

10:30-10:45 Break (15 minutes)

10:45
- J. Csikai (Hungary)
- B. Patrick (UK)

12:00-14:00 Lunch - ANL Cafeteria

14:00
- D.L. Smith (USA)

3. Recent Measurements of Production of Long-Lived Activities in Fusion Materials

- L.R. Greenwood (USA)

15:30-15:45 Break (15 minutes)

15:45 4. Discussions on Technical Problems with the Experiments

17:00 Bus leaves ANL for Holiday Inn

Tuesday, 12 September 1989

08:00 Bus leaves Holiday Inn for ANL

08:30 5. Theory used for the Estimation of Activation Cross Sections Leading to Long-Lived Radionuclides

- C.Y. Fu (USA)
- N. Yamamuro (Japan)

6. Activation Cross Section Data Needs for Fission and Fusion Reactor Technology

- E.T. Cheng (USA)

10:30–10:45 Break (15 minutes)

10:45 7. Review of the Data Status of Activation Cross Sections for the Generation of Long-Lived Radionuclides and its Potential Impact on the Work of the CRP

8. Work Programme of the CRP for 1989–1990

12:30– Lunch - ANL Cafeteria

14:00–15:00 Draft of conclusions and recommendations

15:00 Discussion of the conclusions and recommendations

17:00 Bus leaves ANL for Holiday Inn

Conclusions and Recommendations

(1) Remarkable progress has been obtained in the first year of the CRP as can be seen from Table 1 which shows the status of the data for the 16 reactions originally included in the CRP. Cross-sections have been obtained from either measurements or evaluations of measurements outside the CRP for 8 reactions, for another 5 reactions measurements are in progress.

(2) The existing and ongoing measurements, evaluations and calculations were discussed in detail and the following conclusions were reached:

a) $^{27}Al(n,2n)^{26}Al$. The existing measurements will probably satisfy the data needs but need to be combined in a good evaluation.

b) $^{63}Cu(n,p)^{63}Ni$. The existing and ongoing measurements will probably satisfy the data needs, unless serious discrepancies are found. The situation has to be discussed once more after completion of the ongoing measurements, considering the theoretical calculations done at Oak Ridge and Nuclear Data Engineering Inc., Japan.

c) $^{94}Mo(n,p)^{94}Nb$. One measurement exists from outside the CRP, one more measurement from inside the CRP seems desirable to compare with this measurement. Possibly this will be done in a Jülich-Debrecen collaboration, also, Dr. Ikeda from Japan will be approached on this problem.

d) $^{109}Ag(n,2n)^{108m}Ag$. Two measurements of this cross-section have been performed which are in good mutual agreement. There is, however, a serious discrepancy between these measurements and KRI's results and an evaluation based on the systematics of total (n,2n) cross-sections and the cross section for $^{109}Ag(n,2n)^{108m}Ag$ (see Table 1); a possible cause may be a serious error in the half-life of ^{108m}Ag, which therefore should be checked.

e) $^{179}Hf(n,2n)^{178m}Hf$. Data needs will probably be satisfied by the completed and ongoing measurements and calculations. A final conclusion can be made after completion of the ongoing measurements and should be discussed at the next meeting.

f) $^{151}Eu(n,2n)^{150m}Eu$, $^{153}Eu(n,2n)^{152g}Eu$ and $^{159}Tb(n,2n)^{158g}Tb$. The existing measurements within the CRP and the evaluation of previous measurements are in good agreement and definitely meet the data needs. They should be combined in a common evaluation.

g) $^{193}Ir(n,2n)^{192m2}Ir$ and $^{187}Re(n,2n)^{186m}Re$. Cross-sections have been derived from the systematics of total (n,2n) cross-sections and measured values for the $^{193}Ir(n,2n)^{192g+m1}Ir$ and $^{187}Re(n,2n)^{186m}Re$ cross-sections. These evaluations in principle satisfy the data needs, however, one additional confirmation of the $^{187}Re(n,2n)^{186g}Re$ results seems desirable.

h) $^{165}Ho(n,\gamma)^{166m}Ho$ and $^{98}Mo(n,\gamma)^{99}Mo$. Research contracts for the study of these two reactions have been awarded to the Sichuan University, Chengdu, China, by the IAEA; results can be expected at the next CRP meeting.

i) $^{62}Ni(n,\gamma)^{63}Ni$: No work has been done as evaluated data for this reaction are available within ENDF/B-VI.

j) $^{158}Dy(n,p)^{158g}Tb$ and $^{191}Ir(n,\gamma)^{192m2}Ir$. No work on these reactions could be done because the experimental study of these reactions is extremely difficult due to very small cross-sections and large activities from interfering reactions. In this situation the need for these cross-sections should be critically reviewed by the data users. For the reaction $^{191}Ir(n,\gamma)^{192m2}Ir$ it seems possible to measure the resonance capture integral, however, only after a cooling period of several years.

(3) According to the situation described it was decided to take the following actions in order to fulfill the programme of the CRP as much as possible:

a) Dr. W. Mannhart will ask the International Committee for Radionuclide Metrology (ICRM) to check the half-life of ^{108m}Ag.

b) Dr. Lu Han-Lin will perform a new measurement of the $^{187}Re(n,2n)^{186g}Re$ cross-section.

c) Dr. Lone, Chalk River, will investigate the possibility to measure the resonance capture integral for the $^{191}Ir(n,\gamma)^{192m2}Ir$ reaction at one of the Chalk River reactors.

d) Dr. Cheng will check the needs for the measurement of the very difficult reactions $^{158}Dy(n,p)^{158g}Tb$ and $^{191}Ir(n,\gamma)^{192m2}Ir$ using the estimated upper limits for the cross-sections provided by Prof. Vonach.

e) All participants will work to complete the ongoing measurements prior to the next CRP meeting*.

f) Dr. Qaim and Prof. Csikai will check whether they can do an additional measurement of the $^{94}Mo(n,p)^{94}Nb$ cross-section. Prof. Vonach will ask Dr. Ikeda, Japan, whether he could also measure this cross-section.

(4) As the measurements are to a large extent restricted to a neutron energy of 14 MeV, they should be supplemented by model calculations in all cases in order to get complete excitation functions. Prof. Yamamuro, Japan, has agreed to perform at least a part of these calculations and Prof. Vonach will send him a summary of the existing experimental data for this purpose. In addition all participants are asked to look for cooperation with theorists on this matter.

(5) An amended additional list of neutron-induced reactions important for the nuclear waste problem in fusion reactors (Attachment 1) was presented by Dr. Cheng. The list was discussed in some detail and it was concluded that a large part of the needed information can be obtained from existing measurements. For those cases where the data situation could not be clarified during the meeting, the following procedure was agreed upon:

a) The (n,t) reactions will be reviewed by Dr. Qaim who will send estimates of these cross-sections to Dr. Cheng. These will probably be sufficient for the waste problem.

b) For the remaining cases the data situation will be checked by Dr. Cheng. If new measurements for a few of these reactions are really needed, Dr. Cheng will transmit a request to Prof. Vonach and Dr. Wang DaHai who will try to initiate these measurements within the CRP.

(6) The present knowledge of decay data, especially half-lives for many of the long-lived nuclides is still insufficient and is probably the largest source of uncertainty for a number of the needed cross-sections (see e.g. discussion on the $^{109}Ag(n,2n)^{108m}Ag$ cross-section). The participants would like to bring this fact to the attention of the IAEA and suggest that the IAEA Nuclear Data Section should recommend a decay data table, so that everyone in the CRP should use the same decay data in the analysis of the data.

(7) It is proposed that the next CRP meeting be held in May 1991 in conjunction with the Jülich Conference on Nuclear Data for Science and Technology.

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* The completion of the $^{63}Cu(n,p)^{63}Ni$ measurements at Argonne may, however, only be possible, if additional funding can be found for the necessary liquid scintillation measurements.

Table 1: Data Status of activation cross-sections selected for the CRP on long-lived nuclides for fusion technology.

Reaction	Status	Laboratory	Neutron Energy (MeV)	Cross-Section (mb)
$^{27}Al(n,2n)^{26}Al$ several measurements available from outside CRP, data needs satisfied, evaluation still needs to be done				
$^{63}Cu(n,p)^{63}Ni$	2)	KFA Jülich	7.6	
	2)	ANL/LANL/JAERI	10,14 and D-Be n-source	
		ANL	14.9	54 ± 4*
$^{94}Mo(n,p)^{94}Nb$		ANL	14.8	53.1 ± 5.3*
$^{109}Ag(n,2n)^{108m}Ag$	1)	IAE Beijing	14.77	230 ± 7
	1)	Debrecen	14.50	263 ± 20
	3)	IRK, Vienna	14 - 15	665 ± 73
	2)	ANL/LANL/JAERI	10,14 and D-Be n-source	
	1)	KRI Leningrad	14.9	208 ± 37
$^{179}Hf(n,2n)^{178m2}Hf$	2)	Harwell	14.8	5.9 ± 0.6**
	2)	IAE Beijing	4.2 - 14.8	
	4)	Oxford/LANL	14	2.9
	2)	ANL/LANL/JAERI	10,14 and D-Be n-source	
$^{182}W(n,na')^{178m2}Hf$	2)	Harwell	14.8	
$^{151}Eu(n,2n)^{150m}Eu$	1)	IAE Beijing	14.77	1219 ± 28
	3)	IRK, Vienna	14 - 15	1325 ± 94
	1)	KFA Jülich	9.6 - 10.6	
	2)	ANL/LANL/JAERI	10,14 and D-Be n-source	
	1)	KRI Leningrad	14.9	1090 ± 84
$^{153}Eu(n,2n)^{152g}Eu$	2)	ANL/LANL/JAERI	10,14 and D-Be n-source	
	1)	IAE Beijing	14.77	1544 ± 42
	3)	IRK, Vienna	14 - 15	1442 ± 60
	1)	KRI Leningrad	14.9	1740 ± 145

$^{159}Tb(n,2n)^{158g}Tb$	2)	KFA Jülich	8.6 - 10.6		
	1)	IAE Beijing	14.77	1968	± 56
	2)	ANL/LANL/JAERI	10,14 and D-Be n-source		
	3)	IRK, Vienna	14-15	1930	± 49
$^{158}Dy(n,p)^{158g}Tb$	no work done				
$^{193}Ir(n,2n)^{192m2}Ir$	3)	IRK, Vienna	14 - 15	184	± 44
$^{187}Re(n,2n)^{196m}Re$	3)	IRK, Vienna	14 - 15	591	± 122
$^{62}Ni(n,\gamma)^{63}Ni$	no work done				
$^{98}Mo(n,\gamma)^{99}Mo$	research contract given to Sichuan Univ.				
$^{165}Ho(n,\gamma)^{166m}Ho$	"				
$^{191}Ir(n,\gamma)^{192m2}Ir$	no work done				

1) measurement performed for CRP
2) measurement in progress for CRP
3) evaluation of existing data performed for CRP
4) calculation performed for CRP

* measurement performed outside of CRP by L. Greenwood, ANL
** preliminary value

ATTACHMENT 1

ACTIVATION CROSS SECTIONS NEEDED FOR WASTE DISPOSAL ASSESSMENT OF FUSION REACTOR MATERIALS

Cross Section	Half-Life of Activated Radionuclide (Years)	Remarks[a]
Ag109(n,2n)Ag108m	127	√
Al27(n,2n)Al26	7.2×10^5	√
Bi209(n,2n)Bi208	3.68×10^5	-
Bi209(n,γ)Bi210m	3.0×10^6	-
Ca42(n,α)Ar39	269	-
Ca43(n,nα)Ar39	269	-
Cd108(n,p)Ag108m	127	-
Cu63(n,p)Ni63	100	√
Cu65(n,t)Ni63	100	-
Dy158(n,p)Tb158	150	√
Er166(n,p)Ho166m	1.2×10^3	-
Er151(n,γ)Eu152	13.3	-
Eu151(n,2n)Eu150m	35.8	√
Eu153(n,2n)Eu152	13.3	√
Hf177(n,γ)Hf178m2	31	-
Hf179(n,2n)Hf178m2	31	√
Ho165(n,γ)Ho166m	1.2×10^3	-
Ir191(n,γ)Ir192m2	241	-
Ir193(n,2n)Ir192m2	241	√
K39(n,p)Ar39	269	-
K39(n,α)Cl36	3.0×10^5	-
Mo98(n,γ)Mo99→Tc99	2.1×10^5	√
Mo100(n,2n)Mo99→Tc99	2.1×10^5	√
Mo94(n,p)Nb94	2.0×10^4	√
Mo95(n,d)Nb94	2.0×10^4	-

(a) "√" mark indicates that this cross section has been measured, or is being measured by the participants of the IAEA/NDS Coordinated Research Program.

ACTIVATION CROSS SECTIONS NEEDED FOR WASTE DISPOSAL ASSESSMENT OF FUSION REACTOR MATERIALS

(Continued)

Cross Section	Half-Life of Activated Radionuclide (Years)	Remarks[a]
Mo96(n,t)Nb94	2.0×10^4	-
N14(n,p)C14	5730	√
Nb93(n,γ)Nb94	2.0×10^4	-
Nb93(n,2n)Nb92	3.5×10^7	√
Ni58(n,γ)Ni59	7.5×10^4	-
Ni60(n,2n)Ni59	7.5×10^4	√
Ni62(n,He3)Fe60	1.49×10^6	-
Ni64(n,nα)Fe60	1.49×10^6	-
Pt192(n,p)Ir192m2	241	-
Re187(n,2n)Re186m	2.0×10^5	√
Re185(n,γ)Re186m	2.0×10^5	-
Ta180(n,t)Hf178m2	31	-
Tb159(n,2n)Tb158	150	-
Tm169(n,α)Ho166m	1.2×10^3	-
W182(n,nα)Hf178m2	31	√

(a) "√" mark indicates that this cross section has been measured, or is being measured by the participants of the IAEA/NDS Coordination Research Program.

MEASUREMENT OF THE CROSS SECTIONS FOR THE REACTIONS $^{52}Cr(n,2n)^{51}Cr$, $^{66}Zn(n,2n)^{65}Zn$, $^{89}Y(n,2n)^{88}Y$ AND $^{96}Zr(n,2n)^{95}Zr$ FROM 13.5 TO 14.8 MeV

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ABSTRACT Cross sections for the reactions $^{52}Cr(n,2n)^{51}Cr$ $^{66}Zn(n,2n)^{65}Zn$, $^{89}Y(n,2n)^{88}Y$ and $^{96}Zr(n,2n)^{95}Zr$ were measured in the energy range 13.47 MeV to 14.79 MeV applying the activation technique. Energy of neutrons produced via the $T(d,n)^4He$ reaction was changed by the emission angle. The neutron fluences and energies incident on the samples were determined by the measurements of the ^{92m}Nb and ^{89}Zr specific activities produced in the $^{93}Nb(n,2n)$ and $^{90}Zr(n,2n)$ reactions. The induced γ-ray activities of the irradiated Cr, Zn, Zr and Y_2O_3 samples and their monitor foils were measured by means of calibrated Ge(Li) and NaI well-type γ-ray detectors. The results compared to the corresponding data in the literature show that the uncertainties obtained in this work are considerably smaller in most cases than those given by other authors.

1. INTRODUCTION

Measurements of activation cross sections for production of long-lived isotopes at around 14 MeV neutron energy are of interest for testing nuclear reaction models. Furthermore, the data in the case of structural

materials of a fusion reactor are important for the estimation of neutron multiplication, nuclear heating, nuclear transmutation and radiation damage effects. Cross section data available for such reactions are very scarce and contradictory even in the vicinity of 14 MeV especially in the case of $^{52}Cr(n,2n)$ and $^{96}Zr(n,2n)$ reactions. This work describes precise activation cross section measurements for (n,2n) reactions on ^{52}Cr, ^{66}Zn, ^{89}Y and ^{96}Zr in the 13.5 to 14.8 MeV range.

2. EXPERIMENTAL PROCEDURE

Rectangular high-purity metallic samples of natural Cr, Zn and Zr, with the dimensions 16 mm x 8 mm and thicknesses of 0.75 mm, 1.0 mm and 1.0 mm, respectively, as well as Y_2O_3 powder samples were irradiated at the Cockcroft-Walton neutron generator of the Institute of Experimental Physics, Kossuth University (KFI), Debrecen. The Y_2O_3 powder with 2.0 mm effective layer thickness was contained in thin-walled (0.5 mm) cylindrical perspex containers with an inner diameter of 14.0 mm. The neutrons in the 14 MeV range were produced via the reaction $T(d,n)^4He$, using an analyzed d^+-beam with (190±10) keV mean incident energy. The total neutron yield achieved with an air-jet-cooled 0.5 mm thick Al-backed Ti-T target was $\approx 10^{14}$ neutrons in approximately 275 hours. The scattering free arrangement used for the irradiation of the samples has been described elsewhere[1].

For fluence monitoring metallic Nb foils, 0.65 mm thick, with the same shape as the samples were placed back-to-back behind each sample; the Y_2O_3 samples positioned at 0^o, 55^o and 135^o relative to the incident deuteron beam were sandwiched between two fluence monitor foils. Thus all cross sections were measured relative to the well-evaluated cross sections of the reference reaction $^{93}Nb(n,2n)^{92m}Nb$ in this energy range[2,3]. The distribution of the neutron production in time was monitored by means of a BF_3 long-counter. The neutron-energy scale was verified by measuring the ratio of the ^{89}Zr to ^{92m}Nb specific activities induced in Zr and Nb foils, which were exposed as a sandwich at 12.5^o

and 97.5°. The measured activity ratios were compared to expected ones based on accurate cross section data from Palvik et al.[4] for the $^{90}Zr(n,2n)^{89}Zr$ reaction and on the above mentioned $^{93}Nb(n,2n)^{92m}Nb$ cross section data. Another very sensitive check of the energy scale was the $^{52}Cr(n,2n)^{51}Cr$ excitation function measured in the course of this work.

Neutron energy profiles were calculated for each sample using the Monte Carlo simulation code PROFIL[5], and the full width at half maximum (FWHM) and the average energy were determined. Examples of energy profiles are shown in Fig. 1.

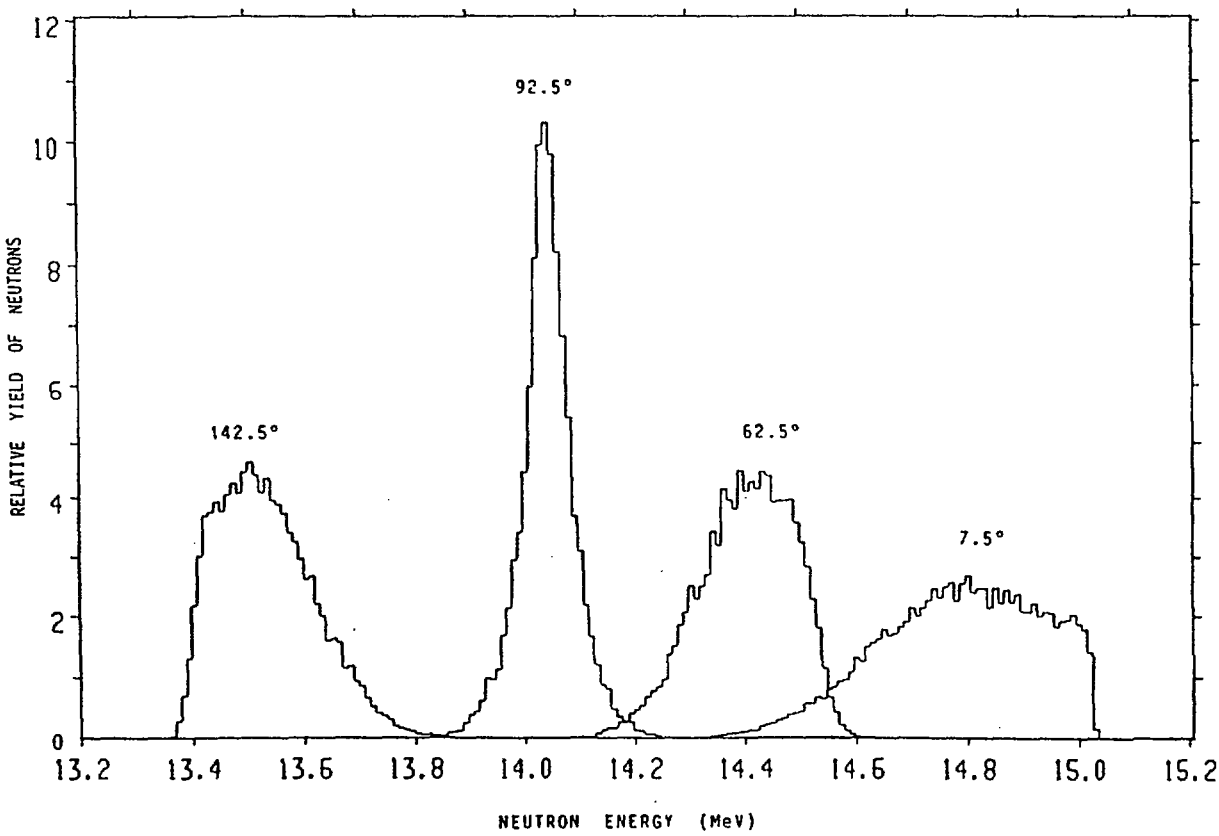

Fig.1. Calculated neutron energy distribution profiles for the different irradiation angles used in this experiment for the chromium samples. All distributions are normalized to equal areas

The activities of the Zn, Zr and Y_2O_3 samples and of the corresponding Nb fluence monitor foils were measured with a Ge(Li) γ-ray detector at the KFI, Debrecen, evaluating the relevant full-energy peak areas and taking into account γ-ray self-attenuation in the samples. The relative efficiency of the Ge(Li)detector has been determined in the

Table 1: Relevant decay data of the product nuclei.

Radio-nuclide	Abundance of the target isotope (%)	Half-life (days)	Ref.	Energies of the principle γ-rays emitted (keV)	Emission probabilities	Ref.
^{51}Cr	83.789±0.012	27.704±0.004	Tuli (1987)	320.08	0.0985±0.0009	Lorenz (1987)
^{65}Zn	27.9 ±0.2	243.9 ±0·1	"	1115.55	0.5065±0.0020	"
^{88}Y	100	106.64±0.08	"	898.04 1836.06 2734.09	0.924 ±0.004 0.9930±0.0005 0.0072±0.0007	"
^{92m}Nb	100	10.15±0.02	"	912.6 934.44 1847.54	0.0178±0.0010 0.9892±0.0010 0.0083±0.0004	Luksch (1980)
^{95}Zr	2.80 ±0.01	64.02±0.04	"	235.69 724.20 756.73	0.0029±0.0005 0.4415±0.0020 0.5450±0.0020	Lorenz (1987)
^{95}Nb		34.97±0.03		765.81	0.9980±0.0002	"

186-2448 KeV energy range by placing a ^{226}Ra source of 19 mm in diameter in different positions to the detector[6]. The absolute efficiency has been determined by ^{203}Hg, ^{137}Cs, ^{60}Co and ^{88}Y standard gamma-ray sources at 60 mm distance from the surface of the Ge(Li) detector. An empirical analytical expression was given for the description of the energy-efficiency curve for three different positions of the sources. The total error of the full-energy-peak efficiency in the 180-1500 keV energy range was found to be 1.0 %.

The relative activities of the Cr samples and their monitor foils as well as those of the Zn and Zr samples were determined with a 15 %-efficiency (relative to a 7.62 cm x 7.62 cm NaI(Tl) crystal) intrinsic Ge γ-ray detector at the IRK, Vienna. For normalization purposes, absolute activity measurements were performed at the IRK on the Cr, Zr and Zn sample with the highest activity, and on some Nb foils, employing a 12.7 cm x 12.7 cm NaI(Tl) well-type detector counting above an energy discrimination level of 22.1 keV[7]. Its efficiency for the radiation of the product nuclei ^{51}Cr, ^{65}Zn, ^{88}Y, ^{92m}Nb and ^{95}Zr-^{95}Nb was determined according to the characteristics of the respective decay schemes, taking into account the self-attenuation and the Compton scattering of the γ-rays in the samples[8]. In the case of ^{92m}Nb the fractional peak area for K-shell X-rays above a discrimination level of 22.1 keV was accounted for[9]. The activities of the Y_2O_3 samples were also measured at the IRK by integral γ-ray counting since higher accuracy and precision of the results could be achieved as compared to the γ-ray measurements in Debrecen using a Ge(Li) detector. The decay characteristics of the product nuclei summarized in Table 1. were taken from the Nuclear Data Sheets and from Tuli[10] and Lorenz[11]. The absolute activity measurements on the mother-daughter pair ^{95}Zr-^{95}Nb required to wait for the decay of ^{89}Zr and were commenced 57 days after the end of the irradiation, approximately at the time of the optimal signal-to-

Table 2: Results of the measurements of (n,2n) cross sections

Reaction	Position of the sample relative to the incident d^+-beam	Average neutron energy (MeV)	Width of the energy distribution (1/2 FWHM) (MeV) [1]	Cross section (mb)	
				IRK	KFI
$^{52}Cr(n,2n)^{51}Cr$	(7.5±0.5)°	14.783±0.013	0.210	400.6±6.8	
	(62.5±0.5)°	14.397±0.011	0.105	339.3±5.2	
	(92.5±0.5)°	14.040±0.010	0.035	270.5±4.8 [2]	
	(142.5±0.5)°	13.528±0.010	0.095	172.9±3.6	
$^{66}Zn(n,2n)^{65}Zn$	(17.5±0.5)°	14.754±0.013	0.190	734.5±11.8	738.1±16.8
	(72.5±0.5)°	14.283±0.011	0.080	652.0±11.9	663.4±15.4
	(102.5±0.5)°	13.922±0.010	0.030	590.6±10.3 [2]	589.8±16.1
	(152.5±0.5)°	13.466±0.010	0.115	503.9± 9.5	495.5±11.2
$^{89}Y(n,2n)^{88}Y$	(0.0±0.5)°	14.789±0.014	0.210	1009.8±14.3	1015.8±21.8
	(55.0±0.5)°	14.476±0.012	0.120	945.8±14.2	917.9±19.9
	(85.0±0.5)°	14.132±0.010	0.054	872.6±12.0 [2]	847.1±25.4
	(135.0±0.5)°	13.586±0.010	0.092	703.6±10.8	686.4±15.1
$^{96}Zr(n,2n)^{95}Zr$ + $^{96}Zr[(n,d)+(n,np)+(n,pn)]^{95}Y \rightarrow ^{95}Zr$ + 6.207* $^{94}Zr(n,\gamma)^{95}Zr$ [3]	(12.5±0.5)°	14.771±0.013	0.185	1506 ±22	1512.2±38.6
	(67.5±0.5)°	14.341±0.011	0.090	1497 ±26	1480.0±37.7
	(97.5±0.5)°	13.981±0.010	0.030	1489 ±23 [2]	1483.3±43.5
	(147.5±0.5)°	13.495±0.010	0.115	1477 ±26	1436.6±39.4

1) FWHM = full width at half maximum

2) Weighted average of the results obtained for the two samples positioned symmetrically to the incident d^+-beam

3) See text.

background ratio. In fact, when measuring the sum of the ^{95}Zr and ^{95}Nb activities produced by 14 MeV neutrons hitting a Zr sample, one measures the sum of the following cross sections:

a) for the $^{96}Zr(n,2n)^{95}Zr$ reaction, which will provide by far the main contribution;

b) for the $^{96}Zr((n,d)+(n,np)+(n,pn))^{95}Y$ reaction, since the radionuclide ^{95}Y decays to ^{95}Zr via β^- emission with a relatively short half-life (≈10.3 min);

c) for the $^{94}Zr(n,\gamma)^{95}Zr$ reaction, weighted by a factor 6.207, which is the ratio of the isotopic abundances of ^{94}Zr to ^{96}Zr in natural Zr.

3. RESULTS, UNCERTAINTIES AND DISCUSSION

The results for the (n,2n) cross sections of the investigated nuclides in the energy range from 13.47 MeV to 14.79 MeV are listed in Table 2, together with the average neutron energies and its uncertainties, and the spread of the energy distributions. As it can be seen in Table 2 cross section data measured in Vienna and Debrecen by two independent methods are in good agreement. The total uncertainty for each cross section value was obtained by adding the uncertainty components in quadrature. Figures 2 through 5 display the results of our work together with those taken from the literature. For the reason of better legibility and demonstration the cross section data obtained for ^{89}Y were split into two parts: the first comprises work performed from 1959 to 1975, the second the more recent results. All data given in the literature were normalised to the latest values of the cross sections for the fluence monitor reactions employed and of the decay data, especially of the intensity of the ^{51}Cr γ-radiation. In general the results agree with those given by a number of other authors, but the uncertainties obtained in this work are considerably smaller in most cases. For $^{66}Zn(n,2n)^{65}Zn$

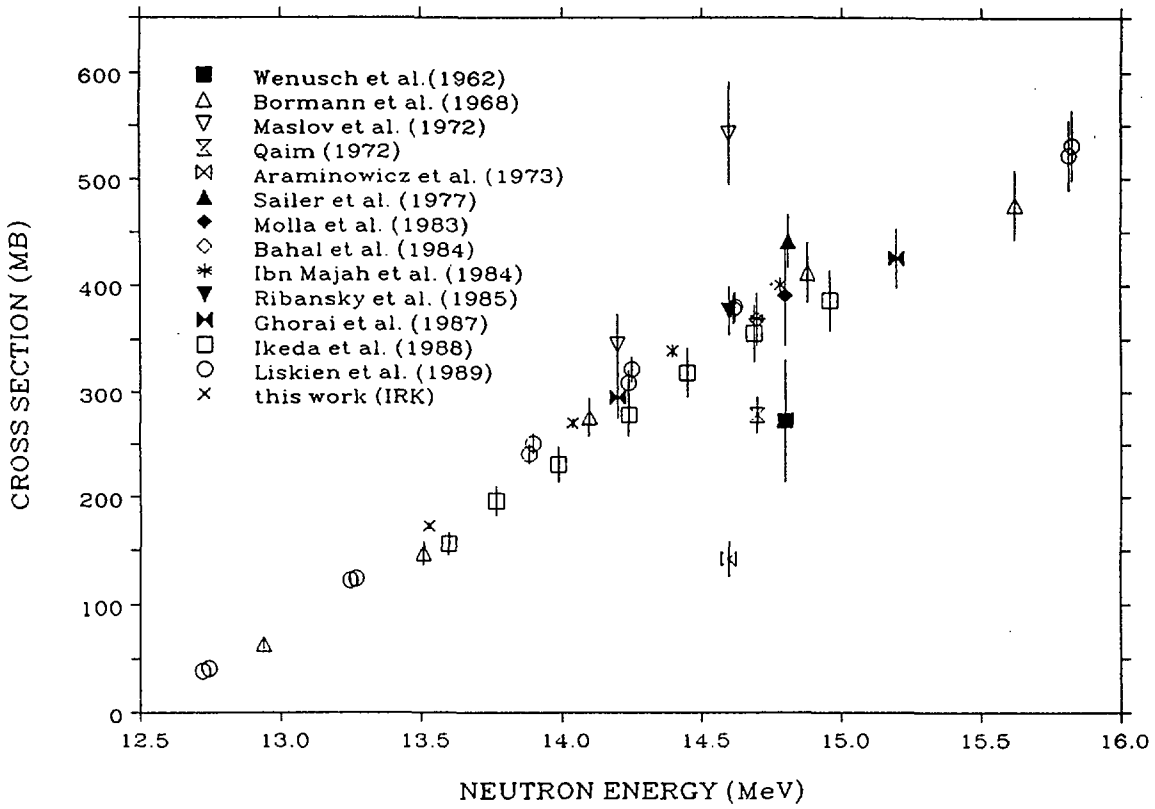

Fig.2. The cross sections for the reaction $^{52}Cr(n,2n)^{51}Cr$ resultant from this work as compared to data from the litereature

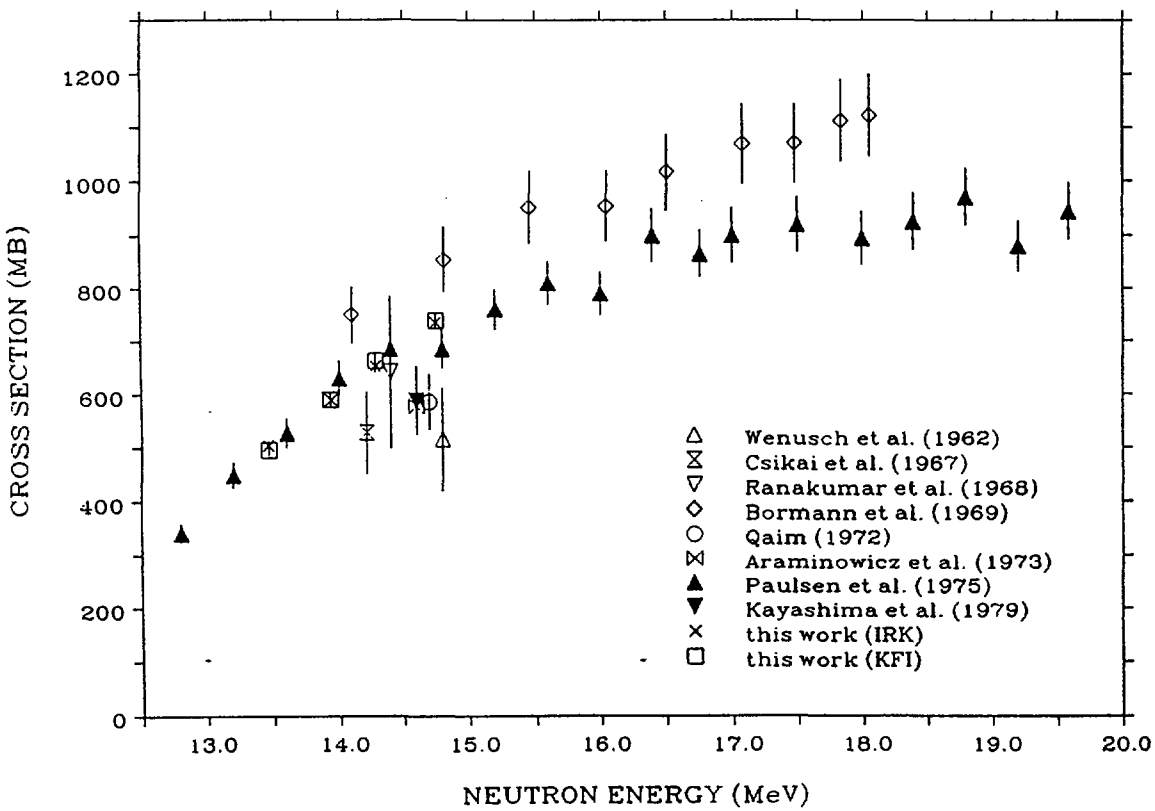

Fig.3. The cross sections for reaction $^{66}Zn(n,2n)^{65}Zn$ resultant from this work as compared to data from the literature

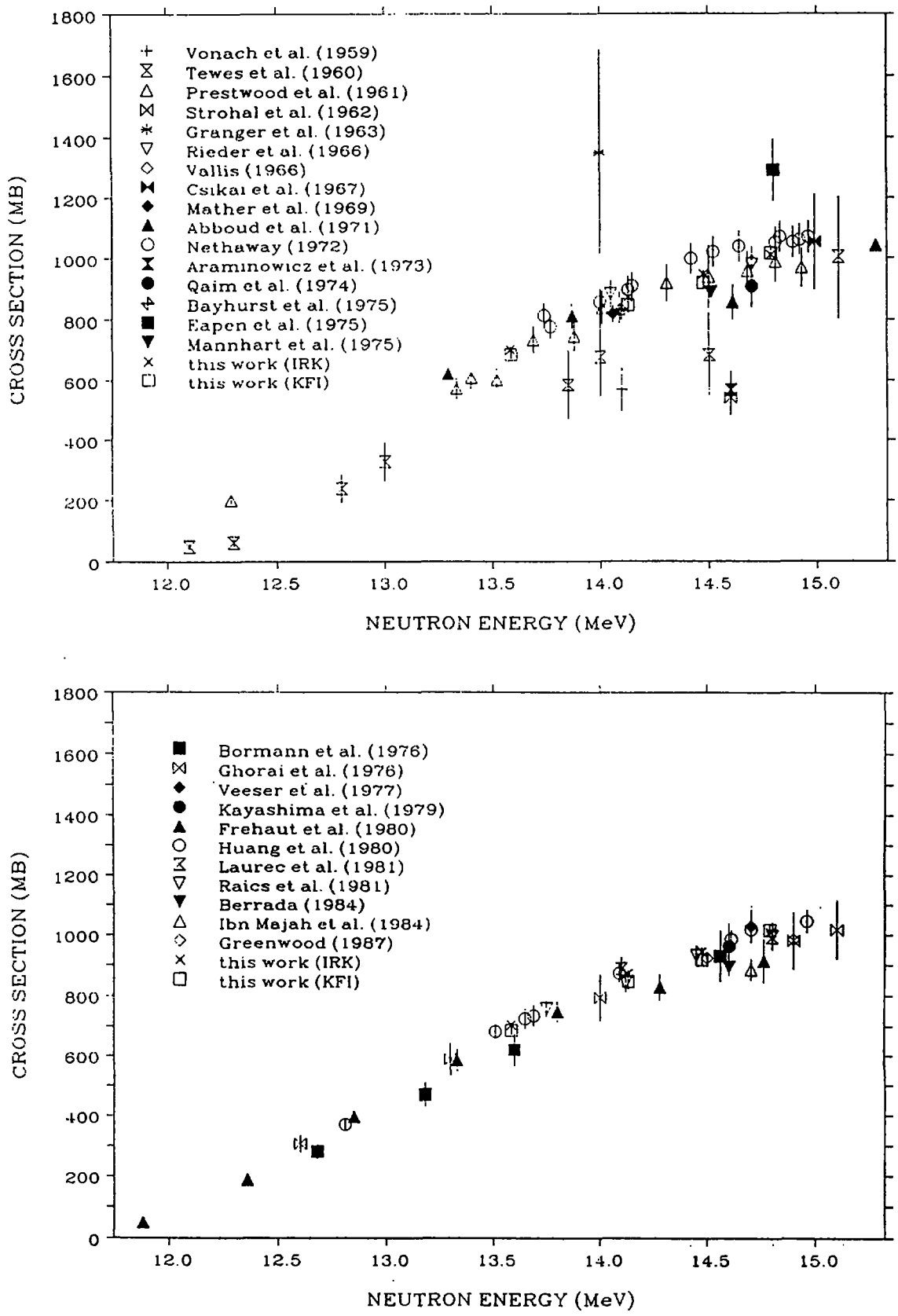

Fig.4. The cross sections for the reaction $^{89}Y(n,2n)^{88}Y$ resultant from this work as compared to data from the literature: published from 1959 to 1975 (upper) and 1976 to 1988 (lower).

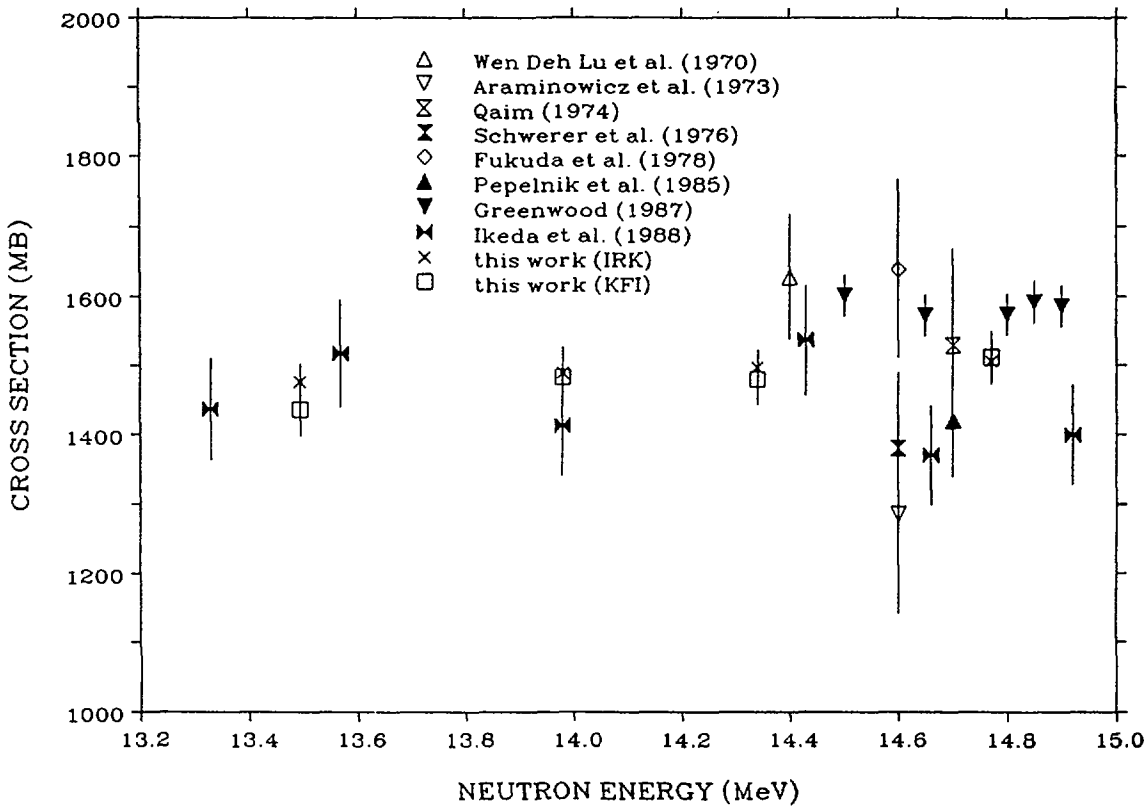

Fig.5. The cross sections for the reaction $^{96}Zr(n,2n)^{95}Zr$ resultant from this work as compared to data from the literature. (Note the suppressed zero!)

our results definitely confirm the measurements of Paulsen[12] and suggests that this work should be used for the whole excitation function rather than the measurements of Bormann and Lammers[13]. For $^{89}Y(n,2n)^{88}Y$ there is now excellent agreement between all recent measurements and a thorough cross section evaluation is now needed more than further measurements. For the $^{96}Zr(n,2n)^{95}Zr$ reaction our cross sections are somewhat lower than the recent results from Greenwood[14], the discrepancy being somewhat larger than the combined uncertainties of both experiments, our results are however in better agreement with the systematic trend of these cross sections with mass number.

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RESEARCH OF ACTIVATION CROSS SECTIONS FOR LONG-LIVED RADIONUCLIDES ON ELEMENTS OF CU MO AG EU AND TB

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ABSTRACT: The cross sections for $^{109}Ag(n,2n)^{108m}Ag$, $^{151}Eu(n,2n)^{150m}Eu$, $^{153}Eu(n.2n)^{152g}Eu$ and $^{159}Tb(n,2n)^{158}Tb$ reactions have been measured by the activation method at 14 MeV. The results were compared with existing data (1-4) and calculations of systematic (5,6) and Code HFTT (7) which was based on the compound nucleus evaporation model and the preequilibrium exciton model.

1. INTRODUCTION

The nuclear data requirements for integral calculations for fist wall, blanket , shielding and activating problems of fusion reactor have been listed by E.T.Cheng and dopted by working group 1 of 16th INDC Meeting as high priority data requests for fusion technology. Especially,the products of long-lived radionuclides are important for waste disposal and maintenance of fusion reactors.

Up to now, only few reports for (n,2n) reaction cross section values in this area were found at 14 MeV. In addition, cross sections for ^{109}Ag $(n,2n)^{108m}Ag$ reaction have not been measured before. Investigetions were initiated on the (n,2n) reactions for Ag,Eu and Tb. The cross sections for $^{109}Ag(n,2n)^{108m}Ag$, $^{151}Eu(n,2n)^{150m}Eu$, $^{153}Eu(n,2n)^{152g}Eu$ and $^{159}Tb(n,2n)^{158}Tb$ reactions have been performed by activation technique at 14 MeV. The calculations of systematic and theoretic also made for Cu, Mo (n,p) reaction and Ag, Eu and Tb (n,2n) reaction.

2. EXPERIMENTAL PROCEDURE

2.1 Irradiation

Irradiations were carried out by neutron source of $T(d,n)^4He$ reaction at Intense Neutron Generator of Lanzhou University (INGL) and Cockcroft-Walton at Institute of Atomic Energy Beijing (IAEB). The irradiation geometry on the INGL, the sample groups were placed at 15°, 30°, 50° and 80° angles relative to beam direction and centered at beam stop on the T-Ti target with distances 5

and 10 cm respectively. The irradiation was lasted to 7.36 hours with intensity of neutron flux about $1.1\text{-}3.3\times10^{12}$n/s 4π. One sample group was sent back to IAEB for analysis and three sample groups were counted in Lazhou University. The efficiencies of two Ge(Li) detecters used in both places were calibrated using the same group of gamma-ray standard sources. The irradiation in IAEB, the stacked sample groups with cadmium cover were placed at 0° and 65° angles with effective distances from 12 to 76 mm. The irradiations were lasted to 50 hours at neutron flux densities $(0.7\text{-}2)\times10^{10}$n/s 4π. The energy of deutron beam was taken 300keV.

2.2 Sample

The samples of 20mm diameter were made by natural plates for Ag, Nb or oxide powder for Eu and Tb. The purity of them were better than 99%. The samples in question were sandwiched between two niobium samples with thickness 0.03mm to evaluate the neutron fluence on the samples. The cross sections of $^{93}Nb(n,2n)^{92m}Nb$ reaction were select as a monitor due to it's high accurate and same shape of excitation functions with reaction investigeted.

In the case of Eu, the sample was wrapped in 0.5mm thickness Cd foils to reduce the activities of ^{152g}Eu from $^{151}Eu(n,r)^{152g}Eu$ reaction of thermal neutron.

2.3 Measurement of activity

After irradiation, the samples were cooled for 2-6 months, then to analyse each foils with a 136cc Ge(Li) detecter. The distance of sample to surface of the detecter was 15.5 cm to reduce the effect of sum peak. The measurements were lasted to 1-6 days for each samples, and repeated for two or thrr times. The activities of Nb monitor foils were mesured in a week after irradiations.

2.3 Correction

In general, the corrections for (n,2n) reactions are very small due to it's high threshold. In principle, the same activity was formed by the (n,2n) reaction and (n,r) reaction on an isotope two mass units less then that of investegated nuclide is small and may be neglected. However, in the case of $^{153}Eu(n,2n)^{152g}Eu$ reaction, there were very high (n,r) cross section for the lower energy neutrons it was necessary to determine the contribution of scattered neutrons. The special experimental arrangement was made for measuring the $^{153}Eu(n,2n)^{152g}Eu$ reaction. The relation of activity as a function of distance square was used to determine the contributionof scattered neutrons in the vicinity of target and sample groups. The results of the measurements are shown in the Fig.1. The least square method was used to get the correction of lower energy neutrons. The corrections were large especialy for heavy mass target assembly and sample groups (4-20%). An

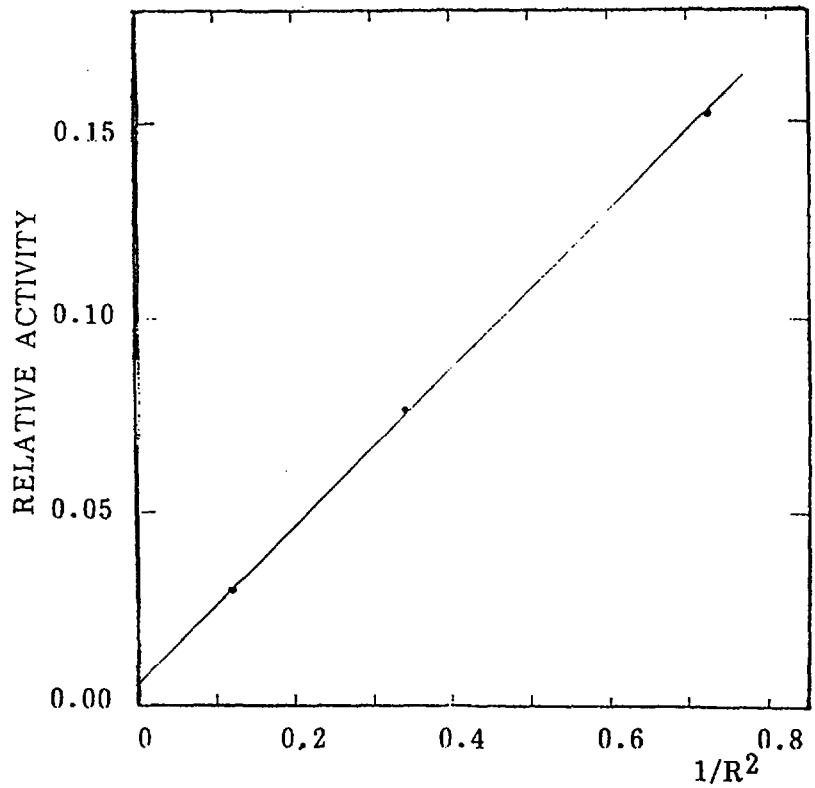

Fig. 1 Dependence 0f the activity on the unproportion of destance squre

another sample group was placed at distance of 2 metres to evaluate the background of the experimental room (about 1% at distance of 3 cm).

Some corrections were also made for the gamma-ray absorption and for scattered neutrons owing to different excitation function shapes for standard reaction and investigated reaction as well as for the effects of the gamma rays with energies close to studied one.

3. THEORETICAL AND SYSTEMATICS CALCULATION

We used the code HFTT (7) for calculating the excitation functions of the (n,2n) reaction on ^{109}Ag, ^{151}Eu, ^{153}Eu, ^{159}Tb and (n,p) reaction on ^{63}Cu, ^{94}Mo. The results are shown in Figs. 2-6. The calculation program used was based on the compound nucleous evaporation model (8) and the preequilibrium exciton model (9). The first and the second emission particles considered in the nuclear reactions were n, p, d, t, ^{3}He, ^{4}He and gamma. The third emission particles considered were n, p and gamma. In preequilibrium theory we chose initial exciton configurations of n_0= 3 (2p, 1h) for neutron induced reactions. The Gelbert-Cameron's formulas were used for calculating the energy level density of nuclei. For computation of inverse cross section of nuclear reaction the optical model was used. The optical paramenters used in the program were recommended by F.Bechetti et al.(10), Lohr (11) and Mefedden (12).

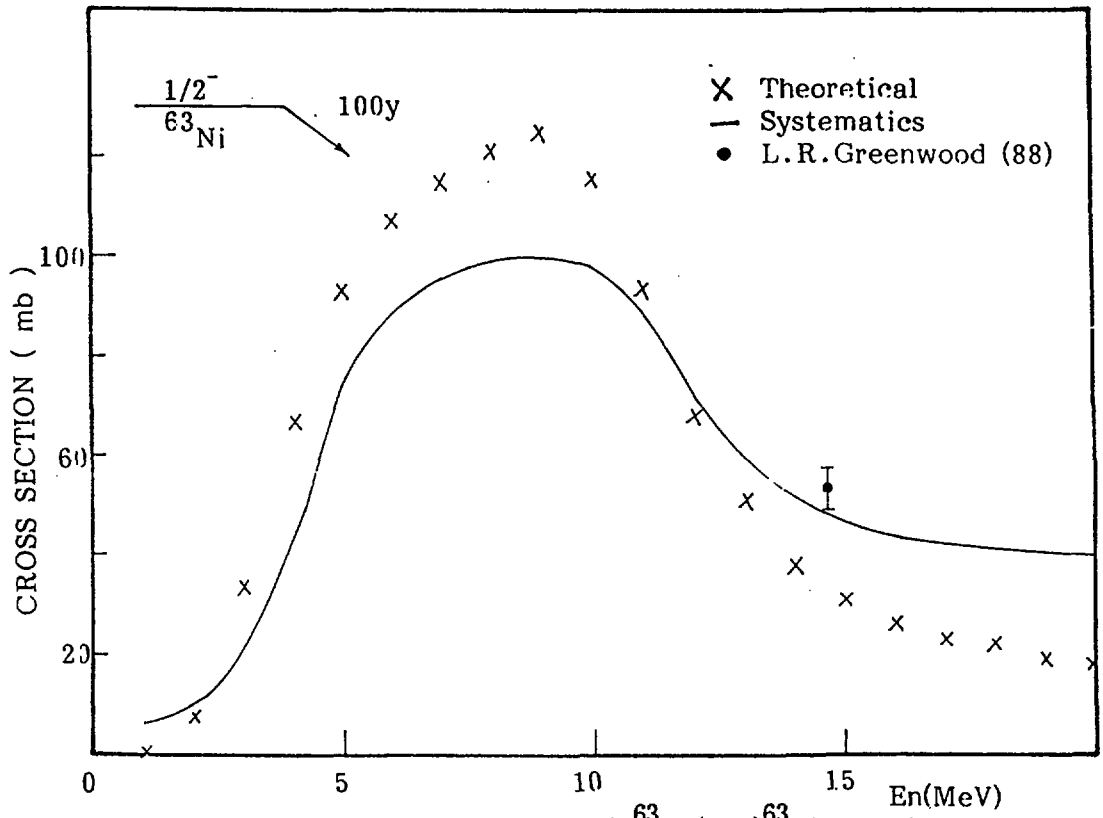

Fig. 2 Excitation function of $^{63}Cu(n,p)^{63}Ni$ reaction

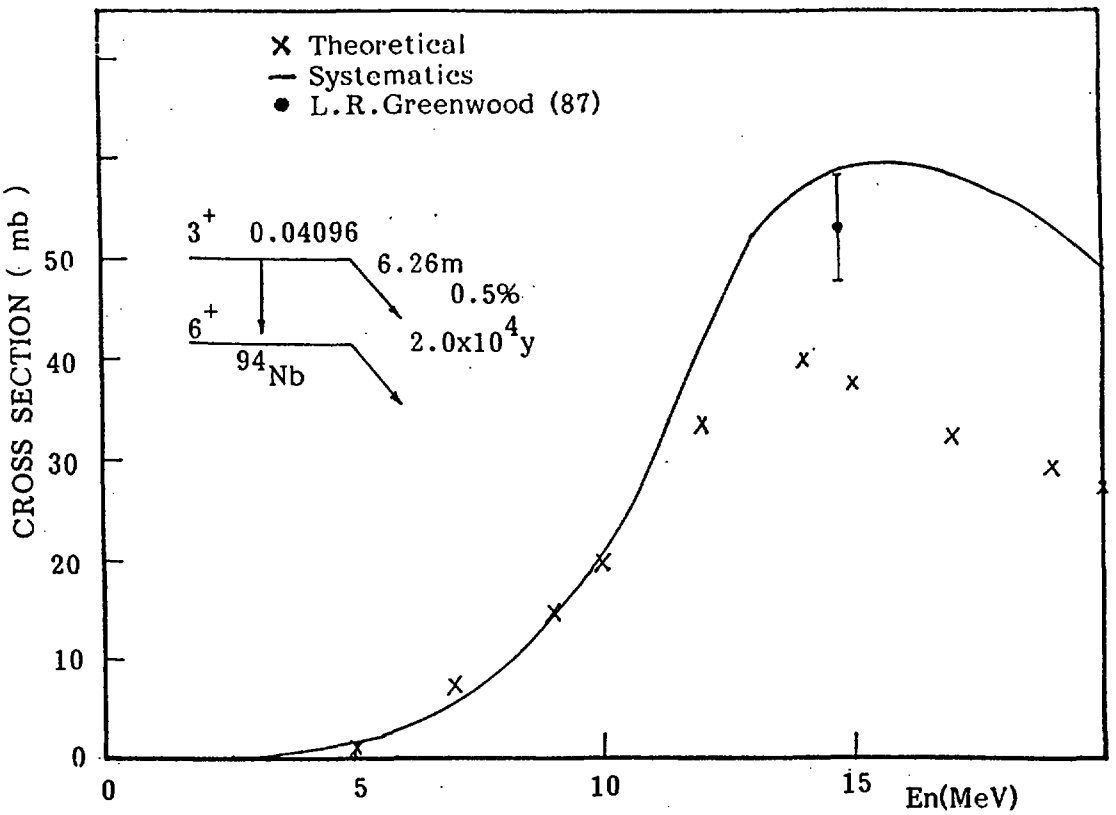

Fig. 3 Excitation function of $^{94}Mo(n,p)^{94}Nb$ reaction

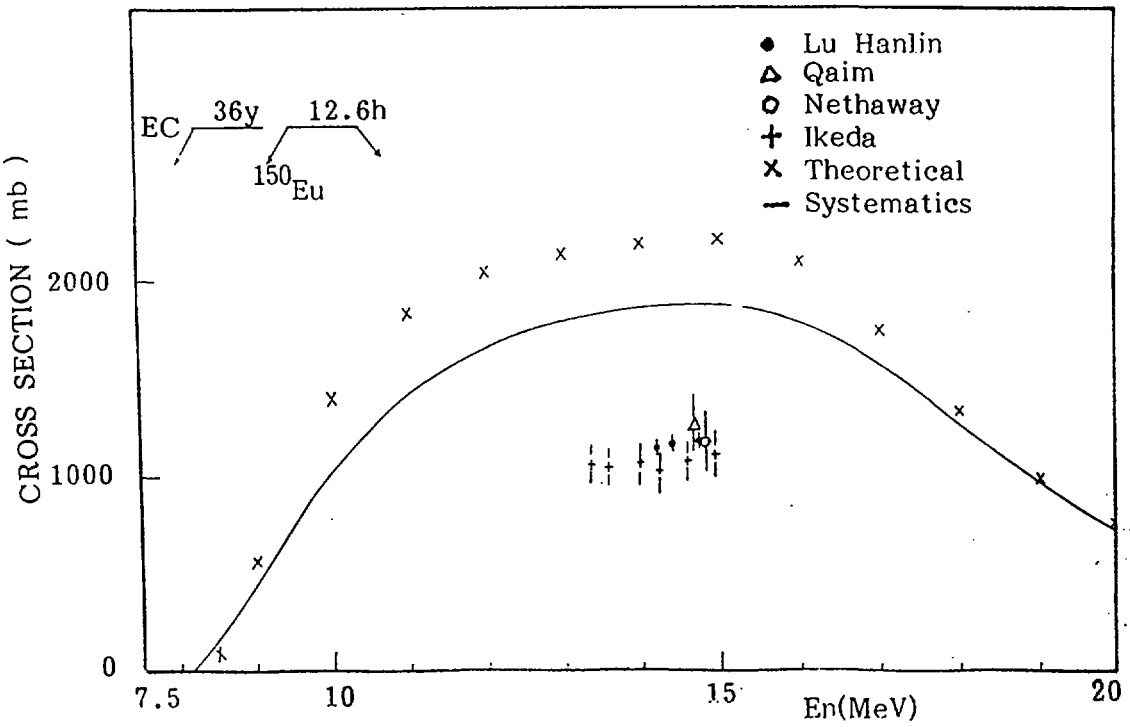

Fig. 4 Excitation function of $^{151}Eu(n,2n)^{150}Eu$ reaction

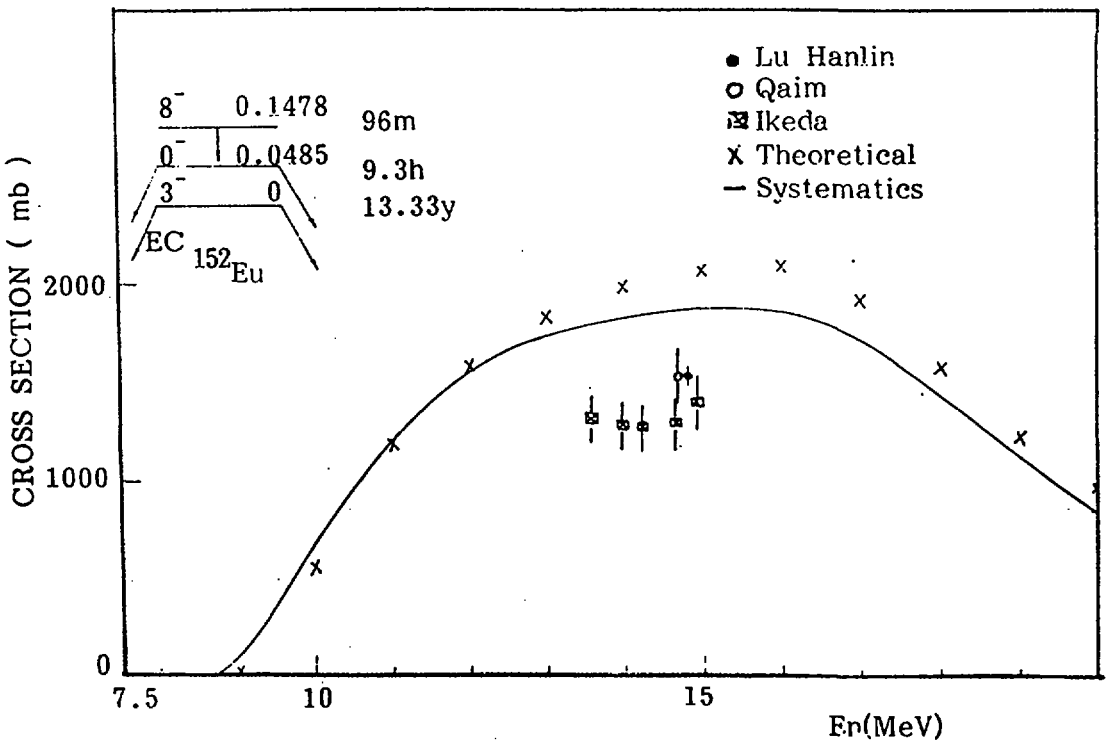

Fig. 5 Excitation function of $^{153}Eu(n,2n)^{152}Eu$ reaction

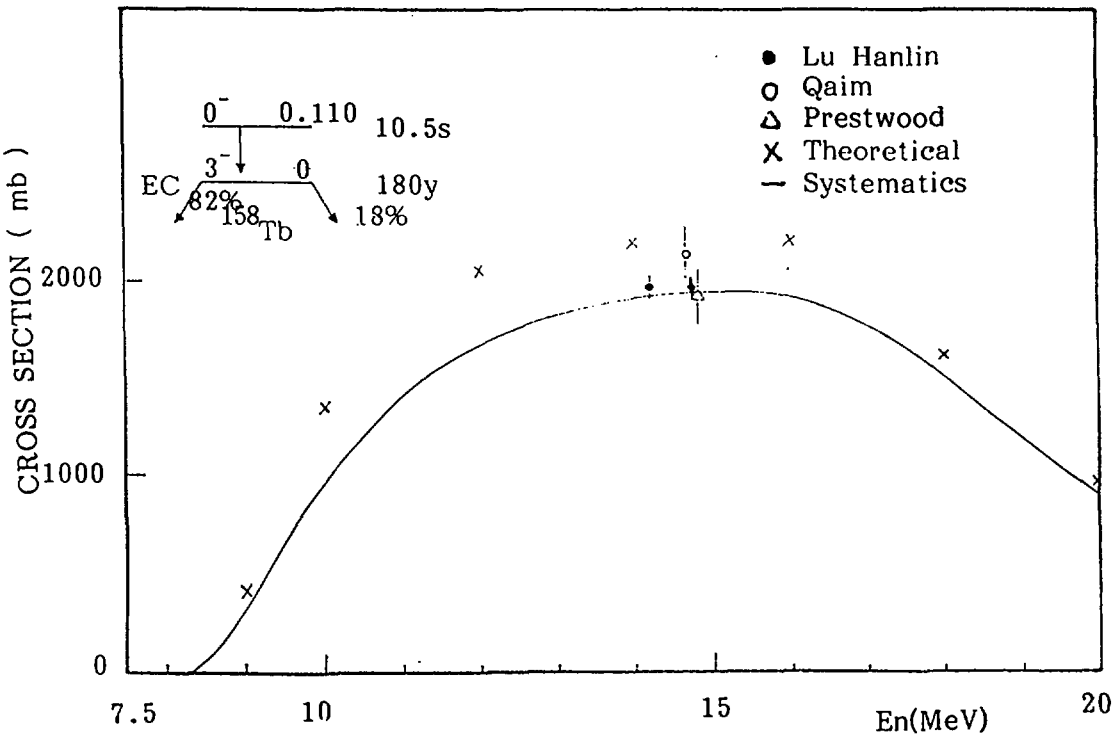

Fig. 6 Excitation function of $^{159}Tb(n,2n)^{158}Tb$ reaction

The calculations of (n,p) and (n,2n) excitation functions using the systematics model were made by Zhao Zhixiang (5) and Zhang Jin (6). The code was based on the constant temperature evaporation model in which preequilibrium emission was considered.

4. RESULTS AND UNCERTAINTIES

The measured results of the cross sections are given in Table 1 and some of them are ploted in Figs. 2-6 with existing data and calculated curves. The error correlations for the measurements of ^{109}Ag, ^{151}Eu, ^{153}Eu, ^{159}Tb and ^{93}Nb are shown in the Table 2. The principal sources of uncertainties are listed in Table 3. For $^{109}Ag(n,2n)^{108m}Ag$ reaction the experiments showed that the activity of ^{108m}Ag was very few produced by $^{107}Ag(n,r)^{108m}Ag$ reaction.

The Table 4 shows the present knowledge of cross sections for long-lived radionuclides with experimental data and semi-systimatical data, which are given through systimatical calculations and experimental data of short-lived. Semi-systimatical data agreed with experimental data except the reaction of $^{109}Ag(n,2n)^{108m}Ag$. Our data are very coincident with D.R.Nethaway and S.M.Qaim for $^{151}Eu(n,2n)^{150m}Eu$ and $^{153}Eu(n,2n)^{152g}Eu$ reactions. Consistent results are given by S.M.Qaim, R.J.Prestwood and us, if the same value of half-lift for the residual nucleus was used for them.

TABLE 1. CROSS SECTIONS AND DECAY DATA

Neutron energy (MeV)	$^{93}Nb(n,2n)^{92m}Nb$	$^{109}Ag(n,2n)^{108m}Ag$	$^{151}Eu(n,2n)^{150m}Eu$ (mb)	$^{153}Eu(n,2n)^{152g}Eu$	$^{159}Tb(n,2n)^{158}Tb$
14.19±0.23	457.5±4.5	224±6	1190±27		1980±56
14.28±0.24	458.0±4.5	215±11			
14.41±0.16	458.7±4.5		1215±36		
14.44±0.26	458.8±4.5	223±10			
14.77±0.48	458.6±5.6	230±7	1219±28	1544±42	1968±56
14.83±0.34	458.6±5.6	233±7			
$T_{\frac{1}{2}}$	10.15d	127y	35.8y	13.33y	180y
E_r (keV)	934.5	433.9	333.9	344.3	944.2
I_r (%)	99.0	90.5	94.0	26.58	43

TABLE 2. CORRELATION MATRIX (14.77 MeV)

Reaction	Error					
$^{109}Ag(n,2n)^{108m}Ag$	3.1%	1.000				
$^{151}Eu(n,2n)^{150m}Eu$	2.4%	0.551	1.000			
$^{153}Eu(n,2n)^{152g}Eu$	2.7%	0.377	0.480	1.000		
$^{159}Tb(n,2n)^{158g}Tb$	2.9%	0.599	0.435	0.370	1.000	
$^{93}Nb(n,2n)^{92m}Nb$	1.2%	0.436	0.639	0.756	0.492	1.000

TABLE 3. THE PRINCIPAL SOURCES OF UNCERTAINTY

Source of error	Uncertainty (%) ^{108m}Ag	^{150m}Eu	^{152g}Eu	^{158g}Tb
reference cross section	1.2	1.2	1.2	1.2
detector efficiency	1.5	1.5	1.5	1.5
counting statistics	1.1-2.4	0.9-2.7	0.9	0.8
gamma absorption	0.4	0.9	0.9	0.6
neutron scattering	0.5	0.5	1.3	1.8
gamma mixture		0.1	0.1	
sample mass including purity	0.1	0.35	0.35	0.5
sum peak	0--4			
Total	2.3--5.1	2.4--3.5	2.7	2.9

* Uncertainties in decay schemas are not included

Table 4. Existing data for experiment and systematic (14MeV)

Nuclear reaction	Present work (mb) exp.	semi-syst.	exp.	Reference ref.	(mb) semi-syst.	
$^{63}Cu(n,p)^{63}Ni$		50	54±4	88 (13)		
$^{94}Mo(n,p)^{94}Nb$		58	53.1±5.3	87 (14)		
$^{109}Ag(n,2n)^{108m}Ag$	230±7	585	263±20	89 (4)	665±73	89 (4)
$^{151}Eu(n,2n)^{150m}Eu$	1219±28	1400	1180±150	72 (1)	1325±34	89 (4)
			1270±149	74 (2)		
			1080±100	89 (4)		
$^{153}Eu(n,2n)^{152g}Eu$	1544±42	1429	1542±138	74 (2)	1442±60	89 (4)
			1300±130	89 (4)		
$^{159}Tb(n,2n)^{158}Tb$	1968±56	1940	*2161±140	74 (2)	1930±49	89 (4)
			1930±135	84 (3)		

* correction to value of half-life 180y

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Radiochemical Studies of Fast Neutron Induced Long-Lived Activation Products

(Research Agreement No. 5061/RO/CF)

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Aims

The aims of the research agreement were defined as:

- Measurement of $^{63}Cu(n,p)^{63}Ni$, $^{151}Eu(n,2n)^{150}Eu$ and $^{159}Tb(n,2n)^{158}Tb$ reaction cross sections in the neutron energy range from threshold to 10.6 MeV
- Hauser-Feshbach calculations

Status Report

Experimental investigations on the three processes mentioned have been under way for over a year. Irradiations have been done with quasi-monoenergetic neutrons produced via the $^{2}H(d,n)^{3}He$ reaction in a D_2 gas target at our variable energy compact cyclotron CV28. The neutron flux densities have been determined via the $^{27}Al(n,\alpha)^{24}Na$ monitor reaction.

In the case of the $^{63}Cu(n,p)^{63}Ni$ reaction a 60 g compact piece of Cu was irradiated with 7.5 MeV neutrons for 25 h. A chemical separation of the weak β^- emitting ^{63}Ni ($T_{\frac{1}{2}}$ = 100 y; β^- = 100 %; E_{β^-} = 66 keV) is envisaged and the radioactivity will be measured either using anticoincidence low-level β^- counting or via scintillation counting. If the techniques are successful, measurements will be done also with 8.5 and 10.5 MeV neutrons.

For studying the $^{151}Eu(n,2n)^{150}Eu$ reaction irradiations have been done at E_n = 9.6, 10.1 and 10.6 MeV, each for 8 h. The product ^{150}Eu($T_{\frac{1}{2}}$ = 35.8 y; EC = 100 %; E_γ = 334 keV; I_γ = 94 %) has been identified and quantitative γ-ray spectroscopic analysis is in progress.

For investigations on the $^{159}Tb(n,2n)^{158}Tb$ reaction, three irradiations with neutrons of energies 9.6, 10.1 and 10.6 MeV have been done. The product ^{159}Tb($T_{\frac{1}{2}}$ = 180 y; EC = 82 %; β^- = 18 %; E_γ = 944 keV; I_γ = 43 %) has been definitely identified. A quantitative γ-ray spectroscopic analysis will be soon initiated.

Hauser-Feshbach calculations on the excitation functions of the three reactions will be performed in due course of time.

NEUTRON INDUCED REACTION CROSS-SECTIONS ON ^{115}In AT AROUND 14 MeV*

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ABSTRACT

A systematic investigation was carried out on ^{115}In isotope to determine the contribution of different reactions to the total non-elastic cross-section in the 13.43 and 14.84 MeV range. All the major component cross-sections of σ_{NE} were measured with exception of the $\sigma^{g}(n,n')$. In the knowledge of σ_{NE}, the energy dependence of $\sigma^{g}(n,n')$ could be deduced. The isomeric cross section ratios both for (n,2n) and (n,n') processes were also determined in the given energy range. The present experiment proves the dependence of $\sigma^{m}/(\sigma^{g}+\sigma^{m})$ ratio on the spin value (I_m) of the isomeric state in (n,2n) reaction. Excitation functions of (n,2n), (n,n') and (n,ch) reactions were compared with results calculated by STAPRE code.

1. INTRODUCTION

The knowledge of the shape and magnitude of excitation functions for fast neutron induced reactions on ^{115}In is of interest for nuclear reaction theory, and it is in connection with the use of indium as a threshold detector for unfolding the neutron spectra. In the case of A≲100, the difference between the nonelastic (σ_{NE}) and the (n,2n) cross sections is mainly due to the inelastic scattering

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because of the negligibly small contributions from any other reactions at 14 MeV neutron energy. Therefore, by studying the (n,2n) and (n,n') cross-sections, one can get information on those properties of nuclei which are dominant in nuclear reactions. The aim of this work is to measure the partial cross-sections of 14 MeV neutron induced reactions on ^{115}In and to deduce the $\sigma^g(n,n')$ and $\sigma^g(n,n')/\sigma^m(n,n')$ values from a comparison of these data with the evaluated σ_{NE} values. The energy dependences and magnitudes of the (n,2n), (n,n') and (n,p) reactions obtained from the present experiment between 13.43 and 14.84 MeV neutron energy have been compared with results calculated by STAPRE code.

2. EXPERIMENTAL PROCEDURES

High-purity (Goodfellow Metals) metallic samples of natural In with dimensions of 15x10 mm^2 and thickness of 375 mg/cm^2 were irradiated by the home-mode neutron generator of the Institute of Experimental Physics. Neutrons of energy between 13.43 and 14.84 MeV were produced via the $^3H(d,n)^4He$ reaction, using an analyzed D^+-beam of 200 keV. In order to reduce the number of secondary neutrons from the generator an air-jet cooled 0.3 mm thick Al-backed Ti-T target was used in a scattering free arrangement[1]. Samples and Nb fluence monitor foils of 0.5 mm thick placed back-to-back were fastened to an aluminium support ring of a 23 cm inner diameter with the beam spot (diameter ~0.5 cm) at the centre[1]. The energy points in the available energy region were distributed roughly equally by the angular positions of the samples. To estimate the effect of scattered neutrons, the yields of $^{115}In(n,\gamma)$, $^{115}(n,n')$ and $^{27}Al(n,\alpha)$ reactions have been measured as a function of distance up to 16 cm from the target spot. The neutron energy versus emission angle has been determined by measuring the ratio of the ^{89}Zr to ^{92m}Nb specific activities produced both in the Zr and Nb foils by (n,2n) reactions[2,3,4]. Cross-section data for the $^{90}Zr(n,2n)^{89}Zr$ and $^{93}Nb(n,2n)^{92m}Nb$ reactions were

taken from Pavlik et al.[5] and Ryves[6], respectively. Neutron energy spreads (1/2 FWHM) at E_n=14.8, 14.4, 14.0 and 13.5 MeV were found to be[7,8] 210, 130, 40 and 130 keV, respectively.

The neutron fluence rate was monitored continuously by a BF_3 long-counter over time intervals much shorter than the half-life of the isotope being measured. Correction for the variation of the neutron output with time was applied both for single isotopes and for a parent-daughter relationship between two isotopes[9,10,11].

The activities of the In samples and of the corresponding Nb fluence monitor foils were measured both by Ge(Li) and NaI gamma-ray detectors. The self-absorption correction factor for the 190 keV gammas has been determined experimentally by measuring the relevant peak areas obtained by indium foils of various thicknesses, having activated the In homogeneously by thermal neutrons.

The absolute full-energy peak efficiency of the Ge(Li)[12] and the NaI[15] γ-ray detectors has been determined by using standard gamma-ray sources and activated samples. Empirical analytical expressions were given for the description of the energy-efficiency curves for different positions and dimensions of both the sources and samples. The total error of the full-energy-peak efficiency in the 180-1500 keV range was found to be 1.0 % for the Ge(Li) detector. The NaI detector has been used only for relative activity measurements.

The parent-daughter ^{115g}Cd-^{115m}In pair required an analysis of the complex decay curve for the determination of the cross-sections for the $^{115}In(n,p)^{115g}Cd$ and $^{115}In(n,n')^{115m}In$ reactions. The contribution of the ^{115g}Cd decay to the ^{115m}In activity-during the irradiation and measurement - was taken into account by using the following expression[9,10,11]:

$$\frac{\sigma_d}{\sigma_p} = \left(\frac{A_d}{A_p}+1\right) \frac{\lambda_d}{\lambda_{d-\lambda_p}} \cdot \frac{D}{B} + f_p \left(\frac{\lambda_d}{\lambda_{d-\lambda_p}} \cdot \frac{C}{B} - 1\right) \qquad (1)$$

where A_p and A_d are the activity of the parent and daughter nuclei at the end of the irradiation, f_p is the fraction of the parent nuclei decays to the daughter nuclei, while B, C and D describe the time variation of the neutron fluence rate as well as the build up and decay of the radioactive nuclei.

The cross-section of the $^{115}In(n,2n)^{114g}In$ reaction was determined by measuring the 72s β^--activity of ^{114g}In and referred to the $^{27}Al(n,p)^{27}Mg$ reaction. Corrections for the G-M counter efficiency and self-absorption in the samples as a function of maximum beta energy were determined by experiment. Cross-section curve between 13.0 and 15.0 MeV for $^{27}Al(n,p)^{27}Mg$ taken from Manokhin et al.[12] was normalized to the recommended data[6] at 14.7 MeV.

The contribution of the $^{113}In(n,\gamma)$ reaction to the ^{114m}In and ^{114g}In activities has been determined experimentally by placing In samples at different distances from the beam spot. The effect of background neutrons was deduced from the distortion of the $1/r^2$ dependence of the ^{114}In activities. The corrections for the low-energy neutrons reduced the measured ^{114m}In and ^{114g}In activities by 1.8 % and 1.0 %, respectively, at the 11.5 cm source-sample distance. The real coincidence correction was below 1 % in all cases.

A significant contribution to the ^{115m}In activity from neutrons produced in D-D reaction was found. This correction has been measured by replacing the TiT target with a Ti plate, assuring the same irradiation conditions as hold for D-T neutrons. The $1/r^2$ dependence of the ^{115m}In activity has shown that the contribution of the target and sample scattered neutrons both for D-T and D-D reactions can be neglected. It was found that neutrons produced by a D^+-beam of 200 keV and 200 μA in a Ti target can contribute to the ^{115m}In activity with about 4 % in average, depending on the position of the sample. The relative angular distribution of D-D background neutrons measured by the $^{232}Th(n,f)$ process using Makrofol KG track-etch detector foils was found to be in agreement with a thick target yield of a point-like source.

Table 1. Nuclear data and detection methods

Radio nuclide	Half-life [15]	Energies of the measured γ-rays [16]	γ-emission probabilities [16]	Detector	Fluence method
^{114g}In	71.9 s	-	-	GM	$^{27}Al(n,p)$
^{114m}In	49.51 d	191.6	0.16	NaI, Ge(Li)	$^{93}Nb(n,2n)$
^{115m}In	4.486 h	336.2	0.458	NaI, Ge(Li)	$^{93}Nb(n,2n)$
^{115g}Cd	53.38 h	336.2	0.497	NaI, Ge(Li)	$^{93}Nb(n,2n)$

Table 2. Cross-section results for ^{115}In

Neutron energy (MeV)	σ(mb)					
	$^{115}In(n,2n)^{114g}In$	$^{115}In(n,2n)^{114m}In$	$^{115}In(n,n')^{115m}In$	$^{115}In(n,p)^{115g}Cd$	$^{115}In_{NE}$	$^{115}In(n,n')^{115g}In$
14.84	268±6	1346±42	55.0±1.0	5.3±0.5	1917	229±21
14.66	271±7	1329±37	55.0±1.0	4.9±0.4	1921	248±23
14.35	263±7	1314±58	57.4±1.0	4.4±0.4	1928	277±27
14.10	263±7	1307±30	60.4±1.2	4.9±0.5	1932	285±28
13.85	259±8	1291±45	61.5±1.3	4.2±0.5	1938	312±40
13.74	-	1278±15	62.5±1.2	4.1±0.4	1940	-
13.64	258±8	1271±18	68.2±1.5	4.1±0.5	1940	328±34
13.48	-	-	-	3.5±0.4	1946	-
13.43	249±8	1229±41	73.0±1.5	-	1947	-

The decay data accepted for the determination of the cross-sections[15,16] are given in Table 1.

3. RESULTS AND DISCUSSION

The results obtained for the $^{115}In(n,2n)$, (n,n') and (n,p) reaction cross-sections between 13.43 and 14.84 MeV incident neutron energies are summarized in Table 2. Uncertainties combined by quadrature have been estimated at 1 σ. The assumed values of the $^{93}Nb(n,2n)^{92m}Nb$ and $^{27}Al(n,p)^{27}Mg$ cross-sections[6] at 14.7 MeV were (460 ± 5)mb and (68.8 ± 0.7)mb, respectively. As it can be seen in Figs. 1 and 2, the shapes and magnitudes of the measured and calculated excitation functions for the (n,2n) and (n,n') reactions are in good agreement. Among the charged particle emission cross-sections,

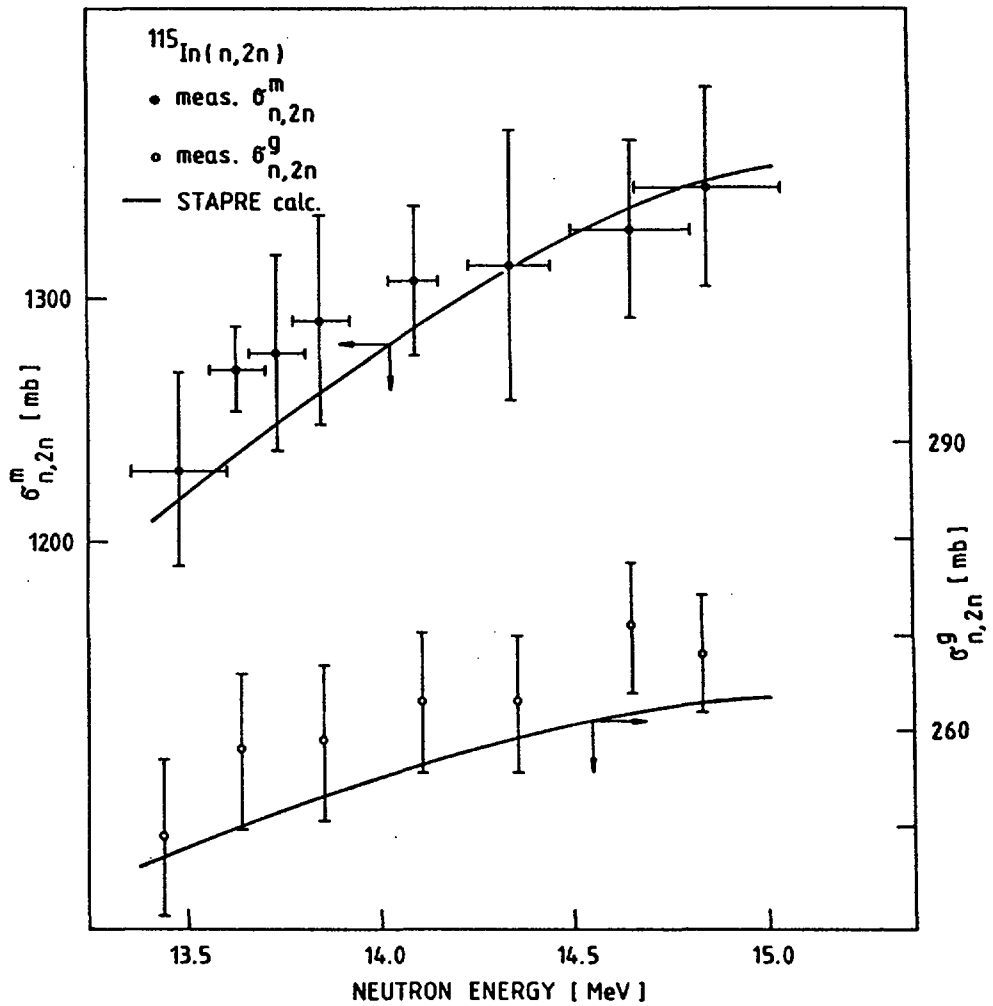

Fig. 1. Measured and calculated excitation functions for $^{115}In(n,2n)^{114m,g}In$ reactions

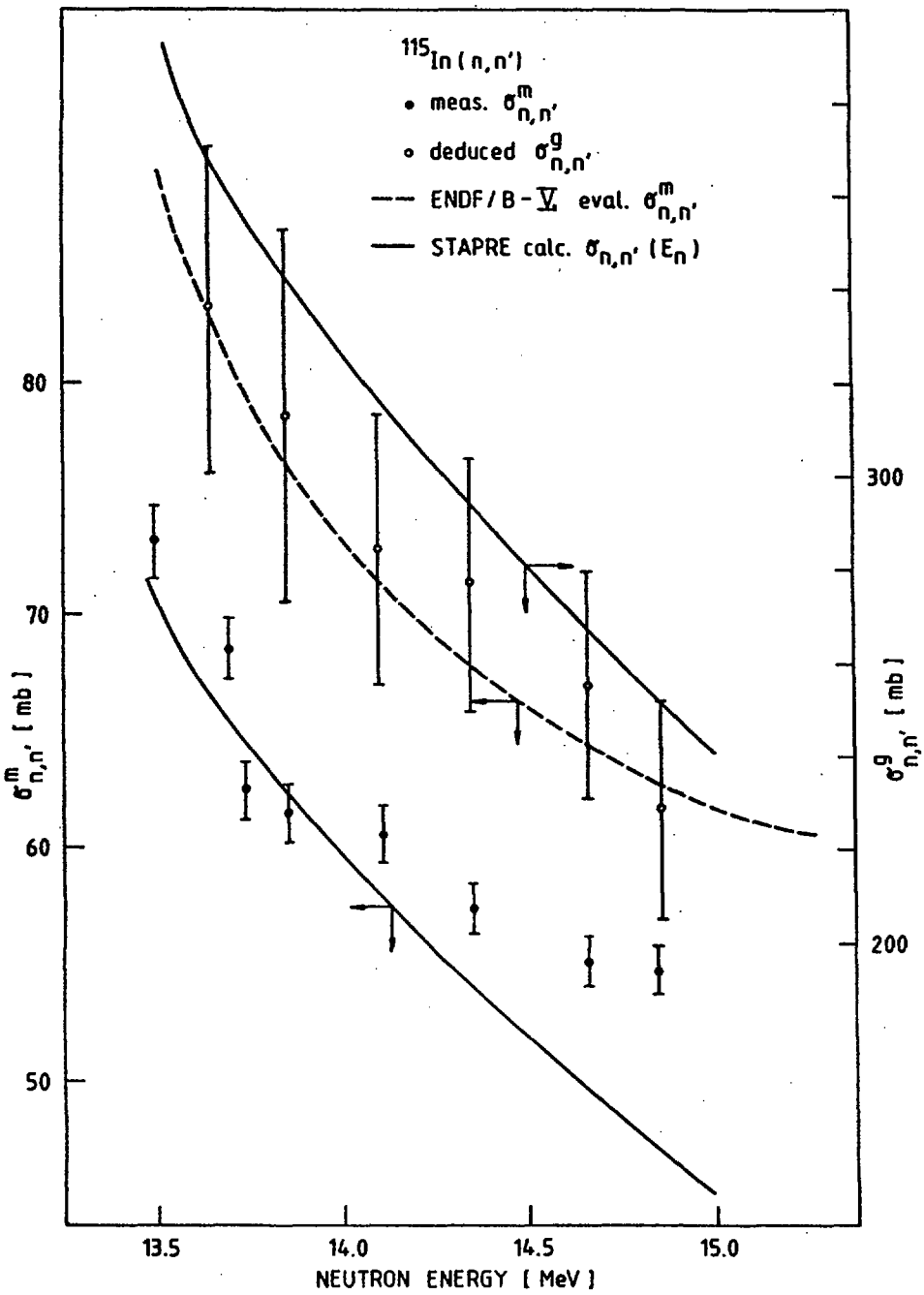

Fig. 2. Measured and calculated excitation functions for $^{115}In(n,n')^{115m,g}In$ reactions

only the (n,p) was measured. The results are shown in Fig.4 together with $\sigma_{tot}(n,ch)$, the (calculated) total charged particle emission cross-section.

In a previous work[17] an analytical expression was given for the description of the energy and mass number dependence of the nonelastic cross-sections. The mass number dependence of σ_{NE} at 14 MeV can be well approximated[18] by the following formula (see Fig.5)

$$\lg\sigma_{NE}(b) = a_o + a_1 \lg A \qquad (2)$$

from which σ_{NE}=1932 mb for ^{115}In is obtained at 14.1 MeV. The σ_{NE} has not been measured for indium up till now. All the major component cross-sections of σ_{NE}, with exception of $\sigma^{g}_{n,n'}$, were measured, therefore, from the difference of σ_{NE} and $\Sigma\sigma(n,x)$ the $\sigma^{g}(n,n')$ could be deduced:

$$\sigma_{NE} - [\sigma^{g+m}(n,2n) + \sigma_{tot}(n,ch) + \sigma(n,\gamma) + \sigma^{m}(n,n')] = \sigma^{g}(n,n') \qquad (3)$$

In eq.(3), all the cross-sections obtained in this experiment were accepted. The calculated $\sigma_{tot}(n,ch)$ is consistent both with the measured values[10,19] and the systematics given by Qaim[20]. A value of (1.2±0.3)mb was assumed for the $\sigma(n,\gamma)$[21]. Though the error bars are large, the deduced $\sigma^{g}_{n,n'}$ data are in surprisingly good agreement with the calculated $\sigma^{g}_{n,n'}(E)$ function.

The isomeric cross-section ratios for ^{115}In(n,2n)^{114m,g}In and ^{115}In(n,n')^{115m,g}In are shown in Fig. 3. The measured

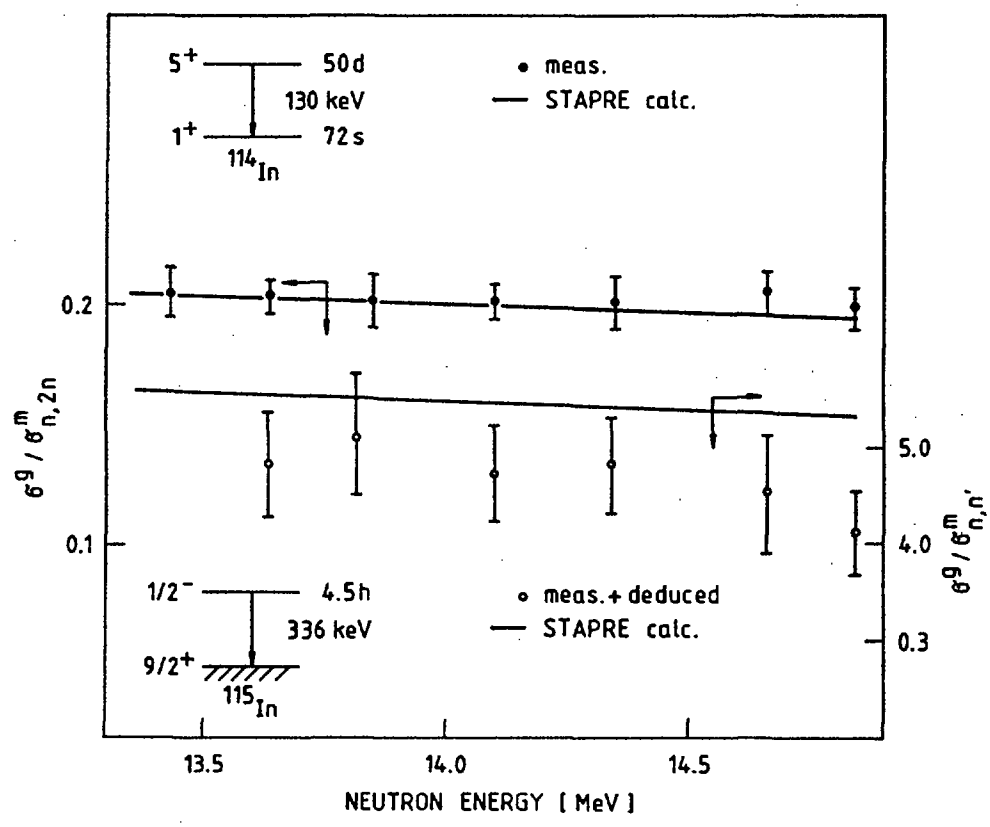

Fig.3. Isomeric cross-section ratios for ^{115}In(n,2n) ^{114m,g}In and ^{115}In(n,n')^{115m,g}In reactions

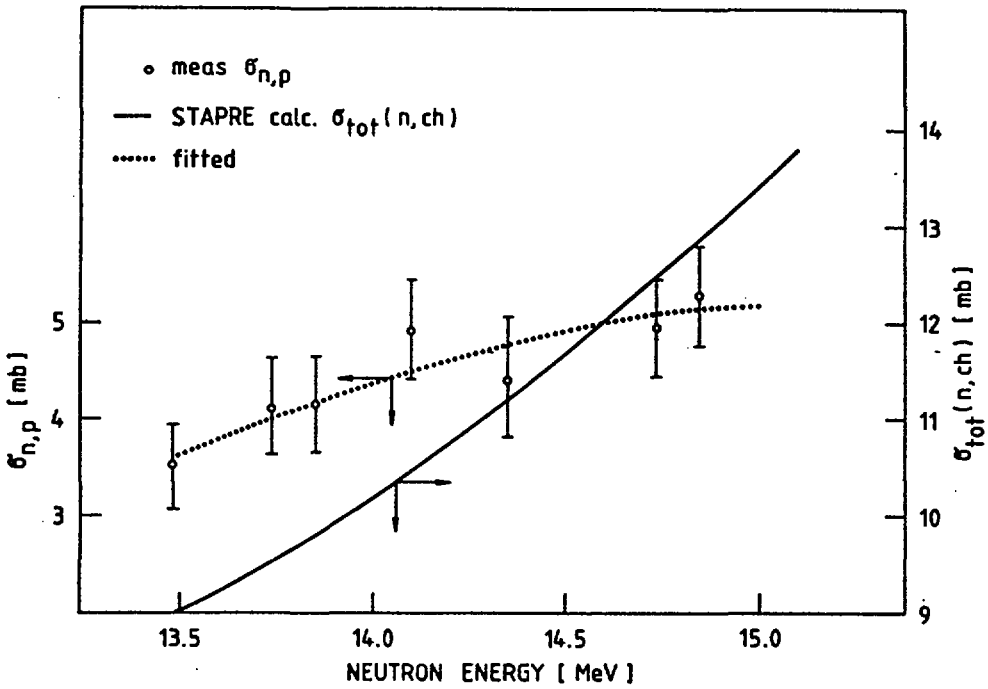

Fig.4. Charged particle emission cross-sections for ^{115}In

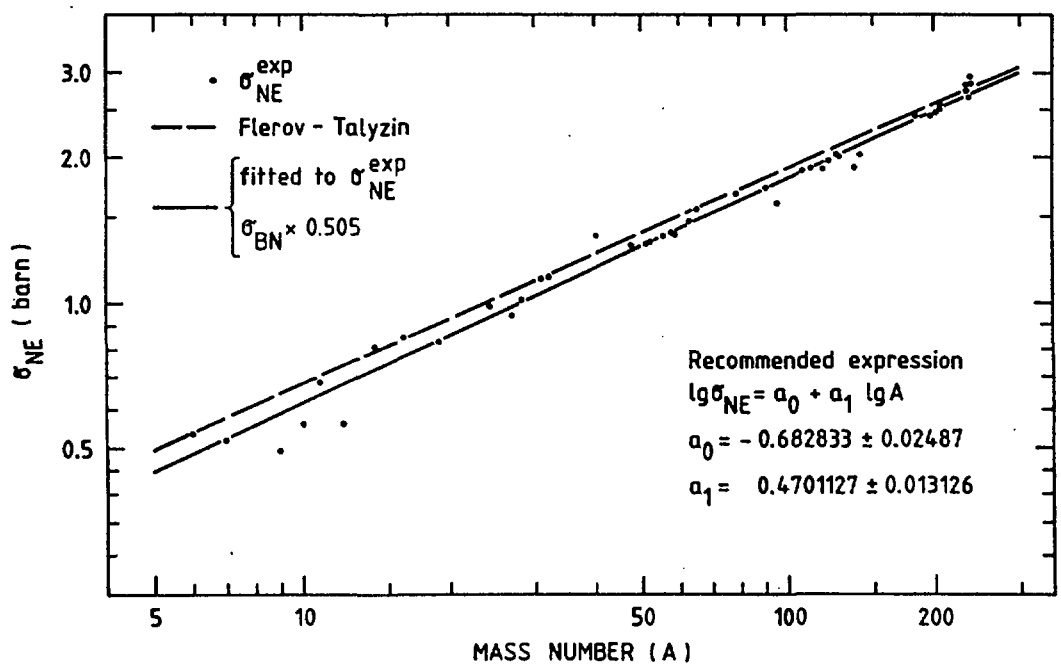

Fig. 5. Mass number dependence of the nonelastic cross-sections at 14 MeV

and calculated σ^g/σ^m values are in good agreement for (n,2n) reaction, while in the case of (n,n') process a difference of about 10-15 % exists, i.e. σ^g/σ^m (meas.)< σ^g/σ^m(calc.).

Table 3. Input parameters used in the STAPRE code

Nucleus	a [MeV^{-1}]	Δ [MeV]	E_d [MeV]	N_d	E_{sep} [MeV]
^{115}In	16.31	- 0.20	1.496	17	6.779
^{115}Cd	16.31	- 0.20	0.962	16	7.447
^{112}Ag	9.90	- 0.90	0.019	2	4.100
^{114}In	15.23	- 0.26	0.969	20	9.029
^{114}Cd	15.74	1.46	2.219	18	6.816
^{111}Ag	9.84	0.04	0.809	15	3.743
^{113}In	14.81	- 0.38	1.471	20	7.313
^{113}Cd	14.81	- 0.38	0.708	10	6.828
^{110}Ag	15.79	- 0.92	0.192	6	3.553

The cross sections for the ^{115}In(n,2n), (n,n') and (n,ch) reactions were calculated by the preequlibrium model using the STAPRE[22] code. Neutron, proton, alpha and gamma emissions were taken into account. The transmission coefficients for these particles have been calculated on the bases of the optical model, using the parameters for neutrons, protons and for alphas as given in[23] and [24], respectively. For the energy and mass dependence of the effective matrix element the $|M|^2=FM\ A^{-3}E^{-1}$ formula was used with a value of FM=250. The separation energies of the emitted particles were taken from the table of Wapstra[25]. The energies, spins, parities and branching ratios of the discrete levels were taken from Nuclear Data Sheets[26]. In the continium region, the level density was calculated by the back-shifted formula[27] using $\sigma = \sigma_{rigid}$. The parameter values accepted in these calculations are summarized in the Table 3.

The experimental results obtained in the present measurements are compatible in general with those published by Ryves et al.[10]. The best agreements with the literature

Table 4. Published ^{115}In cross-sections

Reaction	Energy (MeV)	Cross-section results (mb)	Reference
$\sigma^{m}_{n,2n}$	14.67	1250±30	Ryves et al.[10]
	14.6	1337±120	Barrall et al.[28]
	14.68	1399±81	Santry et al.[29]
	14.96	1393±137	Menlove et al.[30]
	14.6	1331±110	Kayashima et al.[31]
	14.66	1329±37	Present value
$\sigma^{g}_{n,2n}$	14.3	269±7	Ryves et al.[10]
	14.7	269±20	Minetti et al.[32]
	14.35	263±7	Present value
$\sigma^{m}_{n,n'}$	14.3	54.3±2.0	Ryves et al.[10]
	14.5	57.7±2.3	Santry et al.[29]
	14.6	50±7.8	Nagel [35]
	14.3	55±2	Tang Hongqing[33]
	14.35	57.4±1.0	Present value
$\sigma^{g}_{n,p}$	4.3	4.37±0.26	Ryves et al.[10]
	14.6	4.46±0.27	Leschenko et al.[19]
	14.35	4.4±0.4	Present value
$\sigma^{m}_{n,2n}/\sigma^{g}_{n,2n}$	14.3	4.59±0.03	Ryves et al.[10]
	14.7	5.88±0.70	Minetti et al.[32]
	13.86	5.78±0.10	Grochulski et al.[37]
	14.52	5.23±0.08	- " -
	14.35	5.0±0.25	Present value

data are summarized in Table 4. The $\sigma^{g+m}_{n,2n}$ is lower by about 6 % than the BOSPOR evaluation[12] and agrees with the recommended value of Pearlstein[34]. The shape of the $\sigma^{m}_{n,n'}(E)$ function follows the ENDF/B-V. and IRDF evaluations, however, its magnitude lies below the recommended values by 20 %. The present experiment confirms the observed[36] dependence of $\sigma^{m}/(\sigma^{g}+\sigma^{m})$ ratio on the isomeric

state spin value in (n,2n) reaction. Further accurate measurements are needed for other isotopes to confirm the procedure by which the $\sigma^{g}_{n,n'}$ data are deduced.

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ACTIVATION CROSS SECTIONS RELATED TO NUCLEAR HEATING OF HIGH T_c SUPERCONDUCTORS*

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ABSTRACT

Activation cross-sections have been measured for some isotopes of the elements in $Tl_2Ca_1Ba_2Cu_2O_8$ high T_c superconducting oxide. In addition, cross sections for producing long-lived isotopes in Cu and Ag were also determined. Results for the following reactions are given at E_n=14.5 MeV: $^{63}Cu(n,\alpha)^{60g}Co$, $^{90}Zr(n,2n)^{89}Zr$, $^{107}Ag(n,2n)^{106m}Ag$, $^{109}Ag(n,2n)^{108m}Ag$, $^{134}Ba(n,2n)^{133g}Ba$, $^{134}Ba(n,p)^{134g}Cs$, $^{136}Ba(n,p)^{136}Cs$, $^{137}Ba(n,p)^{137}Cs$ and $^{203}Tl(n,2n)^{202}Tl$. Results are compared to the corresponding data published in the literature and given by systematics.

1. INTRODUCTION

During the last decades, interest in the determination of fast neutron cross-sections has been increased because of their use in the designs of fast breeder, fusion, and fusion-fission hybride reactors as well as in radiation damage experiments and cancer therapy. Recent studies on fusion reactors show that the energy will probably be

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Table 1. Data related to the irradiation and measurement

Radio nuclide	Energies of the measured γ-rays[6][keV]	Half-life [5]	γ-emission probabilities [6]	Sample [mm^3]	Cooling time [day]
^{60g}Co	1173.2	5.2719 y	0.999	Cu 12x13x0.7	33
	1332.5		1.0		
^{106m}Ag	717.3	8.51 d	0.291	Ag 12x13x0.2	22
	1527.7		0.164		
^{108m}Ag	433.9	127.7 y	0.907	Ag 12x13x0.2	274
^{133g}Ba	302.9	10.6612 y	0.186	Tl-ceramic	
	356.0		0.623	ϕ 15.3x2.3	40
	276.4		0.0729		
^{134g}Cs	795.8	2.062 y	0.854	ϕ 15.3x2.3	40
^{136}Cs	1235.3	13.002 d	0.198	ϕ 15.3x2.3	40
	818.5		0.997		
^{137}Cs	661.6	30.174 y	0.851	ϕ 15.3x2.3	40
^{202}Tl	439.6	12.232 d	0.914	ϕ 15.3x2.3	40
^{89}Zr	909.2	78.43 h	0.99	Zr 12x13x0.5	22

produced by the D-T-Li fuel cycle. Therefore, the measurements of the activation cross sections with $E_n \leq 15$ MeV for long-lived radionuclides are of primary importance for radioactive waste estimates and nuclear heating of the superconductors[1]. Recently, the nuclear data requirements for fusion energy development summarized in a number of papers[2] indicate that the measurements of activation cross-sections for $^{137}Ba(n,p)$ and $^{109}Ag(n,2n)$ reactions have high priority for dosimetry, too. The aim of this work is to complete the activation cross-sections for long-lived products with high precision.

2. EXPERIMENTAL PROCEDURE

Rectangular and disc shaped high-purity metallic samples of natural Cu, Zr, Nb and Ag as well as $Tl_2Ca_1Ba_2Cu_2O_8$ ceramic were irradiated at the KORONA generator of the GKSS, Geesthacht[3]. The use of high T_c superconducting ceramic sample rendered it possible to measure simultaneously both the cross-sections and the irradiation effects of fast neutrons. The dimensions of the samples are given in Table 1. The total neutron yields were $(1.03\text{-}1.21)10^{14}$ n/cm^2 in approximately 2 hs irradiation time, depending on the positions of the samples. The samples were sandwiched between two Nb fluence monitor foils. The cross sections were measured relative to the $^{93}Nb(n,2n)^{92m}Nb$ reaction for which a value of (460 ± 5)mb was accepted[4] at 14.7 MeV. The neutron energy scale for the thick and extended stack of samples has been determined by measuring the ratio of the ^{89}Zr to ^{92m}Nb specific activities induced in Zr and Nb foils placed back-to-back in different positions inside the sample. The cross-section values obtained for the $^{90}Zr(n,2n)^{89}Zr$ reaction were found to be between 713 and 732 mb for the seven Nb-Zr pairs in the sample stack. On the basis of the cross-section-energy curve, shown in Fig.1., the corresponding energy range is (14.42-14.47) MeV. The average energy (14.44 MeV) is consistent with the (14.5 ± 0.3)MeV value measured in Geesthacht.

Table 2. Cross section results at E_n=14.5 MeV

Nuclear reactions	Cross-section (mb) measured in Geesthacht	Cross-section (mb) measured in Debrecen	Recommended cross section (mb) Qaim[7]	Recommended cross section (mb) Bychkov et al.[8]
$^{63}Cu(n,\alpha)^{60g}Co$	51 ± 2	45.5 ± 2	40 ± 1	(35 ± 8)
$^{107}Ag(n,2n)^{106m}Ag$	570 ± 17	552 ± 20	600 ± 80	400
$^{109}Ag(n,2n)^{108m}Ag$	220 ± 12	263 ± 20	-	27
$^{134}Ba(n,2n)^{133g}Ba$		655^{+60}_{-20}	[1550]	[1640]
$^{134}Ba(n,p)^{134g}Cs$		4.9^{+2}_{-1}	[8]	[7.7]
$^{136}Ba(n,p)^{136}Cs$		6.3^{+2}_{-1}	8 ± 3	8 ± 3
$^{137}Ba(n,p)^{137}Cs$		5.3 ± 0.7	[4.5]	[2.9]
$^{203}Tl(n,2n)^{202}Tl$		1902^{+8}_{-15}	1950 ± 200	(2065 ± 150)
$^{90}Zr(n,2n)^{89}Zr$		723 ± 15	768 ± 78	(768 ± 30)

[ ] from systematics. ( ) from excitation functions.

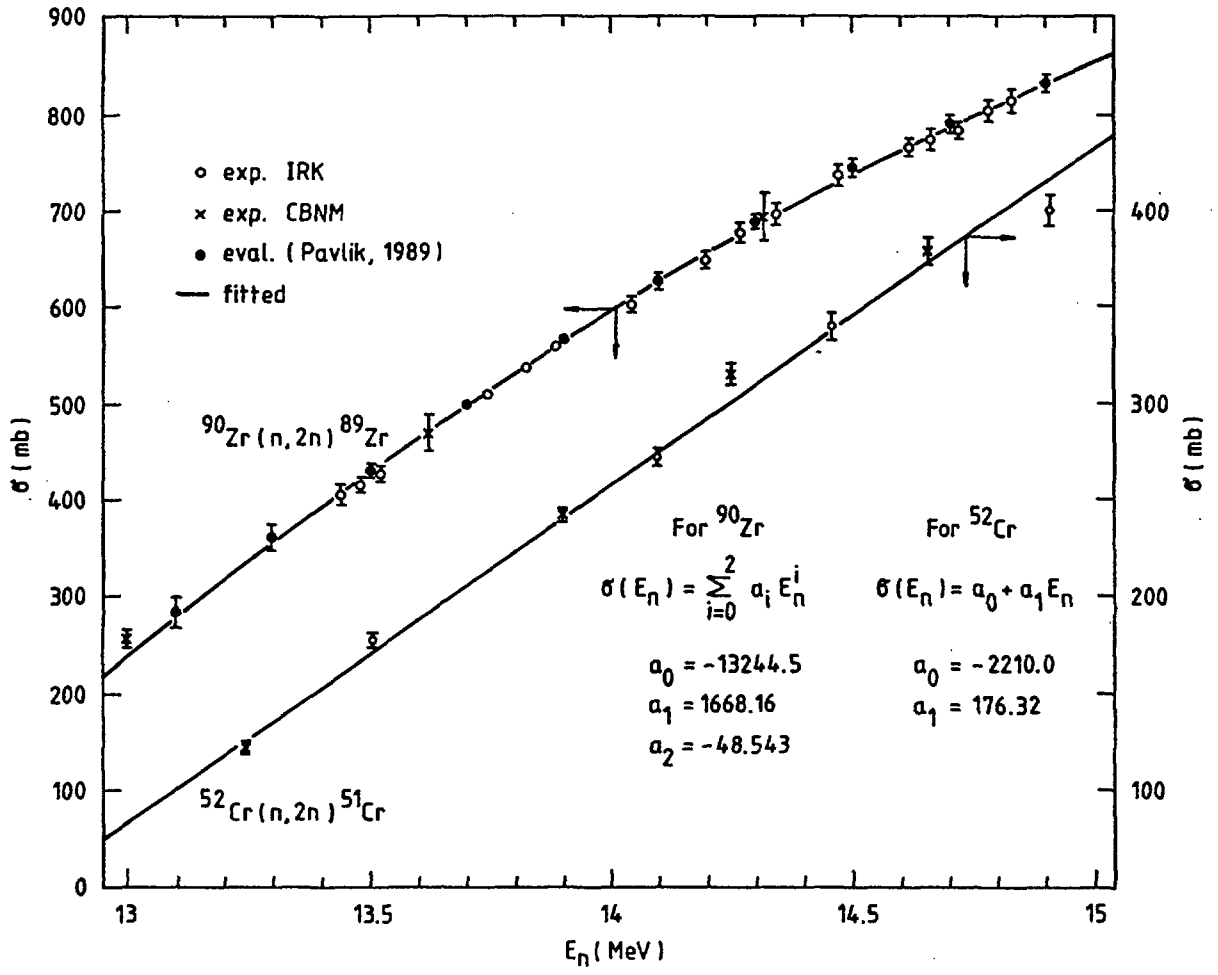

Fig.1. Cross-section curves of $^{90}Zr(n,2n)^{89}Zr$ and $^{52}Cr(n,2n)^{51}Cr$ reactions for the determination of incident neutron energy

The activities of the Ag and Cu samples and the corresponding Zr-Nb monitor foils were measured by calibrated Ge(Li) gamma-ray detectors both in Debrecen and Geesthacht. The activities of the radionuclides produced in the ceramic sample have been measured in Debrecen. Efficiency corrections for different dimensions of the samples were applied. Decay data accepted for the determination of the cross-sections[5,6] are given in Table 1.

3. RESULTS AND DISCUSSION

The cross-sections obtained in the present measurements at 14.5 MeV neutron energy are summarized in Table 2.

Uncertainties combined in quadrature have been estimated at 1σ. As it can be seen in Table 2., cross-sec-

tion data measured in Debrecen and Geesthacht are in agreement within 10 %. Considering the long half-lives and complex decay schemes of the ^{60g}Co, ^{106m}Ag and ^{108m}Ag isotopes, the agreement is satisfactory. The cross-sections for the $^{109}Ag(n,2n)^{108m}Ag$, $^{134}Ba(n,p)^{134g}Cs$, $^{134}Ba(n,2n)^{133g}Ba$ and $^{137}Ba(n,p)^{137}Cs$ reactions were measured the first time in this experiment. Data predicted from systematic trends[7,8] and the measured values differ significantly from each other for these reactions. Therefore, it is needed to improve the systematics by the analysis of the recent more precise data. For Ba the data are in contradiction with the systematic trend observed in the isotopic dependence of (n,p) cross-sections. In general, the results agree better with those recommended by Qaim[7] than by Bychkov et al.[8]. Further precise measurements are needed to complete the data for reaction producing long-lived isotopes and to check the reliability of different systematics[9].

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Measurements of 14-MeV neutron cross-sections for the production of isomeric states in hafnium isotopes

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Introduction

Fusion reactor systems operating on the (d,t) reaction will produce copious numbers of neutrons which will, through nuclear reactions, produce activation products in material subjected to the neutron fluence. In particular, the first wall of a power reactor system and structural materials in that vicinity will be bombarded by very high fluxes which could give rise to intense radioactivity. The neutron fluence is expected to result in radiation damage to the extent that the wall will probably have to be replaced every few years. There is therefore a big incentive to minimise the activity as, if it can be restricted to a low level after a reasonable cooling time, the first wall material could be reused, with obvious economic benefits and minimising the material that might have to be put into a radioactive waste repository.

Some years ago, the UK Fusion Programme initiated a search for viable first wall materials. As activation would be an important criterion in selecting suitable elements, part of that Programme is being devoted to the establishment of a nuclear data library and associated inventory code to enable activation to be calculated with sufficient accuracy. An early library, UKCTRIIIA, was produced by Jarvis (1980) for use with the code ORIGEN.

The neutron flux at the first wall of a fusion power reactor will be of such a magnitude that sequential reactions in a given nucleus will be possible, driving the products well away from the stable region of the periodic table. For this reason, a nuclear data library must contain cross-sections for reactions in unstable nuclides as well as stable ones. To enable the search for low activity materials to be as comprehensive as possible, the nuclear data library also needs to be essentially complete; missing cross-sections could terminate a particular reaction path in calculations of activation, causing misleading conclusions to be reached regarding the suitability of some elements. In the last few years, effort has been directed towards improving UKCTRIIIA, in terms of both the quality of the data and their completeness. The result has been a new library, UKACT1, containing cross-section data in 100-group form for almost 10000 reactions. A complementary library of decay data has also been established and calculations of arisings are carried out with these two data sets using the inventory code FISPACT, a special version of the UK fission reactor inventory code, FISPIN (Burstall (1979)). Further details of the data libraries and FISPACT are to be found in Forrest et al (1988). It is worth noting that FISPACT has the capability to carry out sensitivity analysis of nuclear data and also contains a pathway analysis technique, both of which can be used to identify important reactions.

In studies of potential first wall materials, it was suggested that an alloy containing small quantities of tungsten and tantalum might be worth

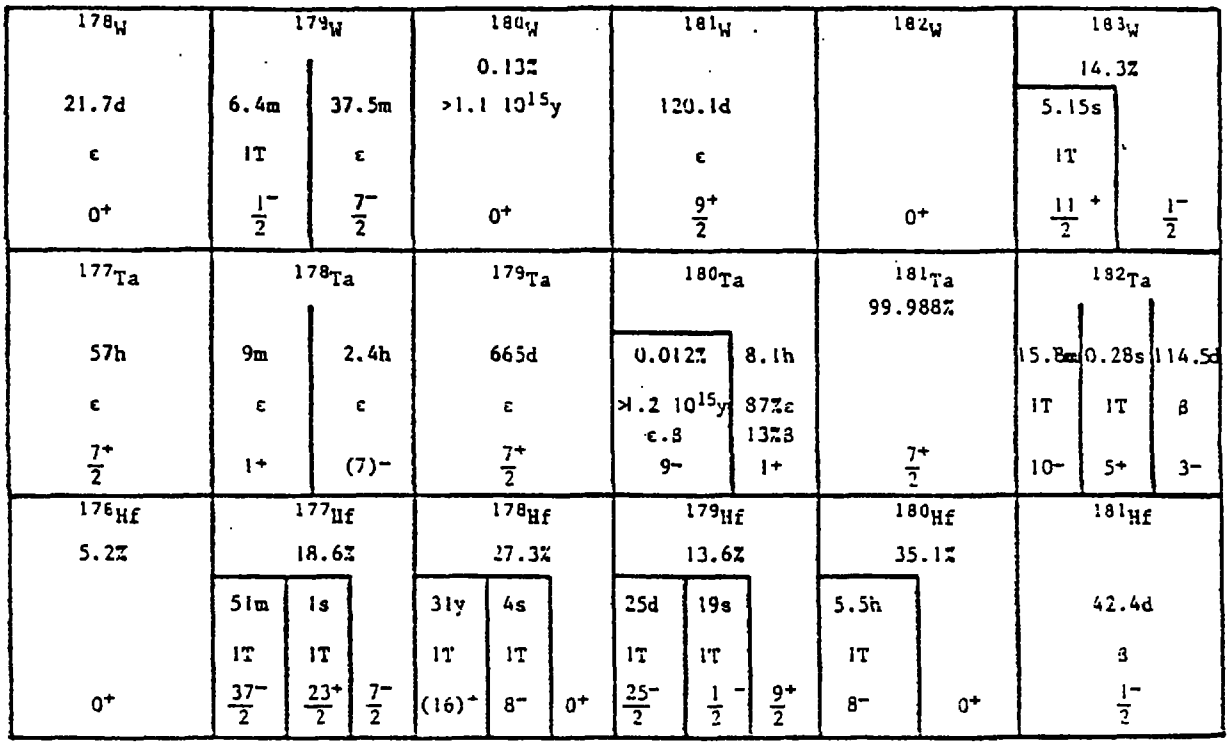

Fig 1. The periodic table in the region of tungsten, tantalum and hafnium

investigating and a request was made for the activation properties of these elements to be investigated. A look at the periodic table in the region of these elements (see Figure 1) identified a possible problem due to the presence of the 31-year isomeric state in Hf-178. The production of significant numbers of nuclei in this state could lead to the first wall being active for many years, making reuse difficult to achieve and therefore raising a potential waste disposal problem. At the time, the production of UKACT1 was still in its infancy and cross-sections for reactions in this region were not fully to hand. Some guesses were made and calculations performed, the conclusion being that production of the isomer in Hf-178 (Hf-178m2) could be sufficient to give rise to an activation problem, but in order to be more specific, better data would be required. An examination of Fig 1 shows that the situation is further complicated by the presence of a 25-day isomeric state in Hf-179 (Hf-179m2). This state could live long enough for an (n,2n) reaction to take place leading to the 31-year state in Hf-178. In fact, the nuclear physics of the situation is such that the cross-section for the reaction Hf-179m2(n,2n)Hf-178m2 in the region of 14 MeV neutron energy could be large as the incident angular momentum required for the reaction to take place is only modest, Hf-179m2 having $J^{\pi} = 25/2^-$ and Hf-178m2 $J^{\pi} = 16+$. Consequently, any Hf-179m2 formed could have a significant probability of being transformed into Hf-178m2.

A search of the literature yielded no relevant measurements or theoretical calculations of cross-sections leading to the isomeric states in Hf-178 and Hf-179, and the question then arose as to how reliable data could be derived. The first solution to this problem came when the Lawrence Livermore National Laboratory (LLNL) offered to irradiate a package of materials, including tantalum, tungsten and hafnium, on the intense 14-MeV neutron generator RTNS-II to enable activation cross-sections to be measured. Later, the co-operation of Los Alamos National Laboratory (LANL) was instrumental in enabling some theoretical calculations of relevant cross-sections to be carried out.

Table 1

Details of materials irradiated on RTNS-II

The foils, 15 mm diameter, were packaged in the order shown, sample number 1 being nearest to the neutron target.

SAMPLE NO	MATERIAL	THICKNESS (mm)	MASS (g)
1	Ni	0.01	0.016
2	Cu	1.10	0.154
3	Au	0.005	0.017
4	Ni	0.01	0.015
5	Co	0.01	0.018
6	Mn/Ni*	0.05	0.060
7	Ni	0.01	0.016
8	Hf	1.00	2.386
9	Ni	0.01	0.017
10	W	1.00	3.170
11	Ni	0.01	0.016
12	Ta	1.00	2.884
13	Ni	0.01	0.015
14	Ti	0.10	0.075
15	SS**	0.14	0.188
16	Ni	0.01	0.016
17	Mn/Ni*	0.05	0.063
18	Co	0.01	0.018
19	Ni	0.01	0.016
20	Au	0.005	0.017
21	Cu	0.10	0.154
22	Ni	0.01	0.014

* 88% Mn, 12% Ni
** stainless steel

Irradiation on RTNS-II

A package of 15 mm diameter foils of various materials was irradiated on the intense (d,t) neutron generator, RTNS-II, at LLNL in March 1987, on the 0° axis close to the tritiated target. The accelerator produced 360 keV deuterons giving a maximum neutron energy of 15.6 MeV. The package was subjected to a fluence of about 10^{18} neutrons/cm^2 over a period of 10 days.

The materials consisted of one foil each of hafnium, tantalum, tungsten, titanium and a low-activation stainless steel, together with neutron fluence monitor foils of nickel, cobalt, gold, manganese and copper, the entire package being wrapped in thin aluminium foil. The package was approximately 6 mm thick, so that there was a significant flux gradient from front to back. Details of the materials are given in Table 1. The monitor foils were chosen on the basis of the accuracy of specific 14-MeV reaction cross-sections leading to states with appropriate half-lives. The package was to be returned to the UK after irradiation and as it was not clear how long shipment would take, short half-lives were ruled out as inappropriate. The

monitor reactions to be used to determine the neutron fluence are shown in Table 2. Care was taken to prevent any possibility of cross-contamination of the foils for which cross-sections were to be measured by ensuring that the foils of these materials were sandwiched between monitor foils. Nickel was used as the primary monitor material, foils being positioned throughout the package to enable the fluence at any point in the package to be determined. The other monitor materials were used to verify the nickel results.

As an independent check on the fluence, the LLNL staff who carried out the irradiation placed foils of niobium on the front and back of the package. The activity in these was measured at LLNL before the package was returned to the UK, the estimated neutron fluences being in agreement with our measurements.

The time distribution of the neutron fluence during the 14-day irradiation period was measured at LLNL by recording the counts in half-hour intervals in a fission chamber placed some distance from the package. This enabled corrections to be made for decay of activation during the irradiation.

Measurement of the activity of the foils

The activity of each foil was measured at intervals over a period of time using a Ge(Li) detector connected to a 4096-channel pulse height analyser system attached to a VAX computer. Dead-time corrections were carried out automatically during the counting of the foils. The areas of peaks corresponding to particular gamma-rays were determined, the half-life being used as a check of the origin of each gamma-ray.

The energy calibration of the analysis system was performed using standard sources, as was the detector efficiency calibration.

The activity of the hafnium foil was dominated for the first year after irradiation by the decay of the 25-day state in Hf-179 and of Hf-175. Only after this activity had largely decayed away did the gamma-rays from the 31-year state in Hf-178 begin to show clearly. Figure 2 shows a spectrum taken several weeks after the irradiation and Figure 3 is one taken two years later. Note particularly the spectra in the region 500-650 keV. In the earlier one, there is virtually no sign of peaks, while in the later one several peaks from the decay of Hf-178m2 are prominent.

To date, only a fraction of the spectra collected have been analysed. The emphasis has been on determining hafnium cross-sections and this paper reports preliminary results. Some corrections have still to be applied to the data (notably for absorption of the gamma-rays in the foils), but these are not expected to be large. It is to be noted that, as natural elements were used in the experiment, it is not strictly possible to obtain unique cross-sections in many cases as there are a number of reactions which could lead to a given activity. However, using a priori knowledge of 14-MeV cross-sections and being guided by theory (see later), it is generally found that one cross-section dominates the route to a particular activity and it has been assumed that all the activity arises from this reaction. However, as will be shown later, this assumption is unlikely to be valid for production of the Hf-179m2 state, where the cross-sections for the reactions Hf-180(n,2n)Hf-179m2 and Hf-179(n,n')Hf-179m2 are believed to be comparable. The consequences of this are dealt with in the section on results.

So far, only the nickel foils have been used to determine the neutron fluence in the region of the hafnium foil and elsewhere in the package. The reactions used are given in Table 2, along with those in other monitor foils

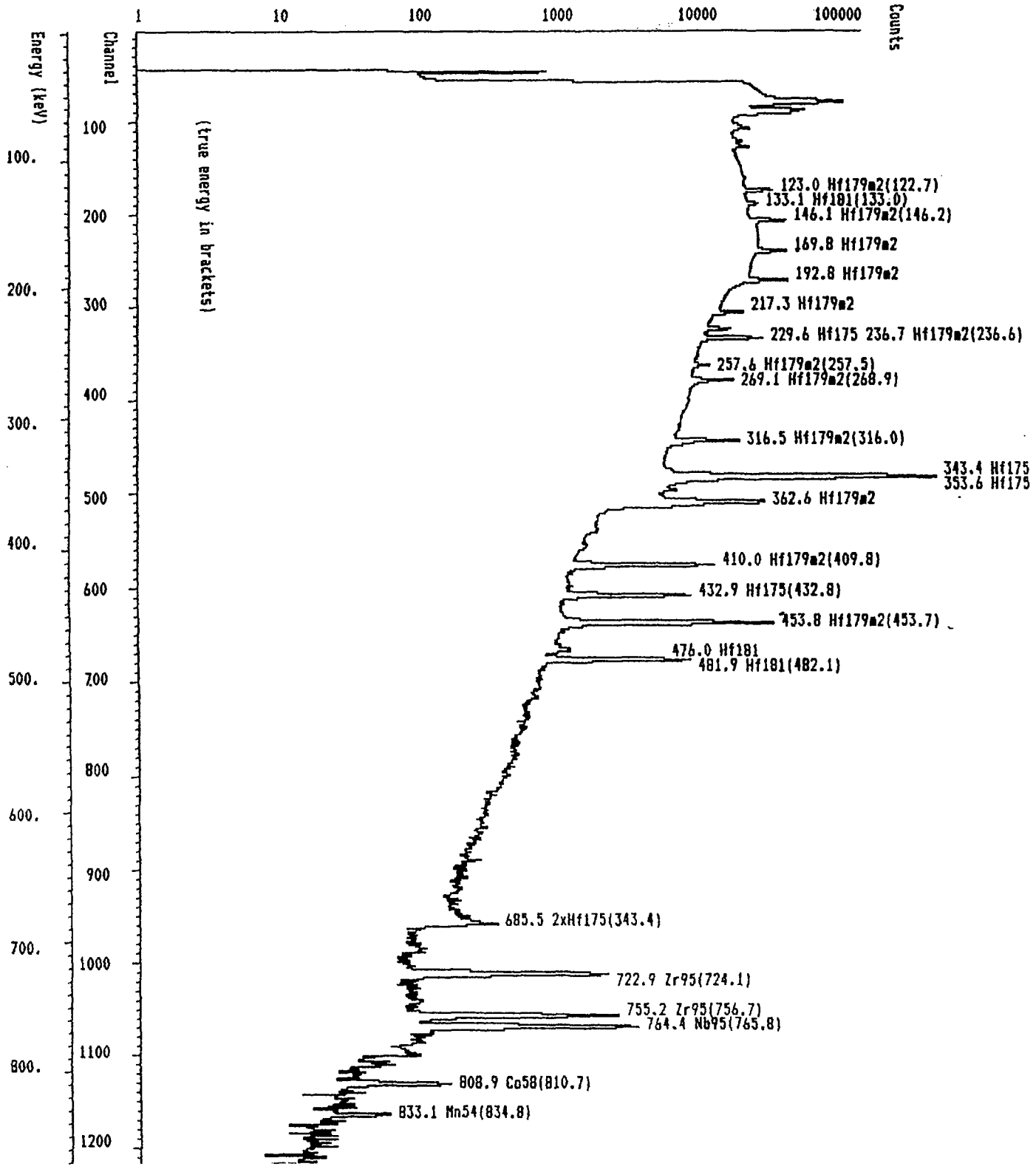

Figure 2 Hf foil spectrum taken 6 weeks after irradiation

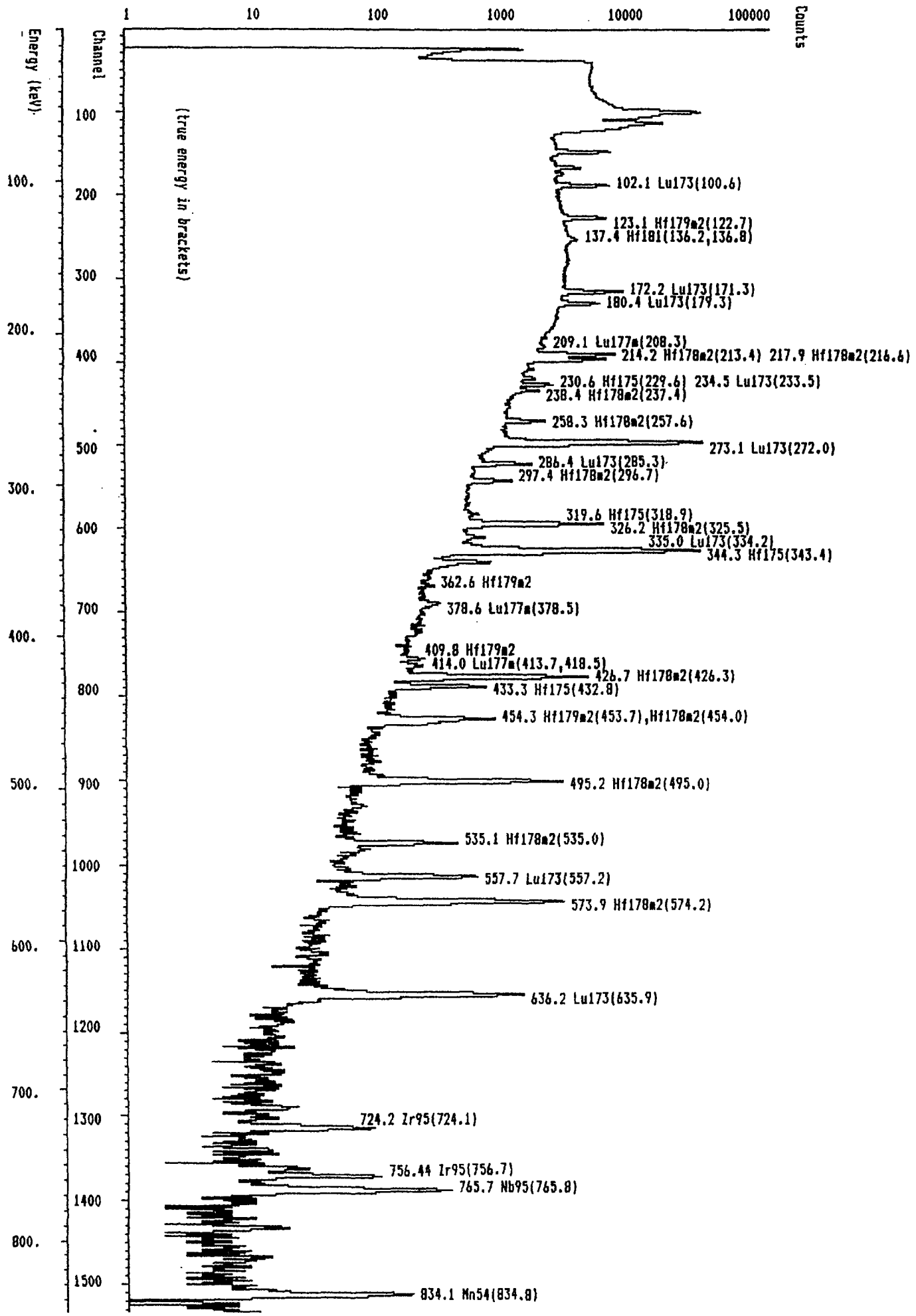

Figure 3 Hf foil spectrum taken 2 years after irradiation

Table 2 Monitor Foils - Reactions Used

Foil Material	Reactions Used
Nickel	^{58}Ni (n,p) ^{58}Co ^{58}Ni (n,pn) ^{57}Co ^{60}Ni (n,p) ^{60}Co
Copper	^{63}Cu (n,α) ^{60}Co
Gold	^{197}Au (n,2n) ^{196}Au
Cobalt	^{59}Co (n,p) ^{59}Fe ^{59}Co (n,2n) ^{58}Co
Manganese	^{55}Mn (n,2n) ^{54}Mn

Table 3 Nickel Monitor Foil Reaction Cross-sections

Reaction	Cross-section* (b)	Gamma Rays Used
^{58}Ni(n,p) ^{58}Co	0.298	511 keV,811 keV 864 keV,1675 keV
^{58}Ni (n,pn) ^{57}Co	0.651	122 keV,136 keV
^{60}Ni (n,p) ^{60}Co	0.129	1173 keV,1333 keV

(* Assuming neutron energy of 15.4 MeV)

which will be used in due course to confirm the fluence values. In order to derive an effective neutron energy for the experiment, fluence values were estimated as a function of neutron energy from the three nickel reactions shown in Table 2, using the known energy dependence of the cross-sections. The variance of the fluence values was then calculated at each energy. The minimum variance was found to lie at a neutron energy of 15.4 MeV and the corresponding cross-sections are given in Table 3. This energy is higher than expected, bearing in mind that the maximum neutron energy is 15.6 MeV. However, the minimum in the variance is fairly shallow and the data are not inconsistent with a lower effective neutron energy when the uncertainties on the cross-sections are taken into account. This point will be examined more closely before the fluences are finalised.

Results

Results of measured hafnium cross-sections are given in Table 4. In deriving these, it has been assumed that the following cross-sections are zero at a mean neutron energy of 15.4 MeV:

Hf-178(n,n′)Hf-178(31-year state)
Hf-178(n,γ)Hf-179(25-day state)

As noted above, there is some evidence that the Hf-179m2 state can be formed

Table 4 Summary of Hafnium Cross-Section Measurements

Reaction	Gamma Rays Used (keV)	Mean Cross-Section	Theoretical Cross-sections Chadwick & Young (1989)
^{176}Hf (n,2n) ^{175}Hf	230 319 343 433	1.77 ± 0.06 b	-
^{179}Hf(n,2n)^{178m2}Hf	258 326 426 495 535 574	5.91 ± 0.64 mb	2.9 mb
^{180}Hf(n,2n)^{179m2}Hf	237 269 316 363 410 454	16.7 ± 1.9 mb*	7.4 mb
^{179}Hf(n,n')^{179m2}Hf	237 269 316 363 410 454	12.8 ± 1.5 mb*	5.7 mb
^{174}Hf (n,2n) ^{173}Hf (^{173}Hf -> ^{173}Lu) β+/EC	272	2.75 ± 0.18 b	-

* The values for these cross-sections were derived from the measured data in the way described in the text.

by two routes with approximately equal cross-sections. Chadwick and Young (1989) carried out a series of calculations of hafnium cross-sections at 14 MeV using the code GNASH at LANL. This code is based on the Hauser-Feshbach statistical model with a pre-equilibrium component.

They estimated the 14-MeV cross-sections for the Hf-180(n,2n)Hf-179m2 reaction to be 7.4 mb and the Hf-179(n,n')Hf-179m2 reaction to be 5.7 mb. Assuming these values are correct in relative terms, the cross-sections for the two reactions have been derived from the measured data, taking the natural abundances of Hf-179 and Hf-180 into account. The results are included in Table 4. Besides deriving cross-sections for the formation of the long-lived isomeric states in hafnium, it has been possible to extract values for (n,2n) reactions on Hf-176 and Hf-174. The former result appears to be about the magnitude to be expected, but the latter is clearly too large. However, it should be noted that Hf-174 has an abundance of only about 0.16%, making accurate measurement difficult. The data will be scrutinised further before final results are issued.

The results of the calculations are also given in Table 4. A comparison of the measured cross-sections with the corresponding theoretical ones shows a very good level of agreement. To calculate these small isomeric cross-sections to within a factor of about two compared with the measurements has to be viewed as remarkably satisfactory and provides some degree of confidence that such calculations can be used to obtain cross-section data of this type for improving fusion cross-section files.

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A SEARCH FOR LONG–LIVED RADIONUCLIDES PRODUCED BY FAST–NEUTRON IRRADIATIONS OF COPPER, SILVER, EUROPIUM, TERBIUM, AND HAFNIUM*

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ABSTRACT

Identical sample packets, each containing samples of elemental copper, silver, europium, terbium, and hafnium, as well as titanium, iron and nickel as dosimeters, have been irradiated in three distinct accelerator neutron fields (at Argonne National Laboratory and Los Alamos National Laboratory in the U.S.A., and Japan Atomic Energy Research Institute, Tokai, Japan) as part of an interlaboratory research collaboration to search for the production of long–lived radionuclides for fusion waste disposal applications. This paper is a progress report on this project. To date, we have detected the following activities, and have obtained preliminary experimental cross section values for several of these: Ag–106m,108m,110m; Eu–150m,152g,154; Tb–158,160; and Hf–175,178m2,179m2,181.

I. INTRODUCTION

A recommendation was made at the 16th Meeting of the International Nuclear Data Committee (INDC), held at Beijing, People's Republic of China, 12–16 October 1987, to establish a Coordinated Research Program (CRP) entitled "Measurements and Calculations of Activation Cross Sections for Long–Lived Radionuclides Important for Radioactive Waste Estimates in Fusion Reactor Technology". The list of reactions to which the attention of this CRP is devoted is given in Table I. These reactions have been identified as being potentially troublesome in fusion nuclear waste disposal considerations. The present investigation was undertaken in support of this project.

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Table I: List of Activation Reactions Included in the IAEA Coordinated Research Program

Reaction	Half–Life (years)
Al–27(n,2n)Al–26	7.2(5)[1]
Cu–63(n,p)Ni–63	100.1
Mo–94(n,p)Nb–94	2.03(4)
Ag–109(n,2n)Ag–108m	127
Hf–179(n,2n)Hf–178m2	31
W–182(n,n'α)Hf–178m2	31
Eu–151(n,2n)Eu–150m	35.8
Eu–153(n,2n)Eu–152g	13.33
Tb–159(n,2n)Tb–158	150
Dy–158(n,p)Tb–158	150
Ir–193(n,2n)Ir–192m2	241
Re–187(n,2n)Re–186m	2.0(5)

[1] 7.2(5) signifies 7.2×10^5.

The original plan of this work was to conduct measurements solely at Argonne National Laboratory (ANL), employing the Fast Neutron Generator Accelerator Facility (FNG) [1] and the thick–target Be–9(d,n)B–10 reaction as an intense neutron source [2]. Using 7–MeV deuterons, this source produces a continuous spectrum of neutrons from < 200 keV to 11.4 MeV. It was reasoned that integral activation data acquired from such measurements would be useful to supplement the 14–MeV results likely to be provided by other participants in this CRP. As the work at Argonne progressed, two collaborators from other laboratories (RCH and YI) joined the Argonne investigators (JWM, DLS and LRG) in this endeavor. The scope of this work was then expanded to include irradiations at Los Alamos National Laboratory (LANL), in the quasi–monoenergetic 10–MeV neutron field produced by the H–1(t,n)He–3 reaction [3] at the LANL Ion Beam Facility [4], and at the Japan Atomic Energy Research Institute (JAERI), Tokai, in the 14–MeV field produced by the JAERI Fusion Neutronics Source Facility (FNS) [5].

As a prelude to the experimental program, nuclear–model cross–section calculations were performed for the reactions in Table I using the pre–compound, statistical model code ALICE [6]. For the (n,2n) processes, the total cross–section strengths derived by this method were allocated among the isomeric states according to the distribution of J in the Gilbert and Cameron level density formula [7]. This is a crude approximation, but it does serve to reduce the weight of high–spin states. These calculations indicated that it would not be practical to produce sufficient activity for reliable measurements (given the available irradiation conditions) for those CRP reactions involving aluminum, molybdenum, dysprosium, tungsten, rhenium, and iridium. Thus, it was decided to abandon these for the present, and to concentrate entirely on investigating those radionuclides produced by fast–neutron irradiation of copper, silver, europium, terbium, and hafnium. Calculated results for several reactions associated with the selected elements are given in Table II, in terms of spectrum–average cross sections for the primary neutrons from two of the accelerator neutron sources employed in this investigation (at ANL and LANL).

Table II: Calculated Spectrum–average Cross Sections (in Millibarns) for Primary Neutrons Associated with the Indicated Neutron Fields[1]

Reaction	H(t,n)[2]	Be(d,n)[3]
Ag–107(n,2n)Ag–106m	87	2
Ag–109(n,2n)Ag–108m	151	3
Eu–151(n,2n)Eu–150m	1264	30
Eu–153(n,2n)Eu–152g	647	12
Tb–159(n,2n)Tb–158	1400	54
Hf–176(n,2n)Hf–175	1571	34
Hf–179(n,2n)Hf–178m2	22	1
Hf–180(n,2n)Hf–179m2	159	5

1 Based on ALICE calculations as described in Section I.

2 For this experiment primary neutrons from the H(t,n) source have energies in the vicinity of 10 MeV [3].

3 For this experiment continuum neutrons from the ANL thick–target Be(d,n) source have energies ranging from < 200 keV to 11.4 MeV [2].

II. SAMPLES

Since it was essential for the success of this experiment to acquire the maximum possible neutron doses in the time available for each of the sample materials, it was decided to irradiate the samples simultaneously in a sample packet that also contained dosimeter foils for determining the neutron fluence. Several identical sample packets were prepared (as shown in Fig. 1) for irradiation at the three accelerator facilities. These packets were 2.54–cm in diameter and $\leq$ 2 cm thick. The europium, terbium, and hafnium samples were in the form of oxides, since these rare–earth materials are otherwise rather reactive. These oxides were encapsulated in sealed plastic containers which defined the sample geometries and prevented any loss of material. All the other sample materials were in the form of metal foils. Several nickel samples were included in each packet in order to measure the neutron fluence gradients (see Fig. 1). A group of copper, nickel, iron, and titanium foils, placed at the rear of the sample packet, provided nine reactions with well–determined excitation functions that could be used to either confirm or determine the neutron spectrum. The chemical purity of all samples was > 99.9%.

III. IRRADIATION PROCEDURES

A single sample packet was placed on the beam line at zero degrees for the irradiations which took place at ANL and LANL. At ANL, the individual samples were located between 5–7 cm from the target, while at LANL the corresponding distances were 4–6 cm. Two sample packets were irradiated at JAERI (one to be retained in Japan for activity analysis and the other to be returned to the U.S.A.). These packets were

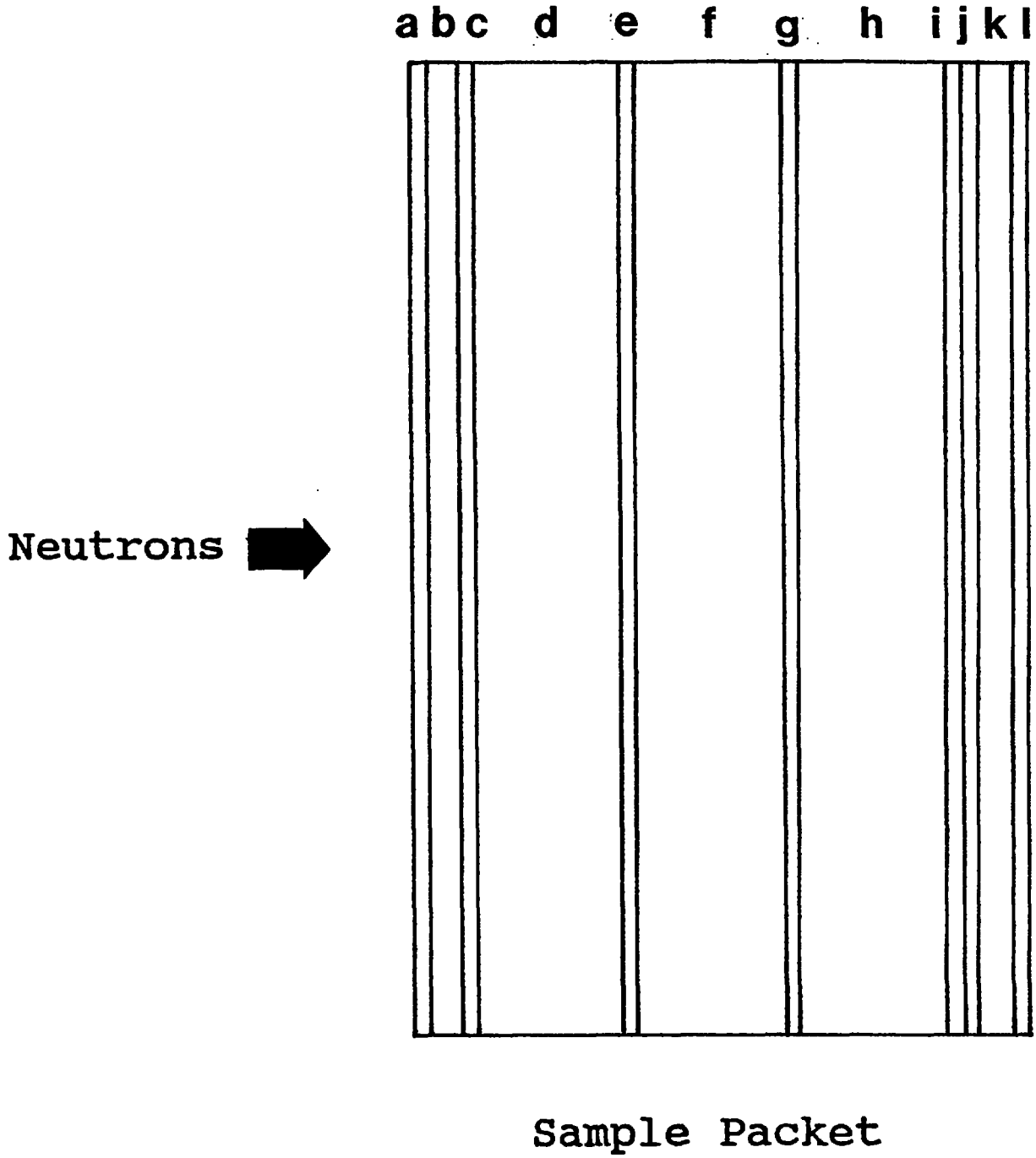

Fig. 1: Schematic diagram for the sample packets used in each of the neutron irradiations. The samples materials are as follows: a = Ni(1), b = Ag, c = Ni(2), d = HfO_2, e = Ni(3), f = Tb_4O_7, g = Ni(4), h = Eu_2O_3, i = Ni(5), j = Cu, k = Fe, l = Ti. The oxide samples are contained in plastic capsules.

Table III

Decay Properties and Other Data [10,11]
for the Radionuclides and Nuclear Reactions
Considered in this Experiment

Activity	Reaction	Half-Life	Principal γ Energy (MeV)	γ Branch (%)	Isotopic Abund. (%)	σ thermal (b)	Iγ (b)	Jπ
Ag–106m	Ag–107(n,2n)	8.46±.10 d	0.512	88±3	51.8	—	—	6+
Ag–108m	Ag–109(n,2n)	127±21 y	0.434	90.5±.6	48.2	—	—	6+
	Ag–107(n,γ)				51.2	0.33	1.2	
Ag–110m	Ag–109(n,γ)	249.8±.04 d	0.658	94.6±1.9	48.2	4.7	72.3	6+
Eu–150m	Eu–151(n,2n)	35.8±1.0 y	0.334	94.0±1.9	46.8	—	—	0–
Eu–152g	Eu–153(n,2n)	13.33±.04 y	0.344	26.6±.5	52.2	—	—	8–
	Eu–151(n,γ)				47.8	5900	1510	
Eu–154	Eu–153(n,γ)	8.8±.1 y	1.275	35.5±.7	52.2	312	1420	—
Tb–158	Tb–159(n,2n)	150±30 y	0.944	43 ± 3	100.0	—	—	—
Tb–160	Tb–159(n,γ)	72.3±.2 d	0.879	29.8±1.5	100.0	23.4	418	—
Hf–175	Hf–176(n,2n)	70±2 d	0.343	87.0±.5	5.2	—	—	—
	Hf–174(n,γ)				0.16	561	436	
Hf–178m2	Hf–179(n,2n)	31±1 y	0.326	94.1±1.9	13.7	—	—	16
	Hf–177(n,γ)				18.6	.0002	?	—
Hf–179m2	Hf–180(n,2n)	25.1±.3 d	0.453	66±3	35.2	—	—	25/2
	Hf–178(n,γ)				27.1	?	?	
Hf–181	Hf–180(n,γ)	42.39±.06 d	0.482	80.6±.7	35.2	13.0	350	—

positioned symmetrically on opposite sides of the beam line at equal distances from the target, so that they would experience equivalent neutron fields and receive about the same dose. The distance from the neutron source to the front surfaces of the packets was 7 cm. They were oriented at an angle of 43 degrees relative to the incident deuteron beam.

The measurements at ANL and JAERI were entirely passive, in the sense that no data were recorded in real time with any active detectors. The dosimeter reactions employed to passively determine the total neutron fluence were as follows: Ti–46,47,48(n,p)Sc–46,47,48, Fe–54(n,p)Mn–54, Fe–54(n,α)Cr–51, Ni–58(n,p)Co–58, and Cu–63(n,α)Co–60. At LANL, a fission chamber fluence monitor was also employed during the irradiations as an auxiliary neutron monitor. This detector contained a uranium deposit with a total mass of 991 μg of uranium. The isotopic composition was: U–238 (99.585%); U–235 (0.415%), others negligible. The irradiation at ANL consisted of about 110 hours of cumulative exposure, occurring over a period of nearly a month during

Table IV

Fluence as Measured by
Uncorrected Spectral Index Reactions[1]

Reaction	Fluence (neutrons/cm^2) ANL (x10^{15})	LANL (x10^{13})
U–238(n,f)	—	2.26
Cu–63(n,α)Co–60	0.578	1.67
Ni–58(n,p)Co–58	0.681	2.14
Fe–54(n,p)Mn–54	0.649	2.12
Fe–54(n,α)Cr–51	0.647	1.47
Ti–48(n,p)Sc–48	0.679	1.30
Ti–47(n,p)Sc–47	0.586	1.44
Ti–46(n,p)Sc–46	0.632	1.74

[1] Note that the Ti, Fe, Ni and Cu foils used for spectral indexing (labeled "i", "j", "k", and "l" in Fig. 1) all have about the same geometry factors since they are close together in the packets. Furthermore, they were positioned very near to the U–238 deposit during the LANL irradiation.

November and December 1988. Preliminary estimates of the neutron doses to the samples from the ANL exposures fall in the range $(0.58 - 0.68) \times 10^{15}$ neutrons/cm^2 (as deduced from the spectral index results given in Table IV). The irradiation at LANL involved about 14 hours of cumulative exposure, gained in two irradiations during October and November 1988. The LANL irradiations involved doses to the samples which appear to be in the range $(1.3 - 2.3) \times 10^{13}$ neutrons/cm^2 (see Table IV). The JAERI irradiations took place during the time period 20–23 June 1989. A total of 32 hours (divided into four 8–hour intervals) of beam time were employed in the experiment, resulting in an estimated dose of 2×10^{14} neutrons/cm^2.

IV. ACTIVITY MEASUREMENTS

One sample packet is being retained at JAERI for activity analysis. Otherwise, all sample counting is taking place at ANL. Since each of the sample irradiations involved time intervals extending from several days to several weeks (often with extensive interruptions), and since the counting of some of the samples was delayed to accommodate sample transport back to Argonne (from LANL and JAERI), no attempt has been made in the ANL counting to study any radionuclides with half–lives shorter than a few days. Table III shows the activities that are being observed at ANL in this experiment and indicates some of their principal features. All of the activity measurements involve gamma–ray counting with germanium detectors, except for Ni–63 which is a beta emitter with no gamma rays. Analysis of the Ni–63 activity has been deferred, pending availability of support to fund the liquid scintillation counting analysis that will be needed

for this purpose. As of this writing, some of the longer–lived gamma–ray activities are still not measurable for the samples irradiated at ANL and LANL because of interference from shorter–lived species. The error for most of the activity measurements is 2–4%. The samples irradiated at JAERI have not yet been counted at ANL.

V. DATA ANALYSIS.

The analyses performed to date on the data include corrections for activity decay and approximate corrections for geometry and neutron absorption. Information required for the determination of these corrections has been obtained from Refs. 8–11. It should be noted from Table III that there are significant uncertainties in the half–lives of several of the radionuclides and, to a lesser degree, in the branching ratios. This will have a marked effect on the ultimate accuracies that can be expected for the derived cross sections.

So far, no corrections have been applied for neutron multiple scattering in the samples, and for the background produced by target–associated reactions or by room–return neutrons. These matters are presently under investigation, and it is clear that they will involve nontrivial corrections. The sample packets are thick (transmissions of about 90%), implying multiple scattering corrections on the order of 10%. Background corrections at the ANL Be(d,n) facility are small. Previous measurements have shown that room–return in the MeV energy range is not a serious problem. Furthermore, neutrons produced by other target associated reactions are overwhelmed by the Be(d,n) yield and, in any case, are included in the measured spectrum. Corrections for these backgrounds are small and are based on previous measurements. For the LANL H(t,n) source, there is a significant neutron yield from (t,n) reactions with the target structure. These neutrons extend to energies well above the H(t,n) peak. The correction for a particular reaction depends on the shape of the excitation function and may be rather large.

Room–return at thermal and epithermal energies does present a troublesome problem in correcting the ANL and LANL data and may, upon examination, turn out to be a problem for the JAERI data. Some activities are produced by both the (n,γ) and (n,2n) reactions. The (n,γ) cross section is quite small in the MeV region and usually causes no serious problem in the measurement of (n,2n) cross sections. However, these materials have rather large thermal capture cross sections and resonance capture integrals (see Table III), so a small thermal and epithermal component in the total neutron spectrum can cause a serious problem. Five of the radionuclides observed can be produced by both reactions. Two of them (Hf–178m2 and Hf–179m2) should present no problem as these states have very high spins (see Table III) and are unlikely to be populated by s– or p–wave capture, but the other three may require correction. Fortunately, four activities were observed that can only be produced by (n,γ) reactions, and these can be used to estimate the corrections for the others. A preliminary estimate of the thermal fluence was made using the thin absorber approximation plus the capture data in Table III. The samples involved are by no means thin, but fairly consistent results were obtained. The thermal and epithermal fluence was about 0.01% of the total for the ANL measurement, and about 0.02% for the LANL measurement. At these levels, only the Eu–152g yield will need a significant correction. Unfortunately, the thermal capture cross section and resonance integral are large and most of that activity is produced by the (n,γ) reaction. It will be very difficult to obtain reliable Eu–153(n,2n) cross sections without using separated isotopes.

The neutron fluence, based on idealized spectra, was calculated for the eight spectral–index reactions, and the agreement indicates how well the actual neutron spectra agree with the assumed spectra. For the ANL measurement, the calculated fluences are fairly consistent (see Table IV). The lowest and highest values differ by less than 20%, and this should improve once the multiple scattering corrections are made. For the LANL measurement, the lowest and highest values differ by nearly 75% (see Table IV). This is much greater than the expected multiple scattering effect, and it is most probably due to the undetermined target–associated neutron background.

VI. CURRENT STATUS

Preliminary cross sections have been derived from the measurements at ANL and LANL, but we choose to not report these data because several important corrections have not been applied and characterization of the neutron spectra associated with these measurements is still incomplete. These results will also eventually be supplemented by data on the production of at least two additional long–lived activities, once the counting of the samples is complete, and by 14–MeV results from the samples irradiated at JAERI.

VII. PENDING WORK

Several matters must be attended to before this experiment will have been completed. The counting of long–lived activities for the samples from the ANL and LANL irradiations is still pending, as well as all sample counting for the JAERI exposures. More detailed corrections for neutron absorption and geometric effects are required, and the effects of multiple scattering must be estimated. The detailed nature of the neutron fields employed in this investigation has to be determined in order to calculate certain corrections associated with both primary and secondary neutron fields, and to refine the estimates of interfering contributions from (n,γ) reactions which come about mainly from lower–energy neutrons.

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MEASUREMENTS OF CROSS SECTIONS OF THE $^{109}Ag(n, 2n)^{108m}Ag$, $^{151}Eu(n, 2n)^{150}Eu$, and $^{153}Eu(n, 2n)^{152}Eu$ REACTIONS AT NEUTRON ENERGY 14 MeV *

(Progress report)

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Abstract: The cross sections of the reactions producing long-lived nuclides: ^{108m}Ag, ^{150}Eu, and ^{152}Eu have been measured at neutron energy about 14 MeV. An activity of irradiated samples was measured with a Ge(Li)-detector. Isotopically pure samples were used.

INTRODUCTION

In fusion reactors the materials should be used, that do not produce large quantity of long-lived radioactivity at irradiation by 14-MeV neutrons. The IAEA specialists' meeting recommended to pay especial attention to a number of reactions which are important in this respect. In the present work the cross sections of three reactions of this list were measured.

EXPERIMENTAL DETAILS

The samples were irradiated at neutron generators NG-400, where fluence about 10^{13} $n \cdot sm^{-2}$ was received by the samples, and NG-200 (fluence about 10^{14} $n \cdot sm^{-2}$). The first irradiation at low beam current was performed in order to clear up the real background conditions, to choose the optimal parameters of the cycle: irradiation - cooling - measurement, and to refine some other experimental details.

The irradiation of the samples was carried out at the arrangement shown in Fig. 1. The construction consisted of four assemblies that were placed at 0°, 60°, 90°, and 120° with respect to the beam. Every assembly contained three isotopes studied, i.e. ^{109}Ag, ^{151}Eu and ^{153}Eu and also five neutron monitor foils made of ^{93}Nb (two foils), ^{90}Zr, ^{58}Ni, and ^{197}Au. The characteristics of the used samples are given in Table 1. The average distance from the sample to the target was 45 mm. At the angle 90° 7000 mg of the natural europium's oxide (Eu_2O_3) were also placed.

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* This work is carrying out under IAEA research agreement

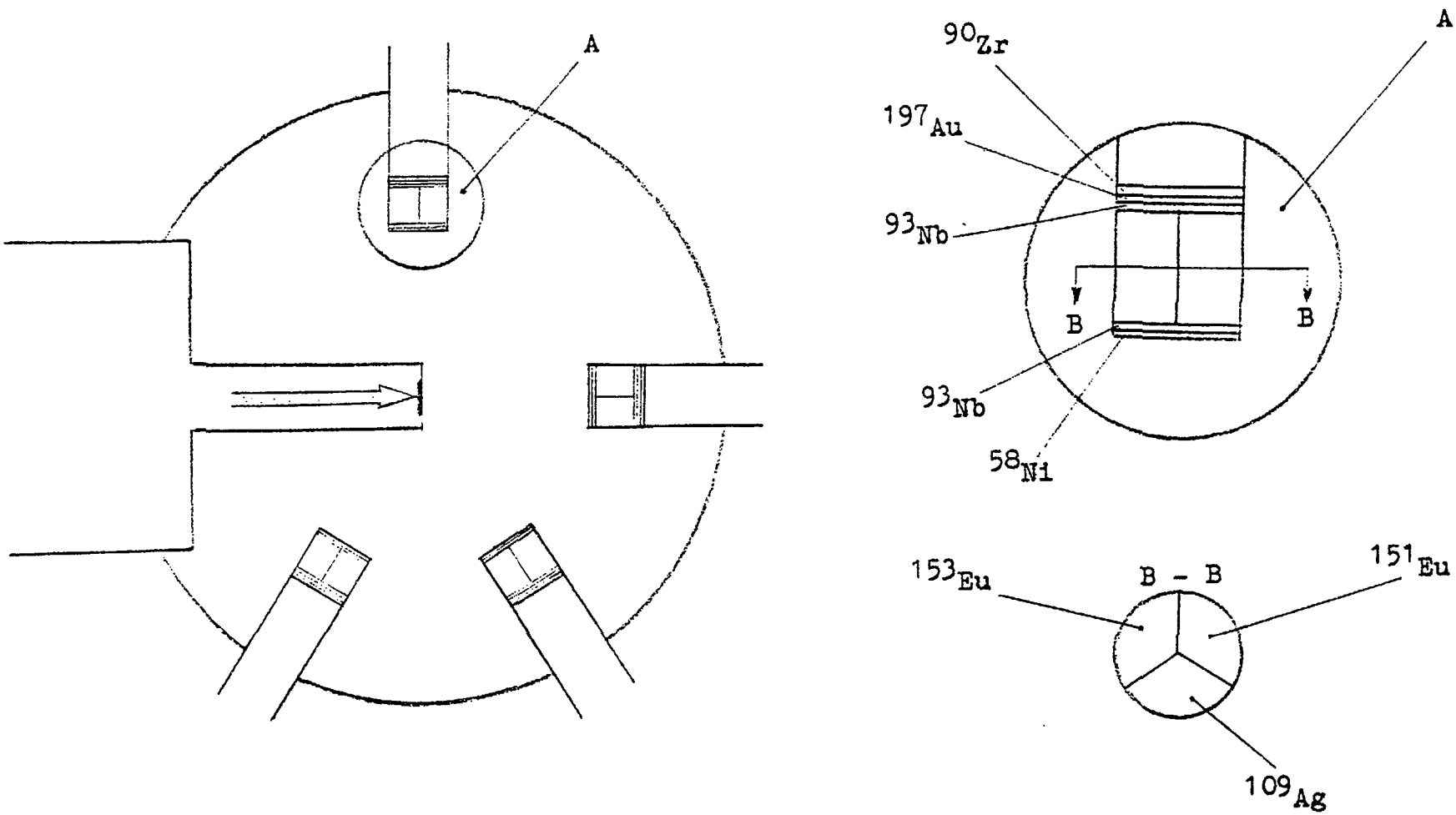

Fig. 1. The experimental arrangement at the NG-400.

Table 1. Isotopic abundance of the samples

Enriched isotope	Chemical form	Isotopic abundance	Pure weight
^{109}Ag	Ag	^{109}Ag - 99.4% ^{107}Ag - 0.6%	4·1250 mg
^{151}Eu	Eu_2O_3	^{151}Eu - 97.5% ^{153}Eu - 2.5%	4·500 mg
^{153}Eu	Eu_2O_3	^{153}Eu - 99.2% ^{151}Eu - 0.8%	4·500 mg

The neutron flux was measured with a long counter and a plastic scintillator counter. To maintain the neutron flux constant the accelerating voltage was gradually increased during irradiation (from 240 kV at the beginning to 280 kV at the end).

The $^{93}Nb(n, 2n)^{93m}Nb$ reaction was used as a standard for neutron fluence determination. The cross section of this reaction was shown to change no more then ±1% in the neutron range 14.1 - 14.8 MeV /1/, an the uncertainty of the evalution of the cross section at 14.7 MeV is 1.6% /2/.

A simultaneous use of foils made of other materials schould increase reliability of the results. Besides, in this case the mean energy of neutrons passed through the sample may be deduced experimentally, since the cross sections of the $^{58}Ni(n, 2n)^{57}Ni$, $^{58}Ni(n, p)^{58}Co$ and $^{90}Zr(n, 2n)^{89}Zr$ reactions depend on the energy quite differently.

The irradiation's arrangement at NG-200, where the neutron fluence about 10^{14} $n \cdot sm^{-2}$ was achieved, differed only in inessential details. There were used two assemblies at 15° and 55°.

The flux, the mean energy, and the energy dispersion of neutrons at the given angle in the laboratory system were calculated by a program using the recommended data on the $^{3}H(d, n)^{4}He$ reaction in the centre of mass system /4/. The program accounted for the real characteristics of the beam (energy, current) and of the target (sorbing material, its thickness, quantity of the absorbed tritium).

The induced gamma-ray activity was measured with a Ge(Li)-detector. Its energy resolution was 2.7 keV and the peak efficiency was 7.9% at 1332 keV gamma-ray energy. Fragments of the measured spectra are shown in Fig. 2-3.

The data were processed by a multichannel analyzer NOKIA LP 4900B, which had a set of programs for treatment of gamma-ray spectra, for de-

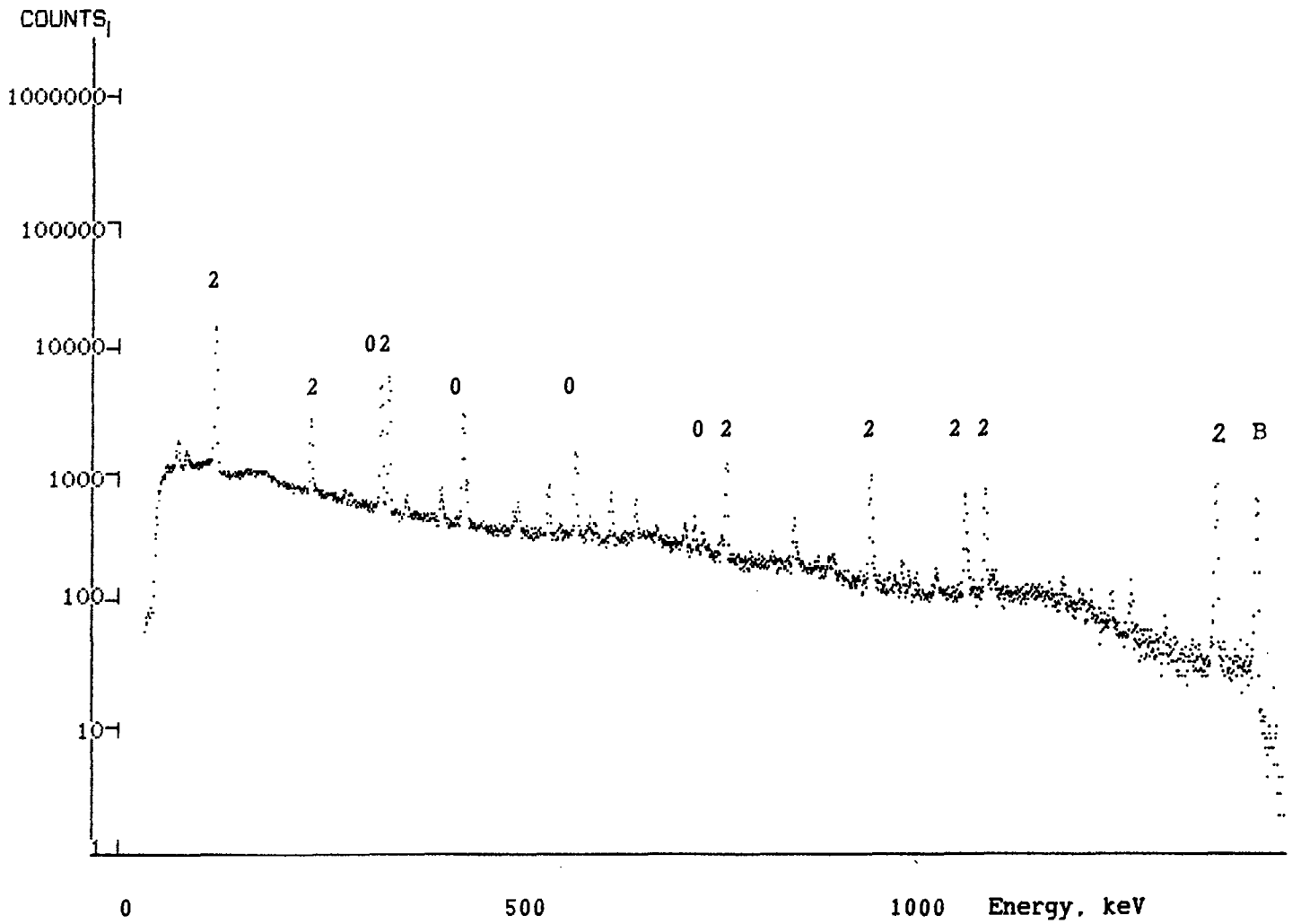

Fig. 2. The γ-ray spectrum from natural europium irradiated by 14.1 MeV neutrons (90^{o}). Cooling is 55 days. Lines from ^{150}Eu are labeled by "0", from ^{152}Eu - by "2"; "B" - background.

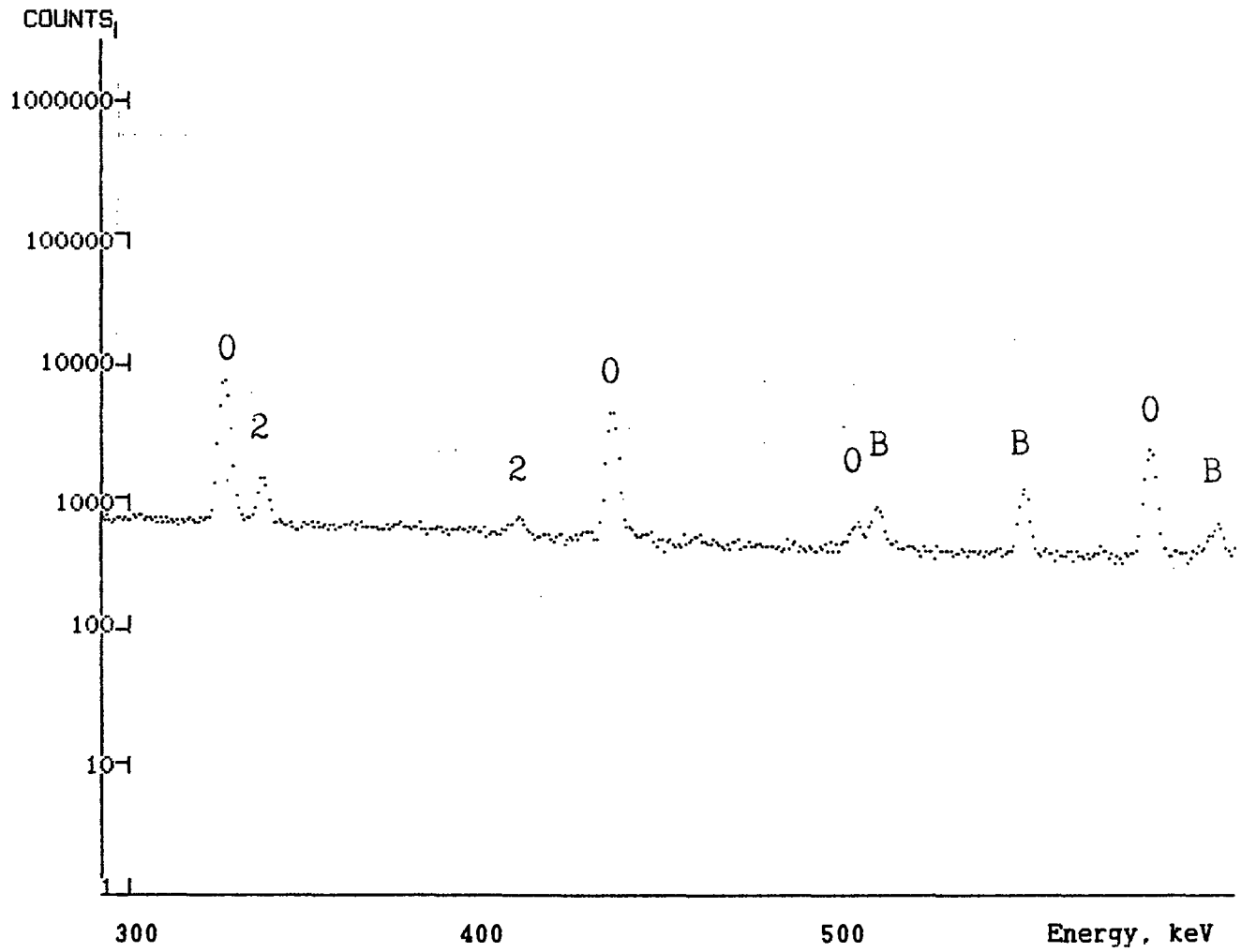

Fig. 3. Fragment of the γ-ray spectrum ^{151}Eu irradiated by 14.1 MeV neutrons (90^{o}). Cooling is 55 days. Lines from ^{150}Eu are labeled by "0", from ^{152}Eu - by "2"; "B" - background.

termination of the spectrometer calibration parameters, and for calculation of volume source activity.

In the present work all gamma-activities were measured in a standard, good calibrated geometry, in which the sample centre was placed at a distance of 62 mm from the effective centre of the detector. The accuracy of the detector efficiency determination and of the source size correction were checked with a number of standard gamma-ray sources. It has been found that the sum uncertainty for these two values does not exceed 3.2% under the experimental conditions used.

RESULTS AND DISCUSSION

The measured cross sections are listed in Table 2. The mean neutron energy given in the first column was obtained by the mentioned program calculation. The mean energy values deduced from a comparison

Table 2. Results of cross sections measurement at neutron energy about 14 MeV

Mean neutron energy (MeV)	Cross section (mb)			
	$^{109}Ag(n, 2n)^{108m}Ag$	$^{151}Eu(n, 2n)^{150}Eu$	$^{153}Eu(n, 2n)^{152}Eu$	$^{93}Nb(n, 2n)^{92m}Nb$
13.7	186(36)	1077(85)	1585(120)	457(9)
14.1	200(39)	1162(93)	1463(110)	461(9)
14.5	204(37)	1147(83)	1482(115)	466(9)
14.9	208(37)	1090(84)	1740(145)	459(9)

Table 3. Nuclide decay data

Radionuclide	γ-ray energy (keV)	Yield (%)	Half-life (Y)
^{108m}Ag	433.937(5)	0.905(6)	127(21)
	614.281(6)	0.899(21)	
	722.938(8)	0.909(21)	
^{150}Eu	333.971(12)	0.960(30)	35.80(10)
	439.401(15)	0.804(34)	
	584.274(12)	0.526(33)	
^{152}Eu	344.281(2)	0.2658(19)	13.33(4)
	778.91(1)	0.1296(7)	
	964.131(9)	0.1462(6)	
	1408.011(14)	0.2085(8)	

of the ^{57}Ni, ^{58}Co, and ^{89}Zr, activities do not contradict to the calculated ones, but have insufficiently high accuracy. This is connected with the fact, that the cross sections of the corresponding reactions are not known with the necessary precision yet. It may be seen, for example, in Handbook on Nuclear Activation Data /3/.

It should be noted, that the data on the $^{93}Nb(n, 2n)^{92m}Nb$ cross section /5, 6/ contained in Ref. /3/ have also rather low accuracy (5-10%) and are in a not very good agreement with the evaluation /2/. Therefore, we decide to measure the cross section of this reaction relative to the well studied cross section of $^{27}Al(n, \alpha)^{24}Na$ /7/. For this purpose, in an arrangement similar to that shown in Fig. 1, some short-time irradiations were conducted, in which the aluminium foils are placed close to the niobium ones. The obtained in such a way

$^{93}Nb(n, 2n)^{92m}Nb$ cross sections are shown in the last column of Table 2. They appear to be constant within 2% in the relevant energy region, according to /1/, and the mean value obtained by us is 461 mb. This differs slightly from the evalution /2/. By neutron fluence determination we have used the cross section value of 461 mb in the whole energy interval 13.7 - 14.9 MeV, assuming its error is 2%.

In Table 3, the gamma-ray energies and yields are shown, that were used at the ^{108m}Ag, ^{150}Eu, and ^{152}Eu activity determination. The data were taken from Refs. /8 - 10/. Every of the listed in Table 3 gamma-lines was searched in the corresponding spectrum, and from its area the radionuclide activity was deduced. Then the mean-weighted value was calculated and the corresponding cross section was obtained.

The main uncertainties of the cross sections listed in Table 2 are the following:

- the uncertainty of the effective fluence determination (including a possible irregularity of the sample mass disiribution) < 6%;
- the uncertainty of the induced activity determination - up to 15% for very weak peaks;
- the half-life uncetaintly - up to 17% for the ^{108m}Ag.

In order to improve the accuracy of the results efforts should be made to increase the neutron fluence received by samples and to obtain a more precise value of the ^{108m}Ag half-life.

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STATUS OF THEORIES FOR CALCULATIONS OF PRODUCTION CROSS SECTIONS OF LONG-LIVED RADIONUCLIDES

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INTRODUCTION

The theories discussed in this paper are confined to those currently being used or considered for the calculation of activation cross sections. The theories are the same regardless of whether the activation product is long lived or short lived. However, the cross sections for the generation of long-lived radionuclides are more difficult or expensive to measure, hence there are fewer data available and the requirement on the predictive capability of the theories used is more stringent.

It is clear from other papers presented in this meeting and the adjoining NEANDC specialists' meeting on activation cross sections that the nuclear theories of interest to this group are those used or needed in modern Hauser-Feshbach (H-F) codes with pre-equilibrium correction and gamma-ray cascades. The H-F formalism is indispensible due to the sensitivity of the calculated results, especially isomeric ratios, to the spins and parities of the discrete levels as well as to the spin distributions in the total and exciton level densities.

Topics included in this paper are the optical model, gamma-ray strength function, total and exciton level-density theories, and the pre-compound model. In each subject, we describe the most commonly used theories first, followed by relatively new developments that are used in at least one model code or the promising theories that do not appear to require a large effort for incorporation into existing H-F codes.

OPTICAL MODEL

The transmission coefficients for the incident and outgoing particles calculated by the optical model are basic ingredients in the H-F formalism. The spherical optical model can calculate such coefficients rather quickly, and most H-F codes now contain an in-house spherical optical model code with built-in global optical model parameters as options. Because the computer cost is still quite large for deformed optical model or coupled-channel calculations, it is more economical to separate the calculation of the transmission coefficients from the rest of the H-F calculations.

For quick calculation of activation cross sections, one would use the global optical model parameters. For more sophisticated analysis, one adjusts the parameters to fit as many experimental data as possible, particularly the total cross sections, s-wave strength function, differential elastic, (p,n), and (α,n) cross sections. Even so, one still encounters troubles such as the 1-MeV minimum in the total cross sections, as discussed below.

The dispersion relations connecting the real and imaginary parts of the optical potential remove many of the anomalies that arise when the spherical optical model is used to analyze accurate neutron data[1]. Of primary concern to the calculation of activation cross sections is the well known fact that the global optical potentials all over-estimate the neutron total cross sections in the region of the minimum around 1 MeV and the calculated

(n,n') and (n,2n) cross sections just above threshold. It is now known that a consistent dispersion analysis gives smaller values of the total cross sections near the 1-MeV minimum in agreement with the experimental data[1,2,3]. Since the dispersion relations also represent the coupling of the elastic and inelastic channels, a consistent dispersion analysis of total cross sections and differential elastic scattering data would also lead to improved reliability and extrapolability in the calculated (n,n') and (n,2n) cross sections as well.

GAMMA-RAY STRENGTH FUNCTION

Theories of gamma-ray strength functions are important for the correct prediction of (n,γ) cross sections where normalization to experimental data is impossible, and for the calculation of (n,z) cross sections above (n,zn) threshold. The giant dipole theory of Brink-Axel[4] is commonly used for E1 and the Blatt-Weisskopf estimates for M1, E2, etc. The predictability of this approach is no better than a factor of two even in the presence of reliable (γ, n) data for the giant dipole resonance. A common remedy is to normalize calculated $\langle\Gamma_{\gamma 0}\rangle/D_0$ values to measured data. Better approaches are emerging to improve the reliability of the calculation in the absence of data, as described below.

Kopecky and Uhl[5] applied the depressed giant dipole model and Gardner and Gardner[6] the energy-dependent Breit-Wigner model to the calculation of (n,γ) cross sections, isomeric ratios, and gamma-ray production spectra. These models give smaller gamma-ray strength functions around 7 MeV than result from using the giant dipole resonance and are in better agreement with the experimental data. These models are easy to implement and may soon be widely adopted in model codes. There is evidence[5,6] that the M1 strength function can be related to a giant resonance around 8 MeV with a width of about 4 MeV.

LEVEL-DENSITY THEORIES

Both the total and exciton level-density formulas are used in modern H-F codes. A common practice is to use the two-Fermion total level-density formula (the Fermi gas formula as in Gilbert and Cameron[7]) for the Hauser-Feshbach part of the calculation and the one-Fermion exciton level-density formula for the precompound part and normalize the sum of the latter to the former at each excitation energy to force consistency. This has been pointed out[8] to be incorrect because the exciton-number dependences in the one- and two-Fermion exciton level-density formulas are also different. It was pointed out that the normalization practice would lead to a harder emission spectrum that may agree with experimental data but in reality only masks the collective component.

Another problem in making a precompound correction to the H-F calculation is the consistency between the total and exciton level densities. It is well known that the total level-density formula has a constant-pairing-energy correction while the pairing correction in the exciton level-density formula depends on both energy and exciton number. An advanced but simple formula for the latter has been developed[9,10,11] and its use in H-F codes is spreading.

Calculations (for example, see Ref. 12) of exciton level densities from realistic single-particle states naturally account for the pairing and shell effects. In particular, such calculations show large differences between the particle and hole excitations and strong isotopic and isotonic effects. A simple method[12] for estimating such effects in the exciton state-density formula has been proposed and is expected to be used soon.

PRE-COMPOUND MODEL

All calculations of activation cross sections presented in this meeting included pre-equilibrium effects. The current approach in H-F codes is to combine the pre-equilibrium results, calculated without angular momentum conservation, with the H-F part that conserves angular momentum. There are several ongoing efforts (for example, see Ref. 13) to make the two parts of the calculation as consistent as possible. Angular momentum conservation in the pre-equilibrium model automatically accounts for the enhanced surface emission and may lead to improved accuracy in the calculated isomeric-ratios. Alpha-cluster formation is also known to be sensitive to angular momentum effects.[14]

CONCLUSION

It is clear that the H-F codes used for the calculation of cross sections are still being improved and meetings such as the present one provide an opportunity for worldwide interaction between the basic and applied physics communities. Many interesting problems in nuclear theories and their implementation in H-F codes exist and call for action. Since the need for activation cross-section data cannot wait for better theories and codes, what we tend to do at present is to over-stretch the model parameters to fit the available data, hence the predictability of the present generation of model codes remains unsatisfactory – probably around a factor of two. When improvements discussed in this paper are implemented, the ability of the codes to predict activation cross sections will improve.

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APPENDIX

LIST OF REGISTRANTS FOR THE

IAEA CONSULTANTS' MEETING

12 September 1989

	Country
Edward Cheng	U.S.A.
Satoshi Chiba	U.S.A. (Japan)
Gyula Csikai	Hungary
Chia-yao (Peter) Fu	U.S.A.
Lawrence Greenwood	U.S.A.
Yukinori Kanda	Japan
Toshio Katoh	Japan
Jiri Kopecky	The Netherlands
Horst Liskien	Belgium
M. Aslam Lone	Canada
Lu Hanlin	People's Republic of China
Frederick Mann	U.S.A.
Wolf Mannhart	Federal Republic of Germany
James Meadows	U.S.A.
Yutaka Nakajima	Japan
Bryan Patrick	United Kingdom
Syed Qaim	Federal Republic of Germany
Thomas Ryves	United Kingdom
Donald Smith	U.S.A.
Jean-Christophe Sublet	United Kingdom
Herbert Vonach	Austria
Maria Wagner	Austria
DaHai Wang	Austria
Nobuhiro Yamamuro	Japan
Phillip Young	U.S.A.
Alan B. Smith	U.S.A.