

International Union of Crystallography

Commission on Neutron Diffraction

An Intercomparison of Neutron Powder Diffraction Instruments*

By A. F. ANDRESEN, *Institutt for Atomenergi, Kjeller, Norway* and T. M. SABINE, *The New South Wales Institute of Technology, Broadway, NSW, Australia*

(Received 2 March 1977; accepted 27 June 1977)

As part of the activities of the Commission on Neutron Diffraction an intercomparison has been made of the performance of neutron powder diffraction instruments. This has been done by flux measurements on irradiated gold foils and by comparing diagrams obtained on standard Al_2O_3 powder samples. Data on 21 instruments in 17 countries have been included.

Introduction

In contrast to X-ray equipment, neutron diffraction instruments are mostly individually built. Since they are also placed at reactors of widely different design and use different means of beam extraction, their performance can be expected to vary considerably. As part of the activities of the Neutron Diffraction Commission it was found of interest to make an intercomparison of a group of such instruments. The powder diffraction instruments were considered a sufficiently uniform group for such an investigation. With instruments of this type one tries to obtain diagrams with high intensity, good resolution and a large peak to background ratio. It is clear that a really good resolution can only be obtained by sacrificing intensity, so a compromise has to be made. On the other hand both intensity and resolution can be improved by increasing the wavelength. This will limit the number of available reflections, and again a choice will have to be made. Which conditions to choose will in the end depend on the type of problems to be solved.

It is the intention of this survey to show what it has been possible to obtain at different type of reactors under different experimental conditions. The data should make it possible for everyone to decide whether in their case optimum conditions have been achieved, and perhaps provide a clue as to what can be done in order to improve the situation.

Data collection

Two different types of data were collected. First the participants were asked to irradiate three gold foils placed in the center of the monochromatic beam at three points along the path. One should be placed in the exact sample location, the two others in front and behind this. The gold foils had a diameter of 6.8 mm and a thickness of 0.15 mm and weighed about 60 mg. They were cut from the same piece of gold at the AAEC Research Establishment, Lucas Heights, Australia. This was to avoid any relative errors due to unknown impurities in the gold. Each foil was individually weighed before being sent out to a participating laboratory. Because of the short half life of gold (2.7 days) the irradiated foils were returned as quickly as possible after irradiation to the nearest one of three counting laboratories. These were: The National Bureau of Standards, Washington DC, USA, Australian Atomic Energy Commission, Lucas Heights, NSW, Australia and Institutt for Atomenergi, Kjeller,

Norway. The counting equipment of these laboratories had been standardized by the exchange of irradiated foils. A correction had to be made for the self-shielding of the γ -rays. This was determined by dissolving a foil in acid and amounted to 8%.

From the corrected number of disintegrations per second, D , the flux was calculated with the formula

$$\Phi = \frac{DMe^{2t}}{\sigma(1 - e^{-\lambda T})N_0m} \quad (1)$$

Here M is the atomic weight of gold, N_0 Avogadro's number, m the weight of the foil, λ the disintegration constant ($1.7828 \times 10^{-4} \text{ min}^{-1}$), σ the cross section of gold at the neutron wavelength, T the irradiation time and t the elapsed time after irradiation.

Secondly, the participants were asked to record a neutron diffraction powder pattern of a standard Al_2O_3 sample. These were sintered cylindrical pellets 13.5 mm in diameter and 16 mm long weighing approximately 9 g. They were made from Linde A grade Al_2O_3 powder isostatically pressed at 30.9×10^7 Pa and sintered for six hours at 1450°C . No preferred orientation could be detected.

Invitation to participate was sent out to 39 laboratories of which 31 declared themselves willing to take part. Each one received one standard Al_2O_3 powder sample and three gold foils for each instrument to be included. Together with the samples there were detailed instructions on how to proceed with the measurements, and a table to be filled in. Each laboratory was asked for particulars about the reactor, the collimators, the monochromator, the counter and the distances between the different parts. It was also asked for information about the beam quality and particular features like the use of filters. A second part of the table contained information about the foil irradiation, time and duration of the irradiation and the placing of the foils.

Complete sets of data was obtained from 21 laboratories. These are tabulated in Tables 1 and 2 and displayed in Figs. 1 to 4. Because of the large variations in the characteristics of the instruments a direct comparison is hard to make. However, several interesting deductions can be made. It should be kept in mind that these data were collected over a time span of three years from 1971 to 1974, that many vital changes in the reactors and instruments took place both during that period and after, and that the conditions may not be characteristic of the respective laboratories today. The main value of the data is to show what it has been possible to obtain under particular conditions at the various type of reactors.

* Prepared on behalf of the Commission on Neutron Diffraction, International Union of Crystallography.

Table 1. Reactors, monochromators, collimators and resolutions obtained

Laboratory	Reactor	Power (MW) ($\times 10^{13}$)	Φ_0 Central flux		Mono- chrom- ator	Plane	Remarks	Mosaic spread	Posi- tion	S_1	S_3	$\Delta d/d$ average ($\times 10^{-3}$)
			λ (\AA)									
Petten	HFR	45	20	2.570	Cu	111	C filter	15'	R	30'	30'	7.5
BNL	HFBR	40	70	2.410	C	002	C filter	24'	R	20'	20'	9.8
Würenlingen	Saphir	5.5	10	2.364	C	002	C filter	40'	R	20'	30'	10.0
Kjeller	JEEP II	2	2	1.863	Ge	111		18'	R	10'	20'	9.5
Risø	DR3	10	15	1.651	Ge	111		16'	R	12'(S ₂)	18'	23.6
Grenoble	HFBR, D1A	57	120	1.507	Ge	533	Guide tube	12'	R	12'	10'	2.0
Harwell	Pluto	23	20	1.480	Ge	331		7'	T	10'	24'	8.1
NBS	NBSR	9	20	1.369	Ge	111		12'	T	20'	20'	25.5
Budapest	VVRS-M	3.5	4	1.140	Pb	111		30'	R	20'	20'	18.0
Harwell	Dido	23	20	1.130	Ge	400		6'	T	23'	20'	11.0
Vinča	TVRS	6.5	6.5	1.124	Al	111		20'	R	13'	20'	14.4
Bucharest	VVR	2	2	1.117	Cu	111		19'	R	28'	23'	17.0
Grenoble	Siloé	30	24	1.113	Cu	200		30'	T	33'	10'	11.7
Pretoria	ORRR	20	20	1.091	Cu	220		35'	T	34'	21'	14.0
Mol	BR2	72	60	1.074	Cu	111		20'	R	35'	35'	22.0
Lucas Height	HIFAR	10	10	1.070	Cu	111			R	19'	27'	15.5
Puerto Rico	TRIGA	1.2	2	1.067	Cu	220		20'	T	20'	20'	17.0
Karlsruhe	FR2	44	5	1.040	Pb	111			T	20'	22'	31.2
Negev	IRR2	26	5	1.016	Cu	111			T	35'	19'	19.0
Tokai	JRR3	10	11	1.000	Ge	111		15'	T	15'	30'	26.9
Oak Ridge	HFIR	100	100	1.070	Co(Fe)	200	Polarized neutrons	12'		20'	15'	18.0

Table 2. Peak intensities and backgrounds

Laboratory	Reactor	Peak intensity (113) (cts/min)	Background (cts/min)	Peak Background	Flux at foil 1 ($\times 10^5$)	Flux at foil 2 ($\times 10^5$)	Flux at foil 3 ($\times 10^5$)	Φ_s ($\times 10^{-9}$)	Core to first collim- ator (cm)	First collim- ator to mono- chromator (cm)	Mono- chrom- ator to sample (cm)	Sample to counter (cm)
Petten	HFR	15912	132	120.5	1.76	1.18	0.89	0.6	475	55	200	40
BNL	HFBR	71087	629	113.0	85.60	88.80	86.00	12.7	350	160	80	130
Würenlingen	Saphir	4493	43	104.5	1.54	0.95	0.63	1.0	331	82	196	66
Kjeller	JEEP II	5171	74	69.9	1.72	1.47	1.21	7.5	240	35	136	44
Risø	DR3	7175	360	19.9	6.08	4.96	1.51	3.3	121	92	176	49
Grenoble	HFBR, D1A	2250	35	64.3	—	2.5	—	0.2	—	—	150	75
Harwell	Pluto	7498	115	65.2	3.05	3.33	—	1.7	243	61	114	90
NBS	NBSR	23689	790	30.0	35.67	23.00	22.76	11.5	229	110	154	69
Budapest	VVRS-M	1381	94	14.7	4.20	1.89	1.79	4.8	250	30	120	60
Harwell	Dido	17024	262	65.0	21.70	19.40	15.90	9.7	198	85	147	121
Vinča	TVRS	6604	95	69.5	12.28	5.54	4.39	8.5	350	160	80	130
Bucharest	VVR	2040	52	39.2	2.70	1.92	1.50	9.5	320	180	165	50
Grenoble	Siloé	7676	213	36.0	25.57	17.87	—	7.5	365	200	190	100
Pretoria	ORRR	6668	284	23.5	6.79	3.93	3.03	2.0	470	90	220	90
Mol	BR2	7140	161	44.3	3.72	4.09	2.18	0.7	342	90	160	95
Lucas Height	HIFAR	4492	136	33.0	10.4	3.8	2.00	3.8	130	129	164	160
Puerto Rico	Triga	590	16	36.9	1.47	0.64	0.55	3.0	215	25	195	30
Karlsruhe	FR2	6324	428	14.8	29.08	13.18	8.89	17.8	585	140	140	90
Negev	IRR2	1538	38	40.5	1.52	1.28	0.92	2.6	507	111	114	67
Tokai	JRR3	679	31	21.9	—	1.65	1.10	17.0	—	—	120	100
Oak Ridge	HFIR	11710	155	75.5	40.86	19.20	6.13	1.9	—	—	193	119

Discussion of the tables

In Table 1 are given details about the reactors, the monochromators, the collimators and the resolutions obtained. The instruments are listed in the order of decreasing wavelength. Three instruments use a very long wavelength (2.364–2.570 \AA) and a graphite filter to cut out the high-order contamination. Two of these instruments also use a graphite monochromator crystal. The monochromator crystals most commonly used are, however, copper and germanium. The Oak Ridge instrument is a polarized neutron diffractometer using a Co(Fe) monochromator crystal. It is therefore not

directly comparable with the others. In the column 'Position' is indicated whether the crystal is used in reflection (R) or transmission (T).

S_1 and S_3 are the opening angles of, respectively, the first (before the monochromator crystal) and the third (before the counter) collimator. The opening angle is given as the full width of a slit divided by its length. In the Risø instrument, which is a triple-axis spectrometer used in a double-axis mode, there is no collimator in front of the monochromator crystal, and the opening angle, S_2 , of the second collimator between the monochromator crystal and the sample is given. The instrument at the Grenoble High Flux

Reactor is placed at a guide tube, which reduces the available intensity. Here it is the opening angle of the tube which is given.

As a measure of the resolution we use the quantity $\Delta d/d$ calculated from

$$\Delta d/d = \frac{1}{2} A_{1/2} \cot \theta_B \quad (2)$$

where $A_{1/2}$ is the full width at half maximum of a peak. This quantity varies with angle and is a minimum in the vicinity of $\theta = \theta_B$. The values given in the table are averages over the angular range where $\Delta d/d$ is the lowest.

In Table 2 the peak intensity of the 113 reflection and the background are taken from the powder diffraction diagrams

corrected for counter efficiency and sample weight. The peak-to-background ratio is highest for the long-wavelength instruments, mainly because of the increase in coherent scattering with wavelength relative to the incoherent background. From the number of disintegrations per second measured on foils 1, 2 and 3 the corresponding fluxes have been calculated with equation (1). These are given in the next columns of Table 2. The flux at foil 2 is the flux at the sample position. The ratio between this flux and the central thermal flux is shown in the next column. The decrease in the flux from foil 1 to foil 3 is a measure of the divergency of the beam. The irregularities which occur are most likely due to the difficulties connected with the placing of the foils centrally in the beam. However, in the case of the instrument at Brookhaven National Laboratory where a curved graphite monochromator crystal was used, the observed larger flux at foil 2 relative to the flux at foils 1 and 3 was expected. Last in the table are given the distances between the different parts of the instrument. There are great differences, in particular, in the distance between the core and the first collimator.

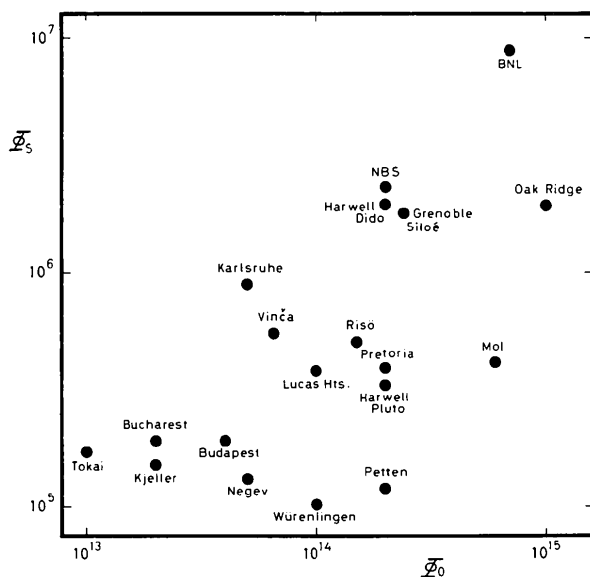


Fig. 1. Flux at sample position, Φ_s , versus central flux, Φ_0 .

Discussion of the figures

In Fig. 1 is plotted the flux at the sample position, Φ_s , versus central thermal flux. If the reactors were similarly constructed, and the efficiency of the beam extraction system the same, one should expect proportionality between these quantities. The plot shows to what extent the instruments are able to utilize the central thermal flux. There is for instance a large difference in this respect between the Brookhaven High Flux Beam Reactor which has been built specifically for beam experiments and the BR2 reactor in Mol which is a materials testing reactor. However, of even greater importance may be the intensity loss connected with an increase in the resolution. Thus the instrument in Petten has a very high resolution, whereas the instrument in Karlsruhe included in this comparison had a rather low resolution.

Fig. 2 gives the resolution of the instruments plotted as a

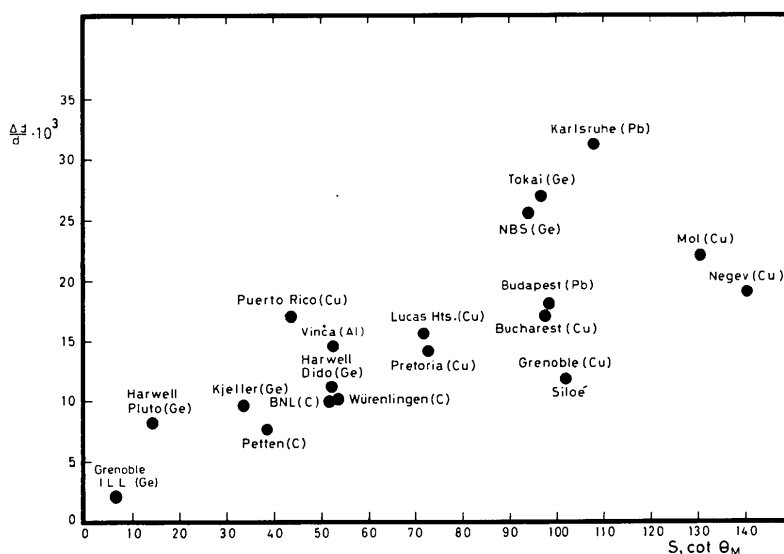


Fig. 2. Resolution $\Delta d/d = \frac{1}{2} A_{1/2} \cot \theta_B$ versus $S_1 \cot \theta_M$.

function of $S_1 \cot \theta_M$. S_1 is the opening angle of the first collimator and θ_M the take-off angle at the monochromator. Small values of S_1 as well as of $\cot \theta_M$ are expected to give good resolution. This is also the general trend of the data plotted in Fig. 2. The instrument at the High Flux Beam Reactor in Grenoble (channel D1A), which has by far the best resolution, uses a very high take-off angle ($2\theta_M = 122^\circ$). Also given in this figure is the type of monochromator crystal being used.

If now, instead of the flux measured with foils, we plot the peak intensity of the 113 reflection as a function of the central flux, we get a different picture (Fig. 3). Owing to the increase in reflectivity with increasing wavelength the instruments using a long wavelength improve their situation. Thus the Petten instrument using a wavelength of 2.57 Å has one of the highest 113 peak intensities. This shows that the increase in reflectivity is able to compensate both for the decrease in flux due to the deviation from the maximum

of the Maxwellian flux distribution and for the loss in the neutron filter. Since this instrument also has one of the best resolutions the situation would look less favourable if integrated intensities were compared.

In the last figure, Fig. 4, we give the data in the most concentrated form which we have been able to find. Here, is plotted the ratio of the integrated intensity of the 113 reflection to the central flux *versus* the resolution. This ratio tells us to what extent the neutrons present in the reactor core are being utilized to produce diffraction peaks, and this is seen in relation to the resolution obtained. The ratio is of course critically dependent on the value quoted for the central flux. The high ratio obtained for the instrument at the DIORIT reactor in Würenlingen may be partly ascribed to the monochromator crystal which is a vertically bent pyrolytic graphite crystal. Instruments using a long wavelength are also favoured in this intercomparison.

Another purpose of this survey was to check the conclusions of Sabine & Weinstock (1969). They had shown that, in a system with only a cylindrical inpile collimator, the flux at the specimen distance z from the monochromator was given by

$$\Phi(z) = \frac{\sigma R_m \alpha_m^2 l^2}{16(l+z+d)^2} \quad (3)$$

where α_m is the angular divergence of the collimator, σ is the source emissivity, R_m is the reflectivity of the monochromator, l is the length of the inpile collimator, d is the distance from the monochromator to the specimen. The relation implies that, while the flux at the collimator exit ($z=d=0$) is dependent only on the angular divergence, the rate of fall-off in flux is very dependent on the actual length of the collimator. While most collimator geometries were fairly complicated this prediction was broadly obeyed and the flux at the specimen was much higher when longer collimators were used.

Summary

It is not the intention of the authors to discuss in detail the results which have been obtained, rather it is left to the reader to extract the information he is seeking. The value of this investigation is mainly to show what it has been possible to obtain at different reactors under particular experimental conditions. This will make it possible for everyone to decide whether in their particular case optimum conditions have been achieved, or whether they should spend more time trying to improve the situation.

It seems, however, appropriate to sum up by stressing some of the more important points which were brought out in the discussion of the tables and figures. It appears that a long wavelength offers advantages with respect to intensity, to resolution, and to peak-to-background ratio. These advantages have been previously pointed out by Loopstra (1966). A long wavelength will strongly limit the number of reflections which can be observed. This is a serious disadvantage in complicated structure work, but is of less importance for magnetic problems, where the form factor decreases rapidly with scattering angle.

To obtain a corresponding increase in resolution by narrowing the collimation or decreasing the mosaic spread of the monochromator crystal a considerable sacrifice in intensity has to be made. With the profile refinement method now in current use, there seems to be less demand for a high resolution. Of considerable importance is, however, the extension of good resolution out to higher angles. This can be

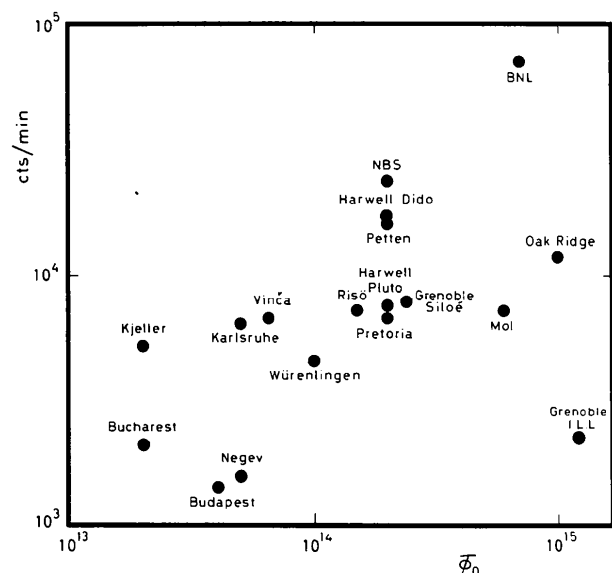


Fig. 3. Peak intensity (113) (counts per minute) *versus* central flux Φ_0 .

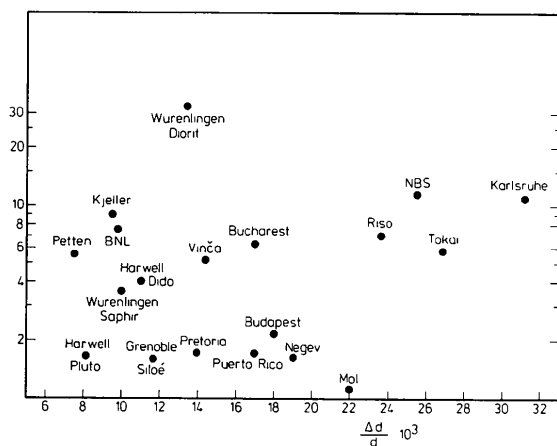


Fig. 4. The ratio of the integrated intensity of the 113 peak to the central flux *versus* resolution.

achieved by the use of a high take-off angle at the monochromator.

Since we have been mainly interested in the intensity and resolution which is available in the twice-reflected beam of a conventional double-axis diffractometer, we have not included specialized techniques on the counter side like multiple counters and position-sensitive detectors. By these techniques it is possible to increase the efficiency and rate of data taking. Thus the instrument at the High Flux Beam Reactor in Grenoble, which has an extremely good resolution out to high angles, relies on this technique to make up for the reduced intensity (Hewat, 1975).

It has been the intention of the authors to avoid making this investigation a competition between the different laboratories. We would therefore like to stress again that the data were collected over a time span of three years (1971–1974), and that they cannot be considered as representative of the situation as it is today. Even during the collection of data significant changes were made both in reactors and in-

struments. This was in fact one of the reasons why it took so long to collect the data and to get complete sets.

The authors would like to express their gratitude to everybody who has been willing to contribute their data. They are greatly indebted to Dr R. P. Ozerov, Mendeleev Institute of Chemical Technology, Moscow for the preparation of samples and for valuable suggestions. The assistance of Dr V. W. Myers, NBS, Washington DC, in measuring gold foils is gratefully acknowledged.

References

- HEWAT, A. W. (1975). *Nucl. Instrum. Methods*, **127**, 361–370.
LOOPSTRA, B. O. (1966). *Nucl. Instrum. Methods*, **44**, 181–187.
SABINE, T. M. & WINSTOCK, E. V. (1969). *J. Appl. Cryst.* **2**, 141–142.