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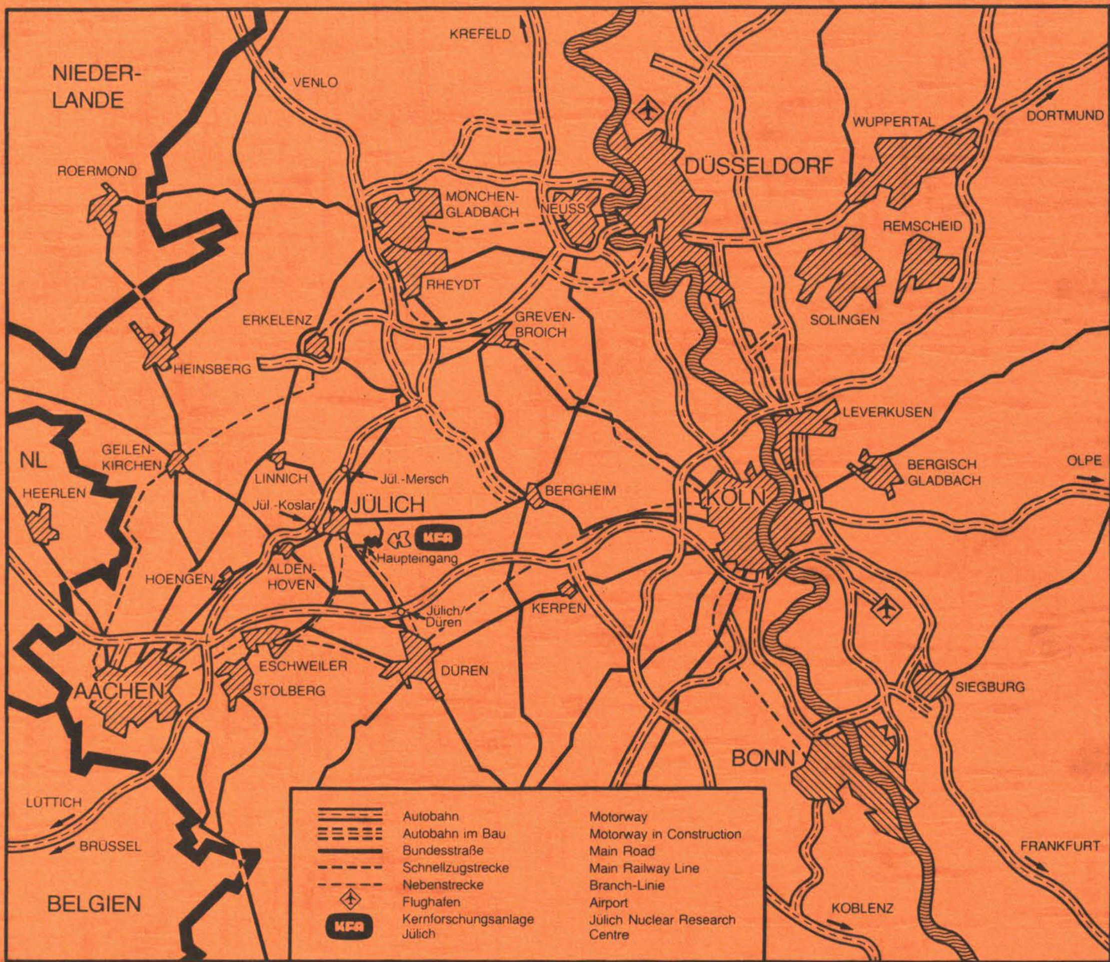

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ABSTRACT

The results of the IAEA "Coordinated Research Programme on Irradiation Embrittlement of Pressure Vessel Steels" were examined again to analyse the reasons behind the reported large scattering and discrepancies in the effect of irradiation on the ductile-brittle transition temperature (DBTT). It is concluded that using specimens from different locations throughout the HSST steel plate thickness was the main reason behind the scattering in these results. Specimens from different locations had different microstructures and different initial mechanical properties which led to different irradiation sensitivities.

The analysis showed that the specimens used can be considered to consist of two sets different in microstructure, mechanical properties and irradiation sensitivity. Set 1 composed of specimens taken from positions between $\frac{T}{4}$ and $\frac{T}{2}$ throughout the plate thickness while set 2 composed of specimens taken from positions between the surface and $\frac{T}{4}$. In this way the scattering became very small and even negligable and the discrepancies were explained.

1. INTRODUCTION

The influence of irradiation on the mechanical properties of metals has a significant impact on the safe behaviour of structural materials employed in nuclear power plants. Of particular concern to the safety of light water reactors is the loss of toughness that occurs in pressure vessel steels as a consequence of their exposure to high neutron fluence levels ($>10^{18} n.cm^{-2}$, E>1MeV) [1].

In October 1967 and October 1968, the IAEA convened two meetings on the problems of pressure vessels which arise during their operation. Both meetings recommended that the IAEA should consider the possibility of organizing a coordinated programme on Irradiation Embrittlement of Pressure Vessel Steels.

The first part of this programme started in 1971 and the results were published and discussed in 1974 [2] and 1977 [3]. The activities continued through the International Working Group on Reliability of Reactor Pressure Components (IWG-RRPC). Programmes are carried out to study the effect of neutron irradiation in weldments, to compare the neutron embrittlement in national steels, and to study the thermal annealing of Pressure Vessel Steels [3, 4].

The main aim of the first part of the programme was to establish that the basis for describing embrittlement, measurement of neutron spectrum, fluence, and mechanical properties are sufficiently standardized to permit direct intercomparison among international programmes without major adjustment of data [2].

Eight institutes in seven countries participated in the programme (see table I). The standard charpy-V impact test has been used for evaluating the increase in ductile-brittle transition temperature due to irradiation. Tests were performed on reference steel ASTM-A 533 Grade 1 (HSST plate O3) [5]. Fig. 1 shows the locations of the blocks used by the participants on the HSST plate O3, while fig. 2 shows the cutting plans for each block into plates and test specimens.

The results showed considerable scattering and discrepancies. In the relation between the increase in ductile brittle transition temperature (DBTT) and the fluence the scattering band width was 65°C. A similar scattering was reported for other plates from A 533 steel and other steels [6, 7].

Nagel [8] attributed the scattering in the IAEA programme results to material-state error and attempted to correct for this error. He reported that the scattering-band width could be reduced to about 50°C.

In the present work the results of the IAEA coordinated programme [2, 3] are examined again and the scattering and discrepancies are analysed.

2. DISCUSSION OF MAIN RESULTS OF IAEA PROGRAMME

2.1 Unirradiated specimens

The DBTT's obtained from Charpy-V notch impact tests for unirradiated specimens are shown in table I.
The DBTT ranged from -28°C to +30°C. However, most of the data lie in the range -6°C to +5°C.

Childress [5] reported the variation in DBTT for the HSST plate 03 as shown in table II and represented in fig. 3. These data were given by the manufacturer and the locations of the test specimens are shown in fig. 1. While DBTT shows large decrease at the plate extremities, i.e. top and bottom, it does not show significant variation across the remaining thickness. Slight DBTT variation was noted related to the position of the blocks along the plate length and no DBTT variation over the plate width was reported by Childress. Stelzman [9] using the same plate showed that there was no DBTT variation over the plate width and confirmed that the variation along the plate length was very small. He also showed that the DBTT variation across the thickness was similar to that shown by the manufacturer [5].

Detailed investigation [10] of the HSST plate 01, which is similar to HSST plate 03 in composition, heat treatment and dimensions, and made by the same manufacturer, revealed a significant dependence of the properties on the depth through the plate thickness. The surface has NDT 65°C lower than the quarter-thickness (1/4 T) or the centre, and 20% higher yield strength. These are about the same variations obtained for plate 03 [5].

Hester and Brooks [10] showed that the above variations are due to variations in the microstructure. The microstructure examination showed that the surface microstructure consists of granular bainite, upper bainite and small amount of lower bainite (and possibly tempered martensite). The microstructure at $\frac{1}{4}$ T and centre consists of granular bainite and a smaller amount of upper bainite. Both the bainite ferrite grain size and the mean free path between carbides are smaller near the surface than $\frac{1}{4}$ T or at the centre. It is concluded that the 65°C lower NDT of the surface and the 20% higher yield strength compared to the $\frac{1}{4}$ T or centre are due to the finer bainite ferrite grain size and/or finer carbide distribution in the microstructure.

The difference in microstructure was attributed to the difference between the cooling rate at the surface and that at $\frac{1}{4}$ T or centre. The surface cooled at about 33°C/s while $\frac{1}{4}$ T at about 0.33°C/s and centre at about 0.22°C/s [10].

The variation of DBTT across the thickness of HSST plate 03 is shown in fig. 3 (A, B). This profile for the variation was selected on the basis of the profile given for HSST plate 01 [10, 11]. This variation was compared with data on the same plate reported by Stelzman [9], shown fig. 3 - curve C, and good agreement is observed. The profile used by Nagel [8] for the same plate, fig. 3 - curve D, indicates the largest variation in DBTT with thickness. He did not show the reason behind adopting this profile.

For the sake of completion the variation of yield strength [5] and fracture toughness [12] across the thickness are shown in fig. 3. Both of the two properties has high value at the surfaces and decreases towards the plate centre.

Based upon the above analysis, the variation in DBTT is expected to take place only in the thickness direction. No variation is expected in the length or width directions. Nagel [8], however, used a correction factor (up to 11°C) in the width direction, which seems to have no physical basis.

It is possible from curves A, B in fig. 3 to estimate the DBTT for the plates 1,2,3,4 and 5. The data are given in tab.III. Therefore, it is expected that the participants using plates 4 and/or 5 will have DBTT different from these using plates 1, 2 and 3. It is important here to note that the ASME Boiler and Pressure Vessel Code [13] requires that acceptable test specimens must be taken out from positions of minimum of $\frac{1}{4}$ thickness from the plate surface. It can be seen from fig. 3 that plate 3 hardly fulfils this condition while plates 4, 5 do not.

Fig. 4 shows the effect of the plate thickness (fig. 4.a) and the plate width (fig. 4.b) on ΔDBTT using the participants date. It is shown (fig. 4.a) that the data obtained by 6 participants lie in the band between the curves by the manufacturer (A) and by Stelzman (C). Fig. 4.b shows that the data obtained by 7 participants lie in a scatter band of only 10°C, and also shows that there is no variation along the width direction. This agrees with the above analysis and shows that no correction is needed along the width direction.

2.2 Irradiated Specimens

The results of the Charpy-V notch tests after irradiation are given in tab. IV and fig. 5. In general ΔDBTT increases with increasing the fluence and/or decreasing the irradiation temperature. The results show large scattering. The scattering band is about 65°C. It is important to note the following cases for specimens irradiated at 290°C:

(a) Comparing the results of participants 1, 4 and 5 for irradiation temperature 290°C and fluence 1.1 - 1.3 x 10^{19}n$\cdot$cm^{-2}, one can notice 100% (±20°C) scattering.

(b) Comparing the results of participants 2 and 8 for irradiation temperature 290°C, fluence 1.8 - 2.0 x 10^{19}n$\cdot$cm^{-2}, and the same reactor type (HWR) a comparable scattering was noticed.

(c) Comparing the results of participants 3 and 5 for the same reactor type (LWR), and irradiation temperature 290°C, it was noted that participant 5 using the higher fluence,gave ΔDBTT which is noticeably lower than that of participant 3 with the lower fluence.

For irradiations at 120°C, the results of participant 4 constitute most of the obtained data. The results show that irradiation at 120°C for a fluence of 1.5 x 10^{19}n$\cdot$cm^{-2} gave ΔDBTT much less than irradiation at 140°C for a fluence of 0.85 x 10^{19}n$\cdot$cm^{-2} or irradiation at 120°C for a fluence of 1.0 x 10^{19}n$\cdot$cm^{-2}. The ΔDBTT for a fluence of 10 x 10^{19}n.cm^{-2} and irradiation temperature 120°C is noticeably less than the ΔDBTT for the same fluence but at 140°C. One can also notice that the smaller ΔDBTT in the first case resulted from irradiation in HWR while the smaller ΔDBTT in the second resulted from irradiation in LWR.

From the above comparison it is clear that the scattering can not be attributed - in the first position - to reactor type, irradiation temperature or fluence. The above scattering in ΔDBTT, however, can be attributed to variation in the initial microstructure and mechanical properties of specimens that resulted from their original locations across the plate thickness.

It was shown that the radiation sensitivity of the steel depends on microstructure [14]. Large grain ferrite-bainite structures show more irradiation sensitivity than those with fine grained structures of ferrite and tempered lower bainite. I was also noted [15], that in general steels having high initial transition temperatures appeared to have higher irradiation sensitivity compared to those having lower initial transition temperatures. Thus, the specimens made from material near the plate surface, where the structure consists of fine bainite and the initial DBTT is low, are expected to have the lowest irradiation sensitivity, i.e. ΔDBTT. The sensitivity increases in the direction from the surface towards $\frac{T}{4}$, and centre, as the structure coarsens and the initial DBTT increases.

This variation in irradiation sensitivity of specimens according to the variation in the location of the specimens throughout the HSST plate thickness can explain the discrepancies revealed in the above comparisons. For the same fluence and irradiation temperature, the irradiation sensitivity (ΔDBTT) obtained by participant 4 using plate 2 was much higher than that obtained by participant 1 using plate 5. Also, for the same conditions, ΔDBTT obtained by participant 2 using plate 2 was higher than that obtained by participant 8 using plate 4.

This also applies to the results of irradiaions at 120°C. Specimens from plate 1 from blocks MD, MQ and GU (fig. 1) showed nearly the same irradiation sensitivity. On the other hand, specimens from plates 4 and 5 showed less irradiation sensitivity than those from plates 1 and 2. Fig. 6 shows the trend of variation in the irradiation sensitivity throughout the thickness. The data used in fig. 6 were chosen to show the trend of variation in irradiation sensitivity throughout the thickness at constant irradiation temperatures and fluences.

Figs. 7 and 8 show the dependence of ΔDBTT on the fluence. Two different straight line relationships were obtained. One for specimens from plates 1, 2 and 3 ($\frac{T}{4} - \frac{T}{2}$) and the other for specimens from plates 4 and 5 (surface - $\frac{T}{4}$). The scattering, specially in the case of specimens from plates 1, 2, and 3, where more data are available, is very small and even negligible. It is clear from figs. 7 and 8 that the irradiation sensitivity (ΔDBTT) for specimens from plates 4 and 5 (surface - $\frac{T}{4}$) is lower than that for specimens from plates 1, 2 and 3 ($\frac{T}{4} - \frac{T}{2}$). The same trend was noticed by examining the irradiation data obtained for HSST Plates 1 and 2 [6, 7] as shown in fig. 9.

Thus, the scattering in the IAEA Coordinated Programme results are explained in terms of irradiation sensitivity of specimens having different microstructure and mechanical properties as discussed previously. Figs. 6 - 9 show that considering that there are two different microstructures the scattering noticed in fig. 5 is no longer present or reduced drastically. Similarly, it can be concluded that the scattering reported before [6, 7] for HSST plates may be due to variation in the irradiation sensitivity between specimens used due to variation in their locations throughout the plate thickness.

Other minor reasons which may cause some scattering in the IAEA results could be differences in neutron spectrum between different reactors and different positions in the same reactor, variations in the methods used to measure and evaluate the neutron fluence. The Fluence data were reported in an unsatisfactory manner which does not facilitate the comparison [16].

There were also differences related to the procedures followed in each test. Only participant, 1, carried out stress relief treatment for the specimens before irradiation. Two participants, 1 and 2, carried out heat treatment for the unirradiated specimens to simulate the thermal conditions of the irradiated ones.

3. SUMMARY AND CONCLUSIONS

The results of the IAEA Coordinated Programme on Irradiation Embrittlement of Pressure Vessel Steels showed considerable scattering and discrepancies. The relation between the increase in ΔDBTT and the fluence showed a scattering band width of 65°C. In an attempt to decrease the scattering Nagel [8] used a correction factor to correct for the material state error. This resulted in reducing the scattering band to 50°C. He neglected the effect of the initial material variations on the irradiation sensitivity of the specimens.

In the present work it was shown that the specimens used by the participants can be divided to two sets different in microstructure and initial mechanical properties depending on their locations throughout the HSST Plate thickness. Set 1 contains specimens from plates 1, 2 and 3, i.e. from positions between $\frac{T}{4}$ and $\frac{T}{2}$ throughout HSST plate thickness, and set 2 contains specimens from plates 4 and 5, i.e. from positions between the surface and $\frac{T}{4}$ (fig. 2.a).

Analysis of ΔDBTT-fluence data showed that a straight line relationship was obtained for each set of specimens. The scattering in each case was small and even negligible. There is a shift of 40-60°C in the case of irradiations at 120°C and of 20-25°C in the case of irradiations at 290°C. The analysis showed that irradiation sensitivity of specimens of set 2 is lower than that of set 1. This was attributed to the variation of initial microstructure and mechanical properties.

The above noticed shift between the two straight lines explains the scattering in the IAEA Coordinated Programme results. Thus the scattering band is attributed to using indiscriminately specimens from different sets having different radiation sensitivities.

It was also noted that there were some variations between the participants related to the specimen preparation and tests procedures in addition to variations in measuring and analysing the fluence data.

In any similar work or programme it is recommended that:

1. The material used must be taken from positions that have (or nearly have) the same microstructural and mechanical properties.

2. A standard procedure must be followed by all participants. The procedures of irradiation, measuring and analysing the fluence data, and the mechanical tests must be carried out according to an agreed upon standard method or methods.

Table I

Charpy-V Transition Temperatures of Unirradiated Specimens

Participant No.	Country	Specimen location[a]	TT (°C)
1	CSSR	1 LJ	- 5
		5 LJ	- 28
2	Denmark	3 LK	0
3	FRG, GKSS	3 LV[b]	- 10
		3 PC	0
4	FRG, KFA	3 MD	18
		3 MQ	30
		3 GU	25
5	France	1-3 LQ	5
6	Japan	3,9 LZ[b]	- 6
7	Sweden	1-4 GX	5
8	U.K.	5 MN	5

a. The letters indicate the block position according to fig. 1, and digits before letters refer to the plate used according to fig. 2.

b. Specimens in the longitudinal direction, others are in the transverse direction.

Table II

Average Transition Temperature for 03 Plate

Specimen Location	TT (°C) Longitudinal	Transverse
Upper Surface	- 94.5	- 77
1/3 T	- 16	- 15
1/2 T	- 15	- 9
3/4 T	- 17	- 9.5
Lower Surface	- 72	- 66.5

Table III

Average Transition Temperature for Plates* in Each Block

Plate No.	TT (°C) Longitudinal	Transverse
1	- 15.5	- 9
2	- 16	- 9
3	- 16.5	- 9
4	- 21	- 14
5	- 28	- 20

* see fig. 2.a for positions of plates

Table IV

Charpy-V Transition Temperature of Irradiated Specimens

Participant No.	Specimen[a] location	Reactor type	Irradiation temperature (°C)	Fluence $\times 10^{19} n \times cm^{-2}$ > 1 MeV	ΔDBTT (°C)
1	1 LJ3	LWR	280	1.3	10
	5 LJ5	LWR	290	1.3	14
2	3 LK2	HWR	290	2.0	57
3	3 PC2	LWR	290	7.0	120
	3 PC4	LWR	290	0.4	22
	3 LV2	LWR	290	7.0	112
	3 LV4	LWR	290	0.4	20
	- MS3	LWR	290	1.6	49
	- MS1	LWR	290	1.6	44
4	3 MD2	HWR	290	1.1	46
	3 MD4	HWR	290	9.2	121
	3 MQ2	HWR	290	1.1	21
	3 MQ4	HWR	290	9.0	94
	3 MD1	HWR	140	0.75	126
	3 MQ1	HWR	140	0.85	120
	3 GU1	LWR	120	1.0	138
	3 GU4	HWR	120	1.5	87
	3 GU2	HWR	140	10	223
	3 GU5	LWR	120	10	185
5	1-3 LQ1-5	LWR	290	1.2	30
	1-3 LQ1-5	LWR	290	8.2	80
	1-3 LQ1-5	LWR	120	1.87	170
	1-3 LQ1-5	LWR	120	17.2	240
6	3,4 LZ3,4	LWR	277	0.85	18
	3,4 LZ3,4	HWR	280	1.3	23
	- GG3,4	LWR	277	1.5	60
	- GG3,4	LWR	277	2.9	92
	GG3,4	LWR	277	8.1	146
	3,4 LZ3,4	HWR	50	1.3	154
7	1-4 GX1-4	LWR	290	3,0-6,0	90
8	5 MN4	HWR	290	1.8	39

a= digits after letters refer to the location of irradiated specimens, others are as in Table I.

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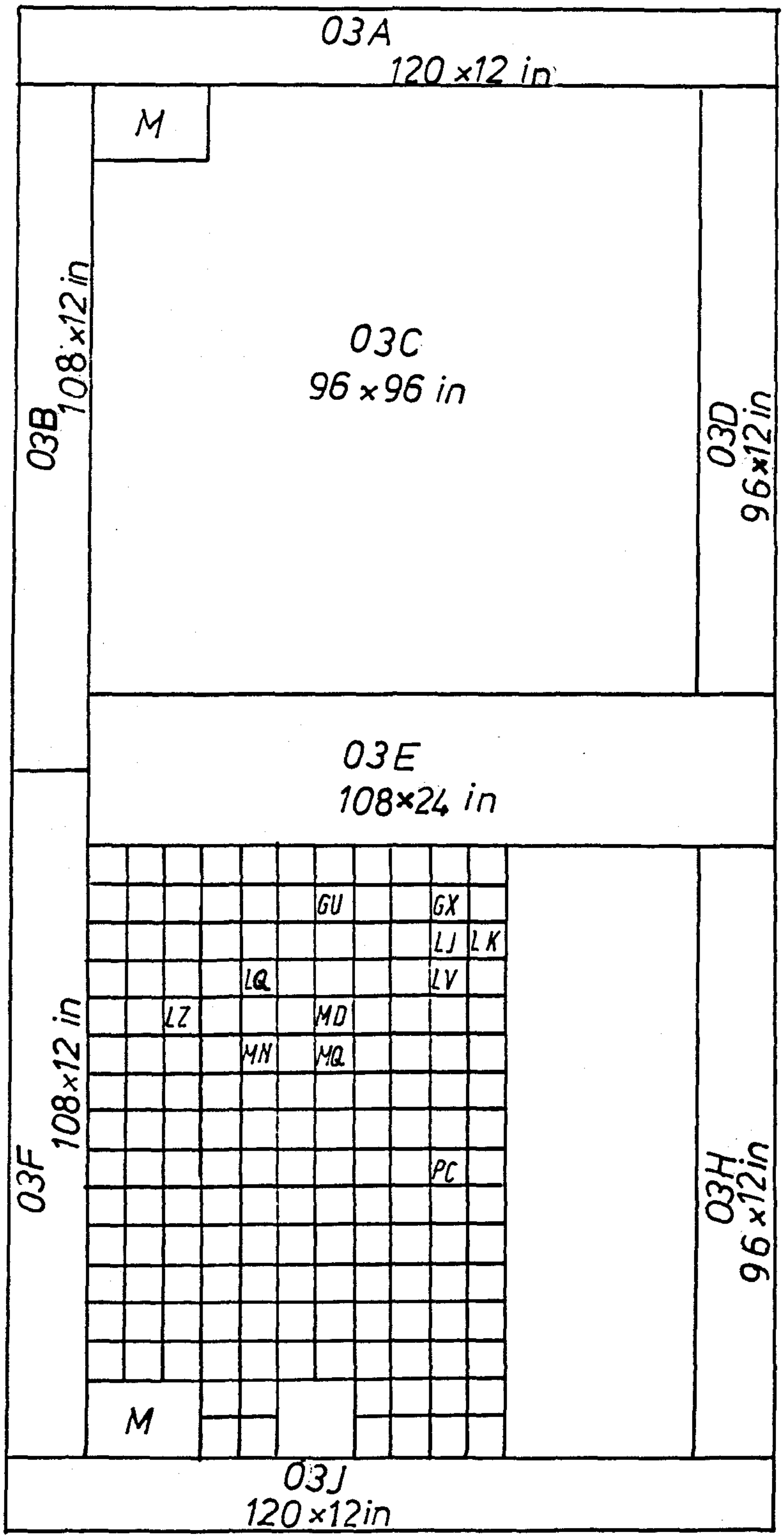


FIG. 1 LOCATION OF THE MATERIAL USED IN THE IAEA PROGRAMME, WITH RESPECT TO THE FULL-SIZE HSTT 03 PLATE. EACH BLOCK USED IS INDICATED BY TWO LETTERS. THE TESTING MATERIAL USED BY THE MANUFACTURERS IS INDICATED BY M

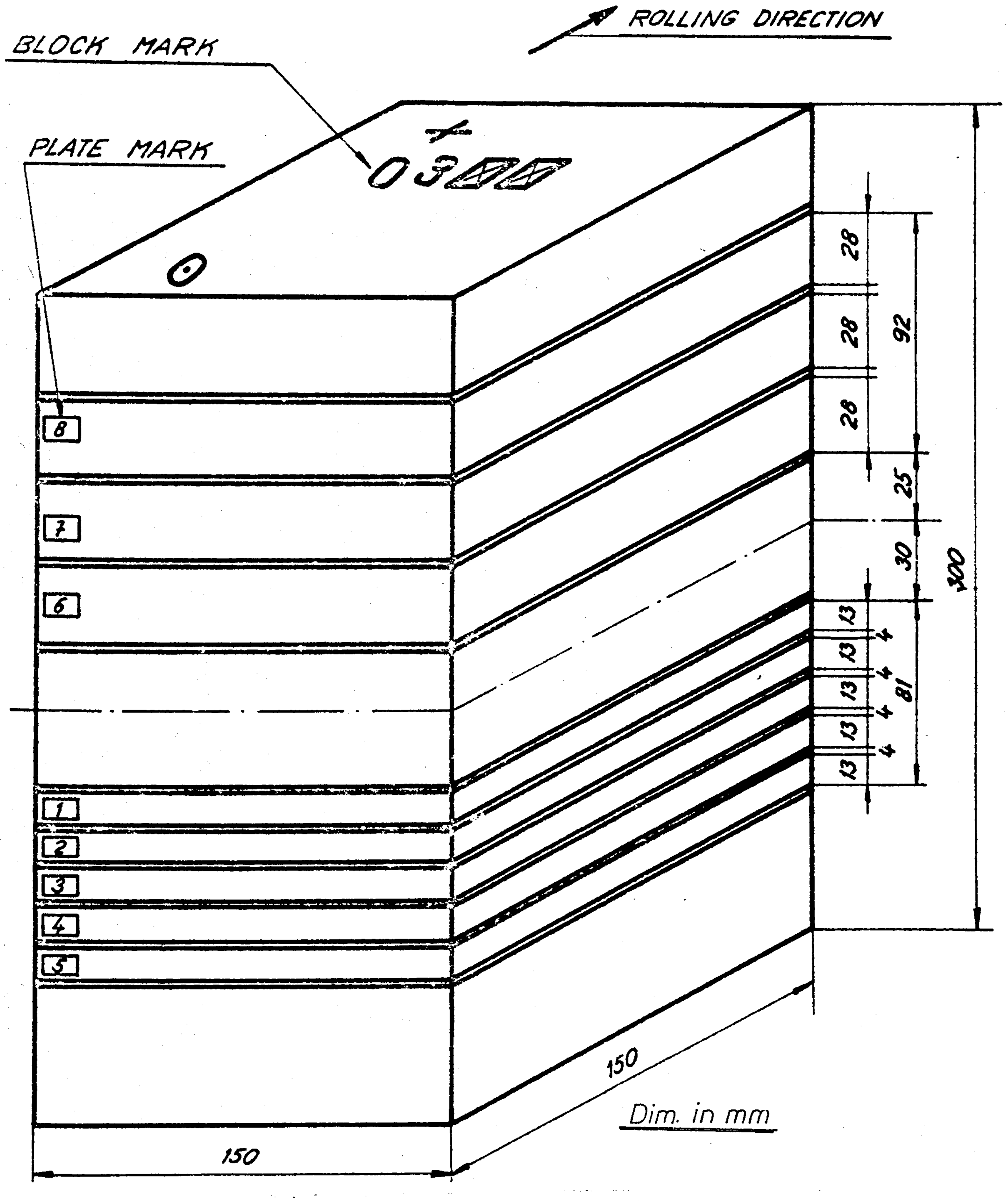
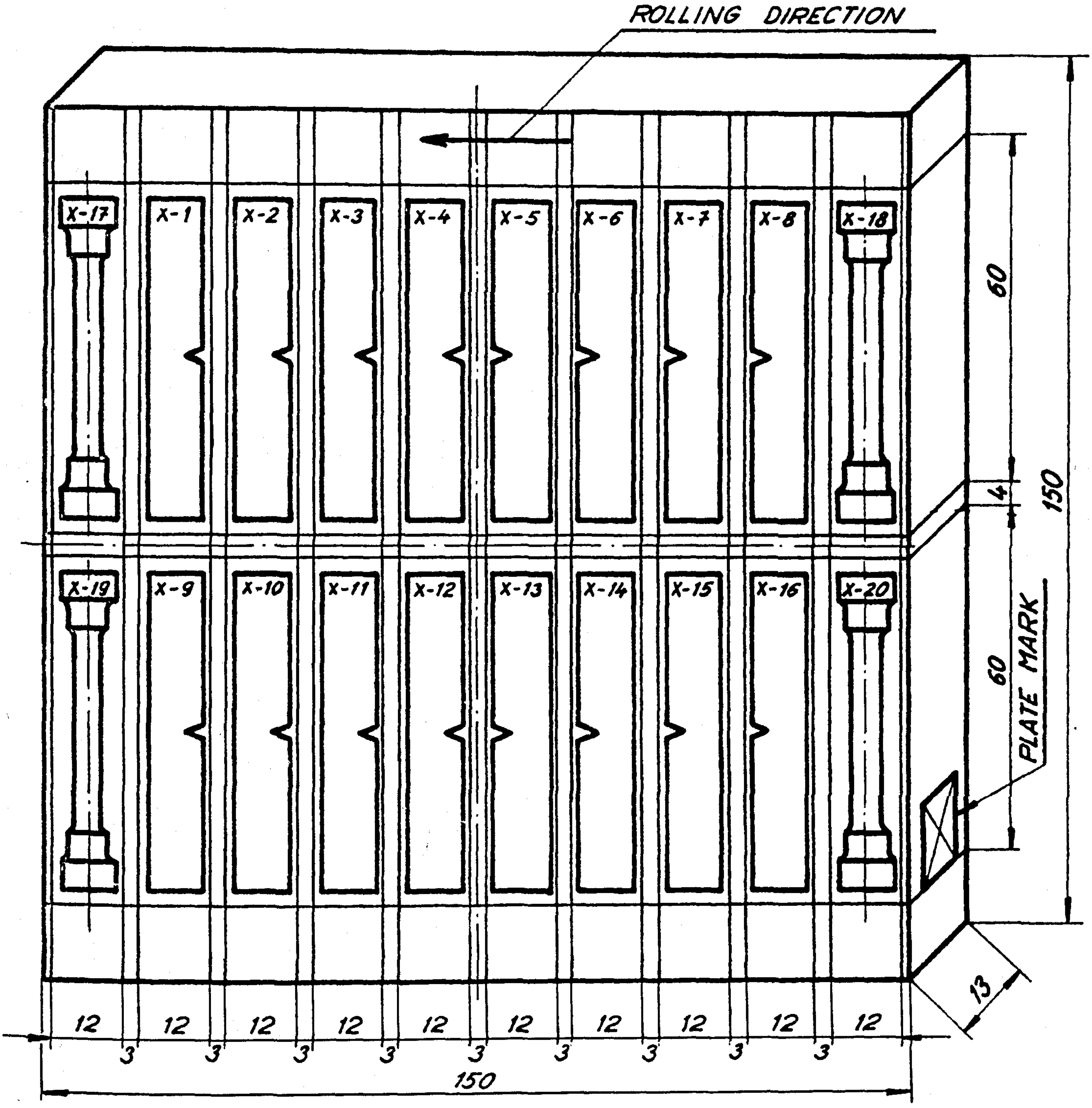


FIG. 2a CUTTING PLAN FOR IAEA BLOCKS



Dim. in mm

FIG. 2b CUTTING PLAN FOR PLATES NO.1-5

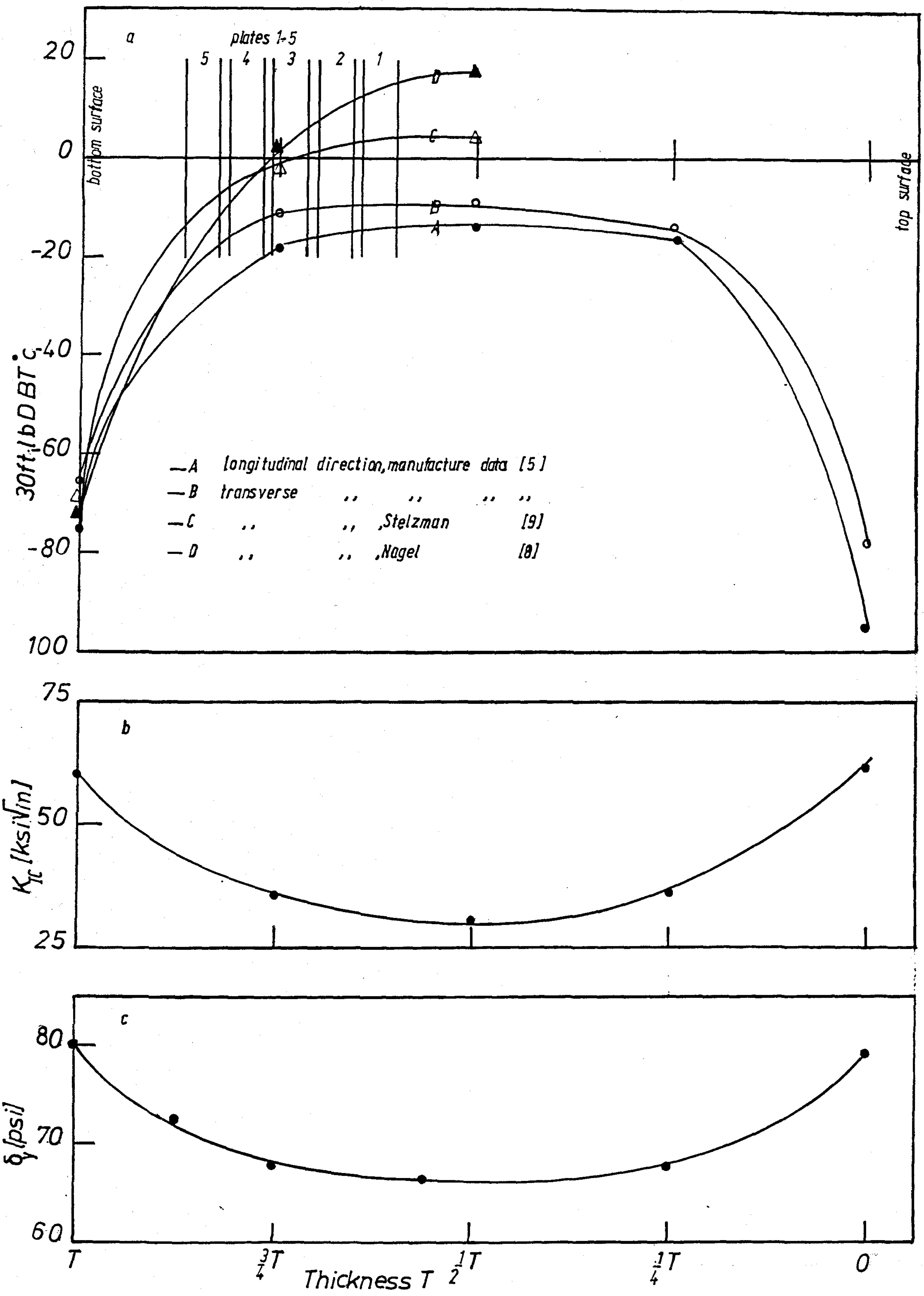


FIG. 3 VARIATION OF THE MECHANICAL PROPERTIES OF HSST PLATE 03 ACROSS THICKNESS (T): a. 30 ft.lb DBTT

b. K_{IC} at -100°C

c. yield strength

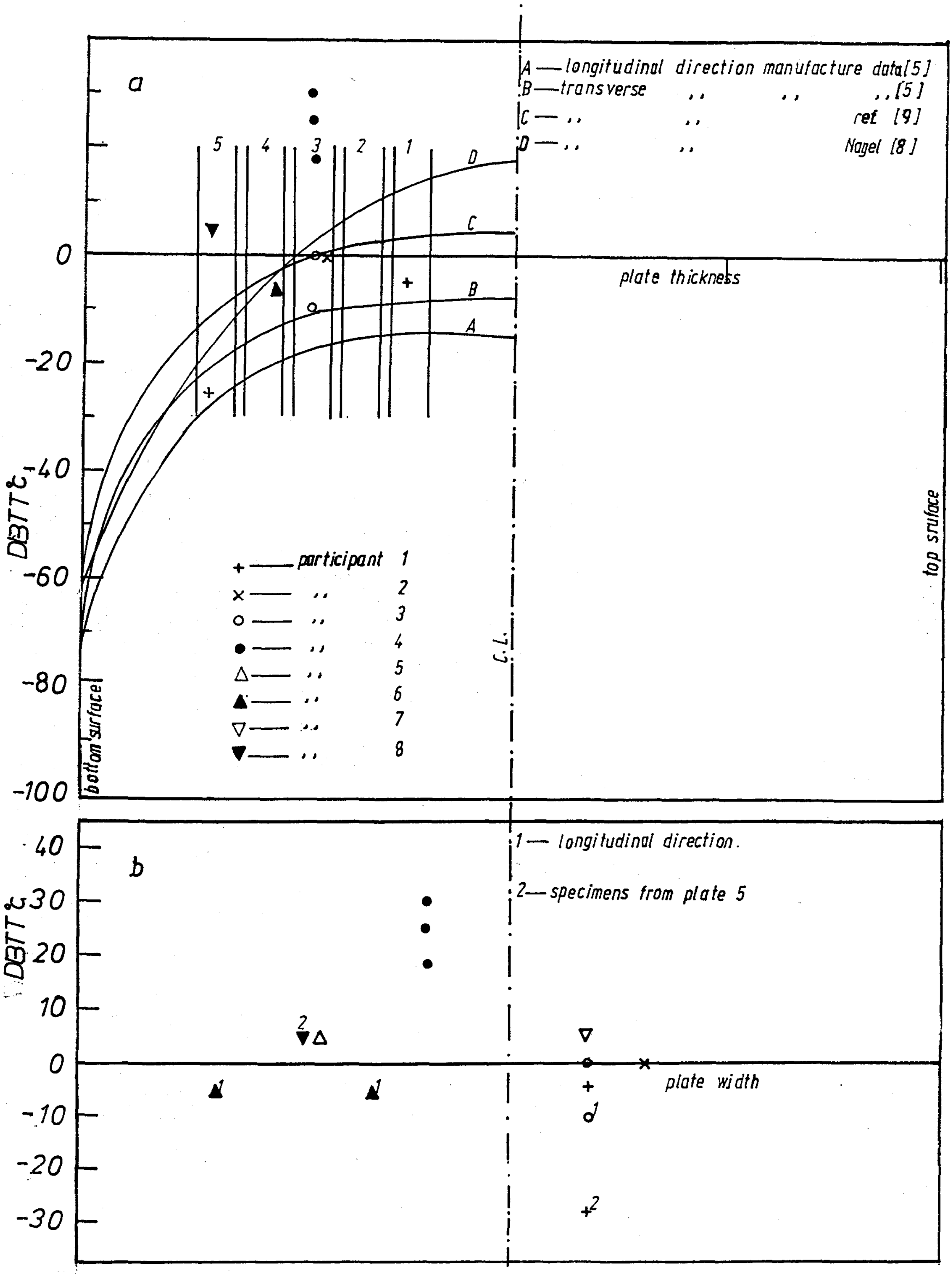


FIG. 4 VARIATION OF PARTICIPANTS' DATA ACROSS HSST PLATE 03 THICKNESS (a) AND WIDTH (b)

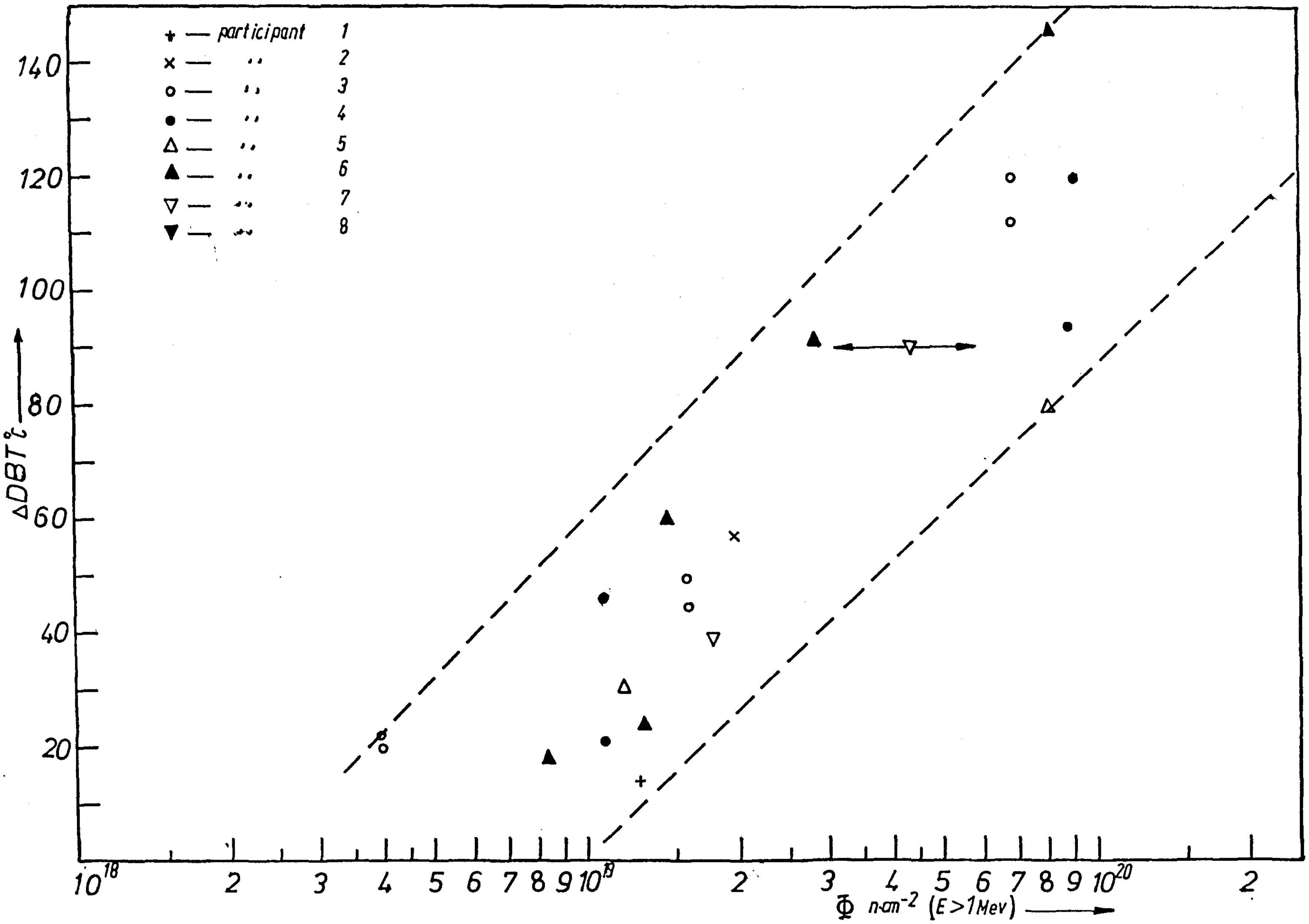


FIG. 5 VARIATION OF ΔDBTT WITH NEUTRON FLUENCE.
IRRADIATION TEMPERATURE ~290°C
IAEA PROGRAMME RESULTS [2,3]

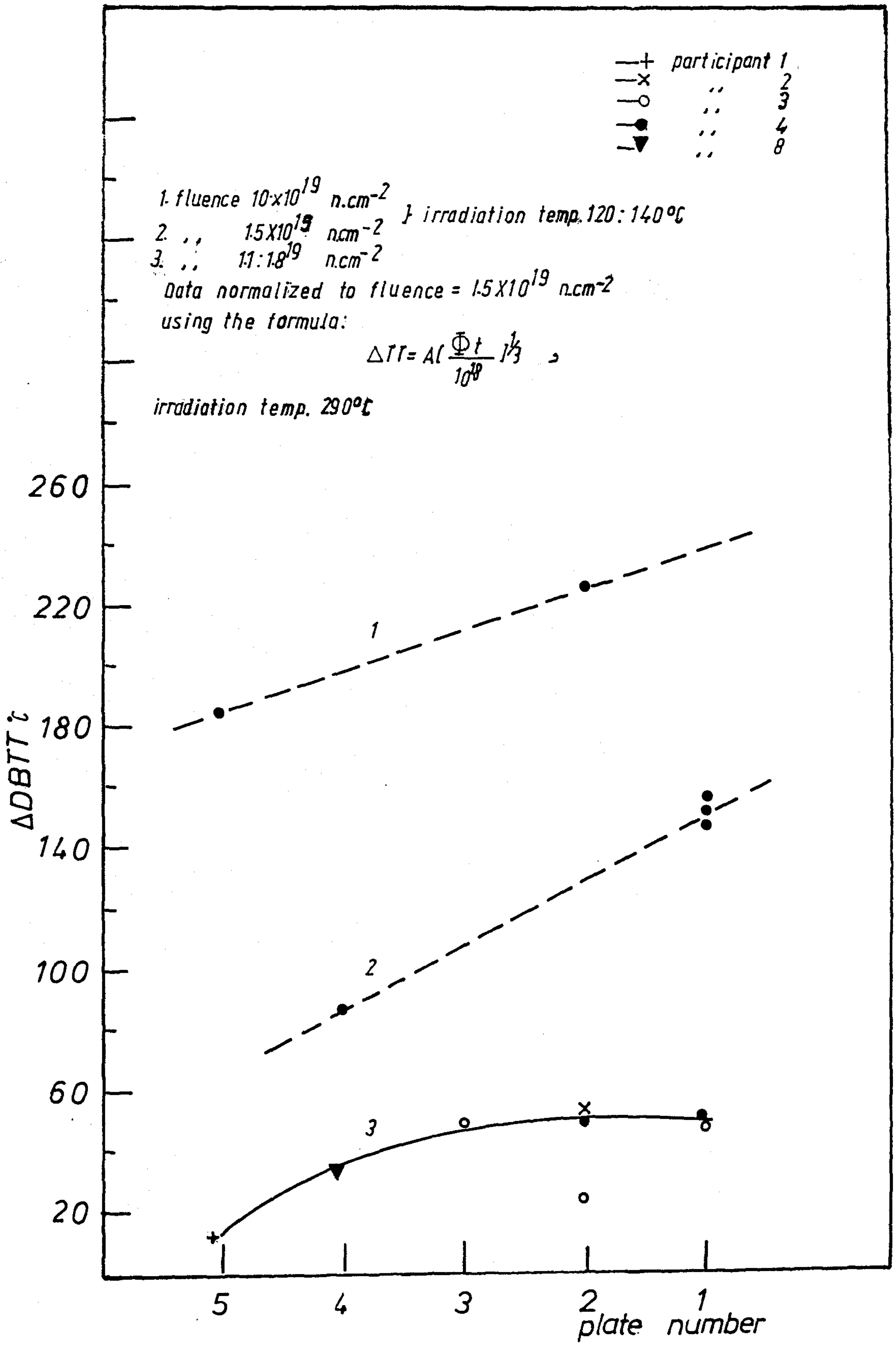


FIG. 6 VARIATION IN IRRADIATION SENSITIVITY ACROSS THE HSST PLATE 03 THICKNESS

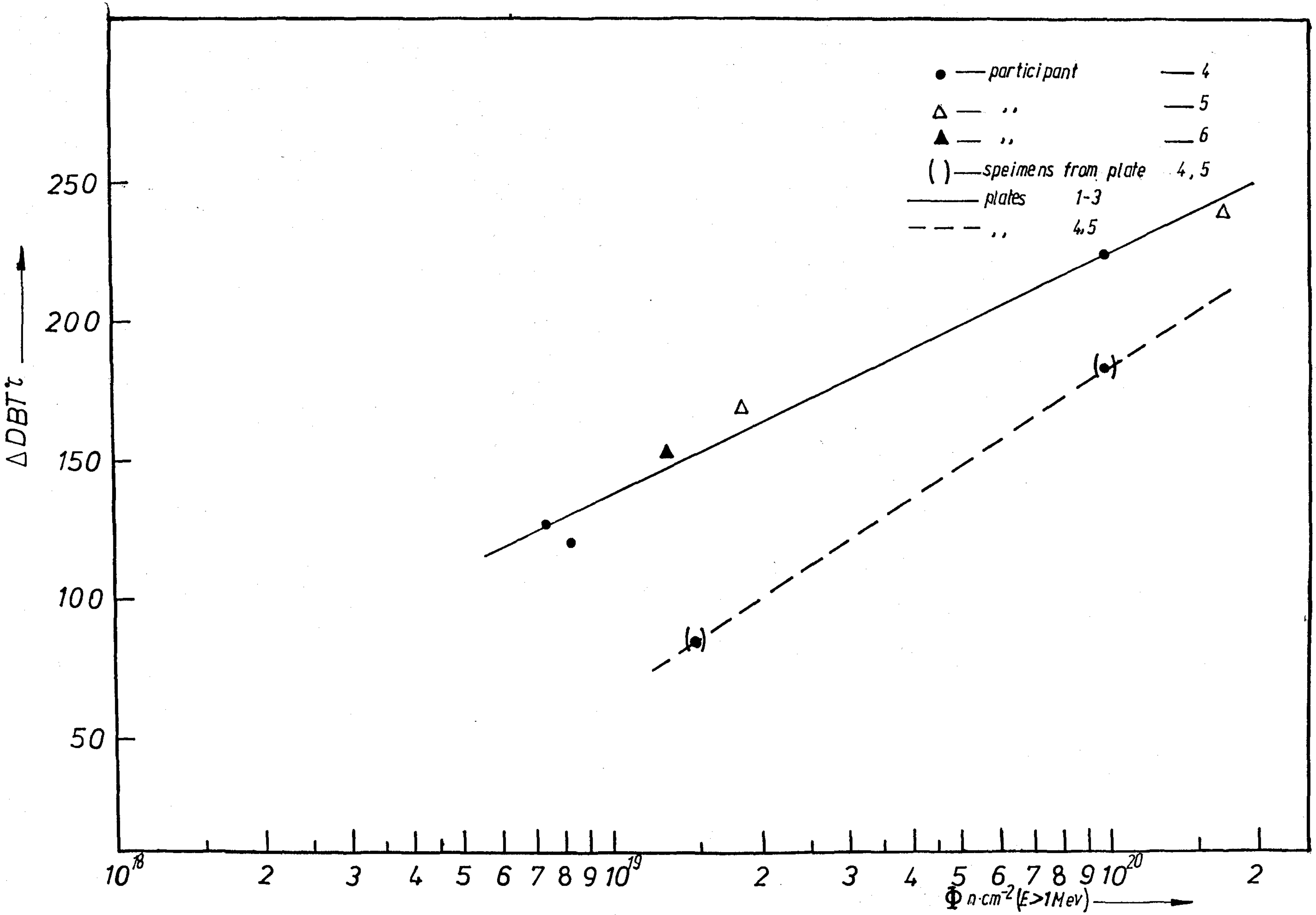


FIG. 7 VARIATION OF Δ DBTT WITH NEUTRON FLUENCE AT IRRADIATION TEMRERATURE~120°C

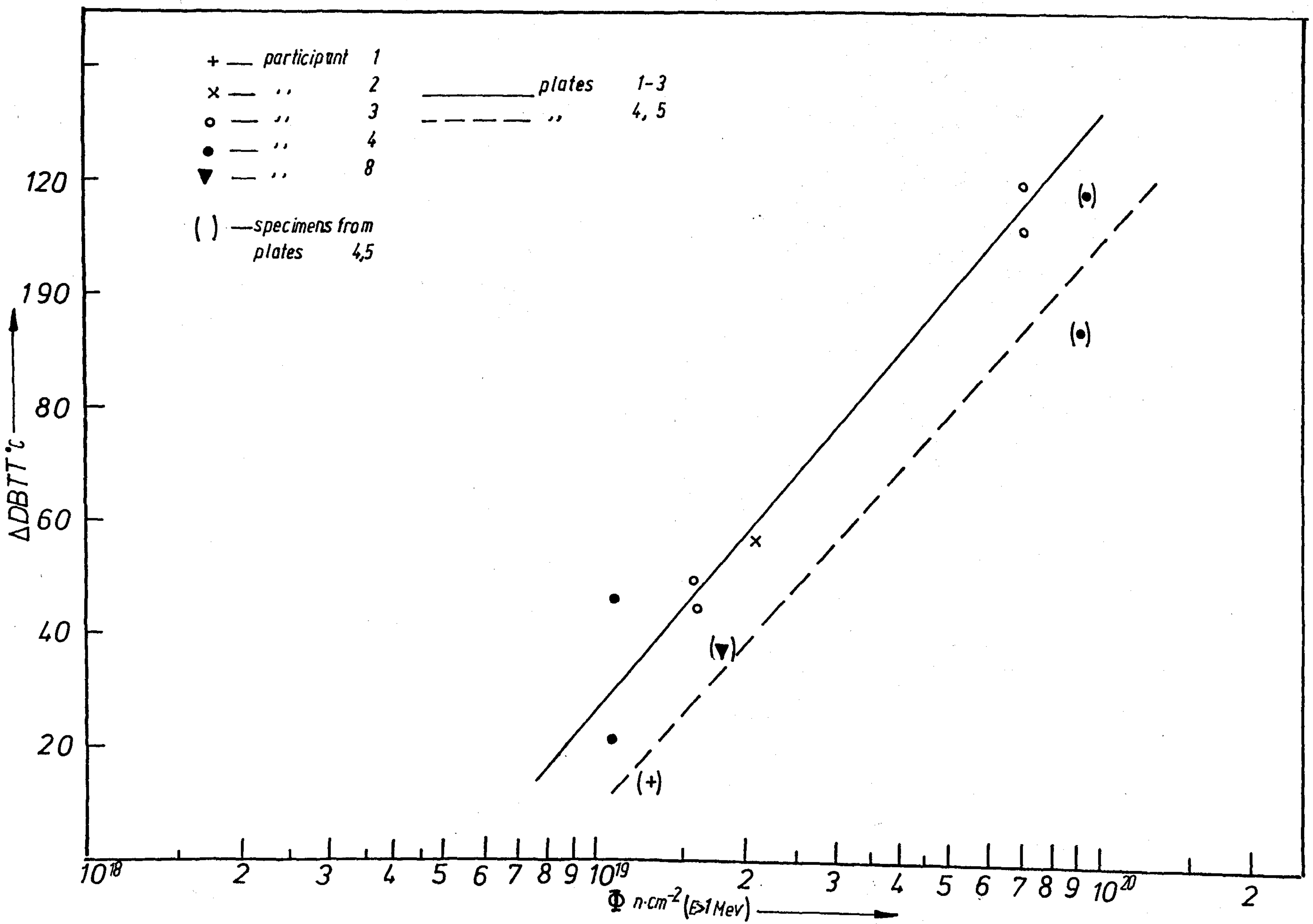


FIG. 8 VARIATION OF ΔDBTT WITH NEUTRON FLUENCE AT IRRADIATION TEMPERATURE 290°C

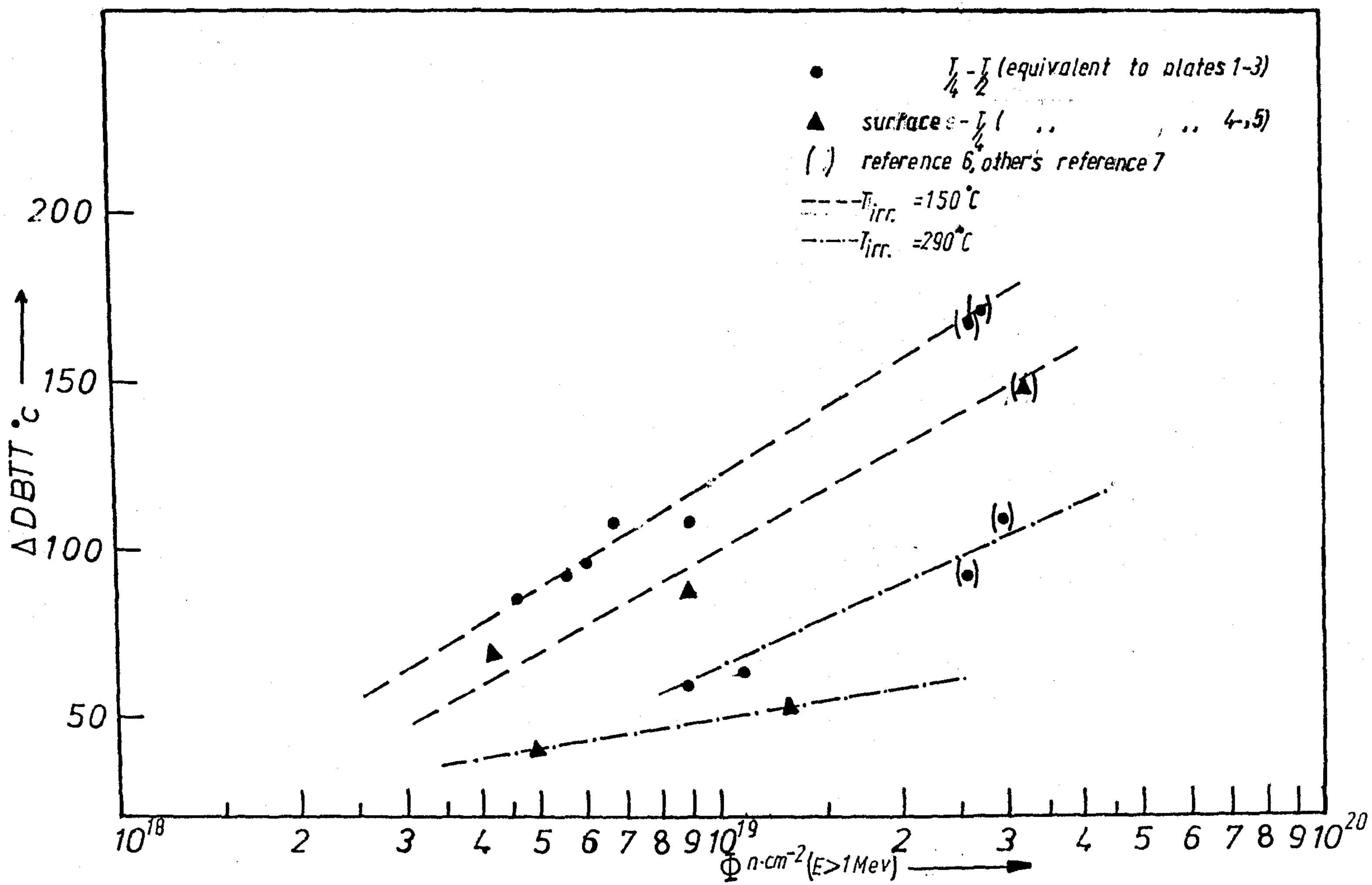


FIG. 9 VARIATION OF ΔDBTT WITH NEUTRON FLUENCE
FOR HSST PLATE 02