ELECTRONICALLY FOCUSED POWDER DIFFRACTOMETERS AT IPNS-I

J. D. Jorgensen and J. Faber
Argonne National Laboratory, Argonne, Illinois 60439

ABSTRACT

Two powder diffractometers have been operated at IPNS-I since August 1981. The diffractometers achieve high resolutioin with large detector solid angles for scattering angles from ± 12 to 157° by electronically focussing the events from individual detectors in an on-line microprocessor.

INTRODUCTION

During the operation of the ZING-P' prototype pulsed neutron source at Argonne National Laboratory (December 1977 to August 1980) considerable data were taken with a time-of-flight diffractometer known as the High Resolution Powder Diffractometer (HRPD).[1] The HRPD clearly demonstrated the high and nearly constant resolution which could be obtained by the time-of-flight technique at a pulsed neutron source, but suffered from one important limitation. The long incident and short scattered flight paths rendered mechanical time-focusing techniques impractical except in back scattering. (90° detectors were provided on the HRPD, but with a much smaller solid angle than at 160°.) For this reason, the HRPD was ineffective for studying magnetic structures and indexing unknown structures where complete data are required at large d-spacings.

The two powder diffractometers at IPNS-I, the General Purpose Powder Diffractometer (GPPD) and the Special Environment Powder Diffractometer (SEPD), achieve focusing, which allows events from separate detectors to be summed, by processing signals from a large number of individual detectors in a dedicated microprocessor before data histogrms are constructed. This technique allows detector arrays of large solid angle to be constructed at any desired scattering angle. Moreover, the focusing of the instrument can be software controlled which allows the detector configuration to be optimized for a particular experiment or the initial flight path to be changed to achieve a different overall resolution.

DIFFRACTOMETER DESIGN

The GPPD and SEPD are of identical basic design but are positioned on different initial flight paths and have different detector configurations. A schematic of the instrument is shown in Fig. 1. The instrument consists of a large octagonal shielded enclosure with the sample position at the center and available detector positions from ± 12° to ± 157° at a constant radius of 1.5 meters. Final collimation of the incident beam occurs just prior to entering the sample chamber. The final collimators are cast from boron carbide and epoxy resin and are supported in an iron "wheel" 61 cm. in diameter and 8.9 cm. thick which can be rotated to select three incident beam sizes up to a maximum of 2.5 × 7.6 cm. The nominal beam size which is used for routine powder diffraction and upon which the design calculations were made is 1.3 × 5.0 cm.

The sample chamber is an aluminum tank 61 cm. in diameter and 122 cm. long. The beam enters through a thin (0.4 mm) aluminum window which is located within the collimator shielding wedge where it is not viewed by any

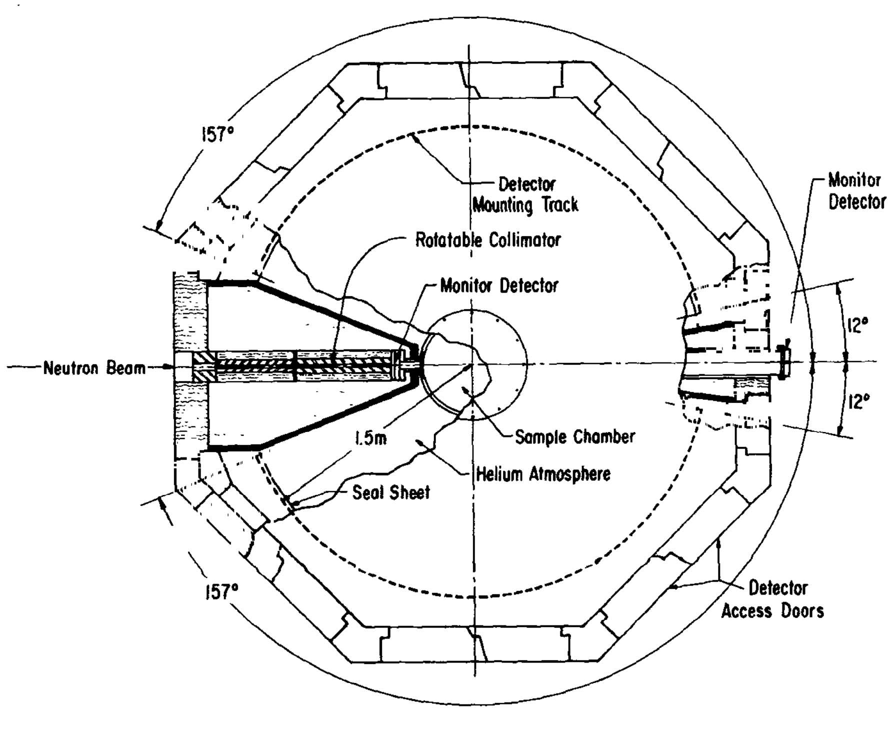


Fig. 1. Schematic layout of the General Purpose and Special Environment Powder Diffractometer.

of the detectors. The exit window is located outside the instrument shielding at the end of a 15 cm diameter pipe connected to the chamber. The sample chamber is evacuated throguh this exit pipe. The wall of the chamber has been thinned to 0.3 cm in the scattered neutron path leading to the detectors. Samples are mounted on an arm extending from the center of the top cover plate. Separate cover plates and adapters are available for mounting furnaces, cryostats, displex refrigerators and pressure cells.

Each instrument presently contains approximately 140 10-atmosphere ^3He proportional counters 1.27 cm. diameter and 38.1 cm. long. The detectors are grouped ito arrays centered around specific scattering angles as listed in Table I. The detectors and their individual preamps are supported in modules which clamp onto the constant radius detector mounting track. The

TABLE I

Performance parameters for the General Purpose and Special Environment Powder Diffractometers at IPNS-I. (May 1982).

GPPD

Incident flight path: 20 m
Useful thermal flux on sample: 2×10^5 n-cm^{-2}-sec^{-1}

2θ	d_{min}(Å)	d_{max}(Å)	$\Delta d/d$	Det. area (ster.)
152	0.2	2.9	0.0022	0.10
90	0.3	3.9	0.0040	0.086
60	0.4	5.5	0.0075	0.052
30	0.9	11	0.025	0.034

SEPD

Incident flight path: 14m
Useful thermal flux on sample: 4×10^5 n-cm^{-2}-sec^{-1}

2θ	d_{min}(Å)	d_{max}(Å)	$\Delta d/d$	Det. area (ster.)
145	0.2	4.0	0.0035	0.086
90	0.3	5.4	0.006	0.086
57	0.4	8.0	0.01	0.052
22	1	19	0.035	0.034

entire detector chamber is dehumidified to reduce electrical noise. Easy access to the detectors is obtained through the hinged, shielded access doors. Shielding is an integral part of the instrument structure and consists of polyethylene, borax, and boron carbide.

DATA ACQUISITION SYSTEM

Signals from the individual detectors are discriminated, time-encoded, and mapped into histograms in a data acquisition system built around a Z8000 microcomputer coupled to a PDP 11/34A minicomputer. (Fig. 2).[2] The Z8000

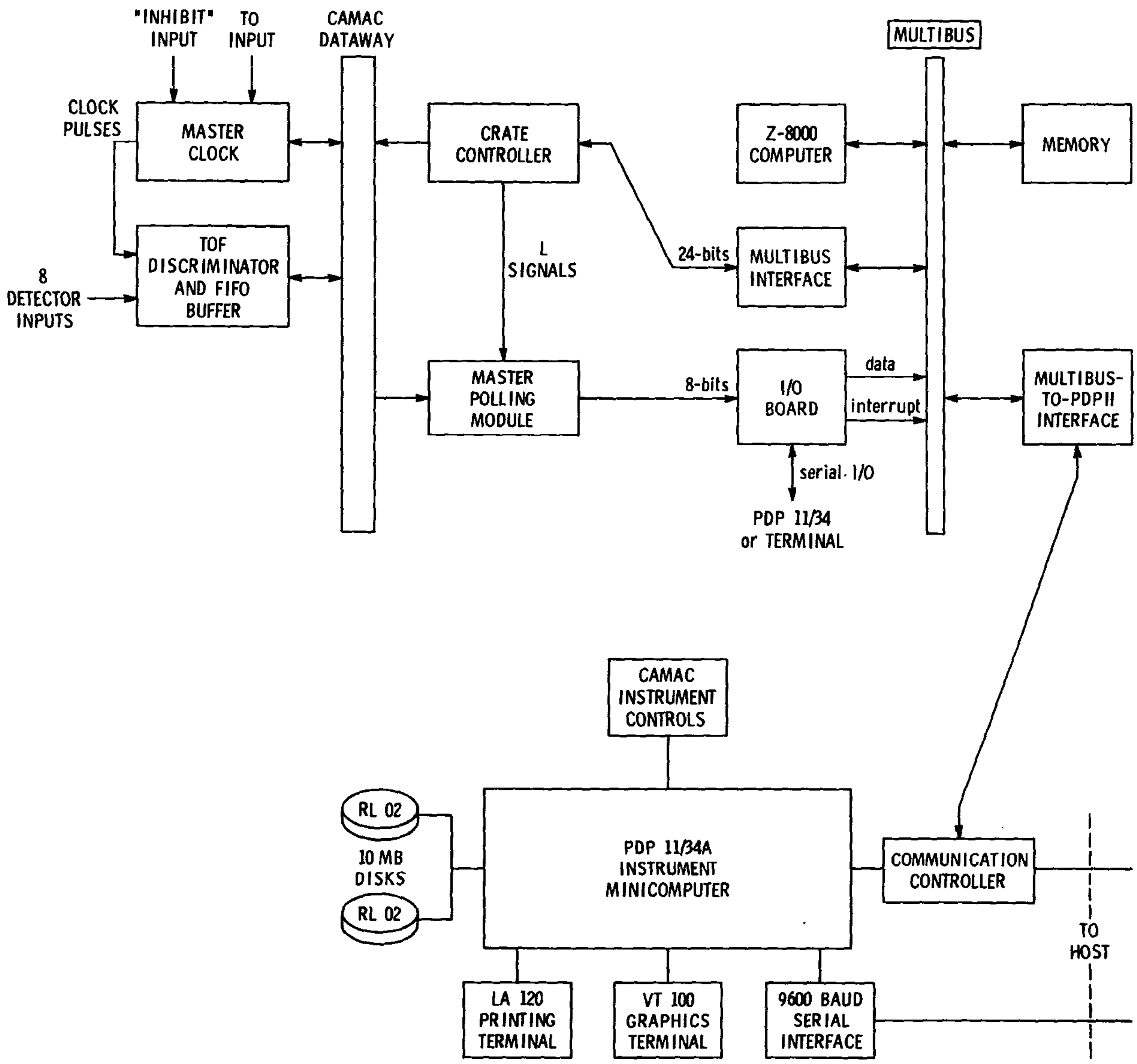


Fig. 2. Block diagram of the data acquisition system.

microcomputer is dedicated solely to data acquisition and histogram construction, and directly accesses semiconductor random accesses memory in which data histograms are stored. All other instrument functions, e.g., input, output and display of data, instrument control, etc., are handled by the PDP 11/34A minicomputer which also supervises the Z8000 microcomputer and has access to the histogram memory.

Discrimination and time-encoding occur in modules in a CAMAC system with 8 detector inputs per module. All of the time-of-flight discriminator modules are connected to a single 8 MHz master clock. Whenever one of the inputs of a TOF discriminator module receives an analog pulse within the discriminator window, a 20 bit time word (125 ns. resolution) is combined with 3 bits of input identification and loaded into a first-in-first-out (FIFO) buffer in the module. Each FIFO buffer can store sixteen 24-bit words (the 24th bit is used to indicate FIFO overflow). A polling module scans the FIFO buffers and identifies those which are over half full. The 8 bit addresses of FIFO buffers to be read are passed through a multibus interface to the Z8000 microcomputer which then reads the data from the buffer. Each event is then represented by a 32-bit word containing 20 bits of time information, 11 bits of detector identification and one overflow bit.

Before constructing histograms, the microcomputer performs the arithmetic operations on the raw time-of-flight data required to achieve time-focusing of detectors at different angles. The standard time-focusing algorithm mimics mechanical time-focusing where path length, ℓ, and scattering angle, θ, are constrained to achieve

$$\ell \sin \theta = \text{constant} \ ,$$

so that d-spacing becomes a linear function of time with a single contant,

A, for an extended array:

$$d = \frac{\lambda}{2\sin\theta} = \frac{ht}{2m\ell\sin\theta} = A\, t \quad .$$

In the case of electronic time-focusing, a pseudotime, t^*, is calculated from the measured time-of-flight, t, in order to make each detector in an extended array appear as if it were at some reference scattering angle, θ_r, and path length, ℓ_r. The pseudotime, t^*, for the detector at angle θ_n and path length ℓ_n is

$$t^* = \frac{\ell_r \sin\theta_r}{\ell_n \sin\theta_n} t = K_n t \quad .$$

The constants K_n (one for each detector) are calculated by the PDP 11/34A minicomputer during the setup of a run and are stored in a lookup table in memory where they can be accessed by the microcomputer to perform the focusing calculations.

Since the time resolution prior to focusing is 125 ns, no significant contribution to overall resolution is introduced by the focusing calculation. Having calculated the pseudotime for each event, time channels of the desired length (typically 2-20 μs) can be constructed and data from different detectors can be summed into the same time fields in memory. System software is written so that more than one historgram may be constructed from the same data if desired. The maximum data processing rate of the Z8000 microcomputer is about 3 KHz.

INSTRUMENT PERFORMANCE

Instrument performance characteristics for the GPPD and SEPD in their present configurations are summarized in Table I. During the first year of operation, the two powder diffractometers have viewed opposite sides of a 10 × 10 × 5 cm thick polyethylene moderator poisoned at the center (2.5 cm) with 0.5 mm thick cadmium. The GPPD is located on a 20 meter and the SEPD on a 14 meter indident flight path. Time-averaged thermal neutron fluxes at the sample position given in Table I are based on Monte Carlo calculations of the target-moderator assembly and have been confirmed by gold foil activation. With the large detector area available on these instruments, typical data can be collected in 6 - 24 hours depending on the complexity of the structure under study and the type of information desired.

Comparisons of unfocused and focused data show that no significant peak broadening or change in peak shape is introduced by the focusing process except at small scattering angles where the resolution of a detector becomes a strong function of angle. Figure 3 shows raw data for the end detectors

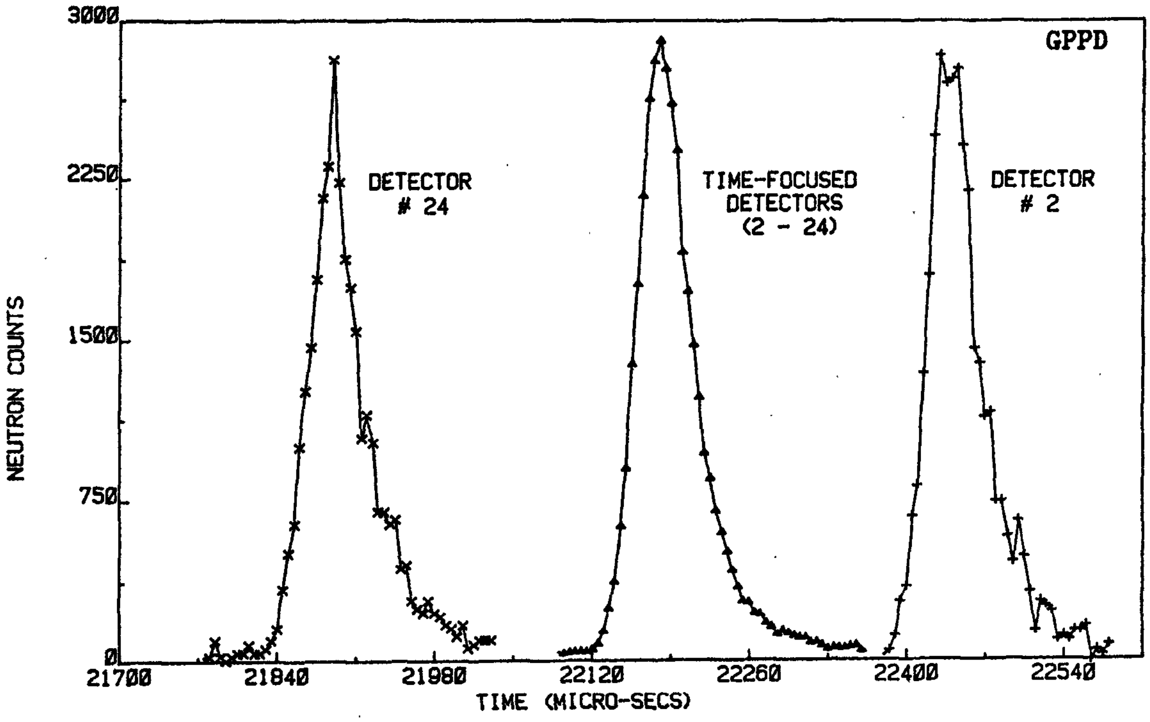


Fig. 3. Raw time-of-flight data for the first and last detectors of a 23 detector extended array on the GPPD and the electronically-focused sum for the array.

of an extended array of detectors along with the focused sum for the array.

A substantial number of data have been collected and analyzed on the GPPD and SEPD during the first year of operation at IPNS-I. Where detailed structural information is desired, the Rietveld refinement method has been used, usually concentrating on back scattering data where resolution is highest and the largest number of peaks are observed. The raw 152° data and Rietveld profile for a standard sample of Al_2O_3 run for 8 hours on the GPPD is shown in Fig. 4. The lower Q data obtained at smaller scattering angles have been successfully used to index unknown or hypothesized nuclear and magnetic structures and to extend the range of measurements on amorphous solids and liquids.

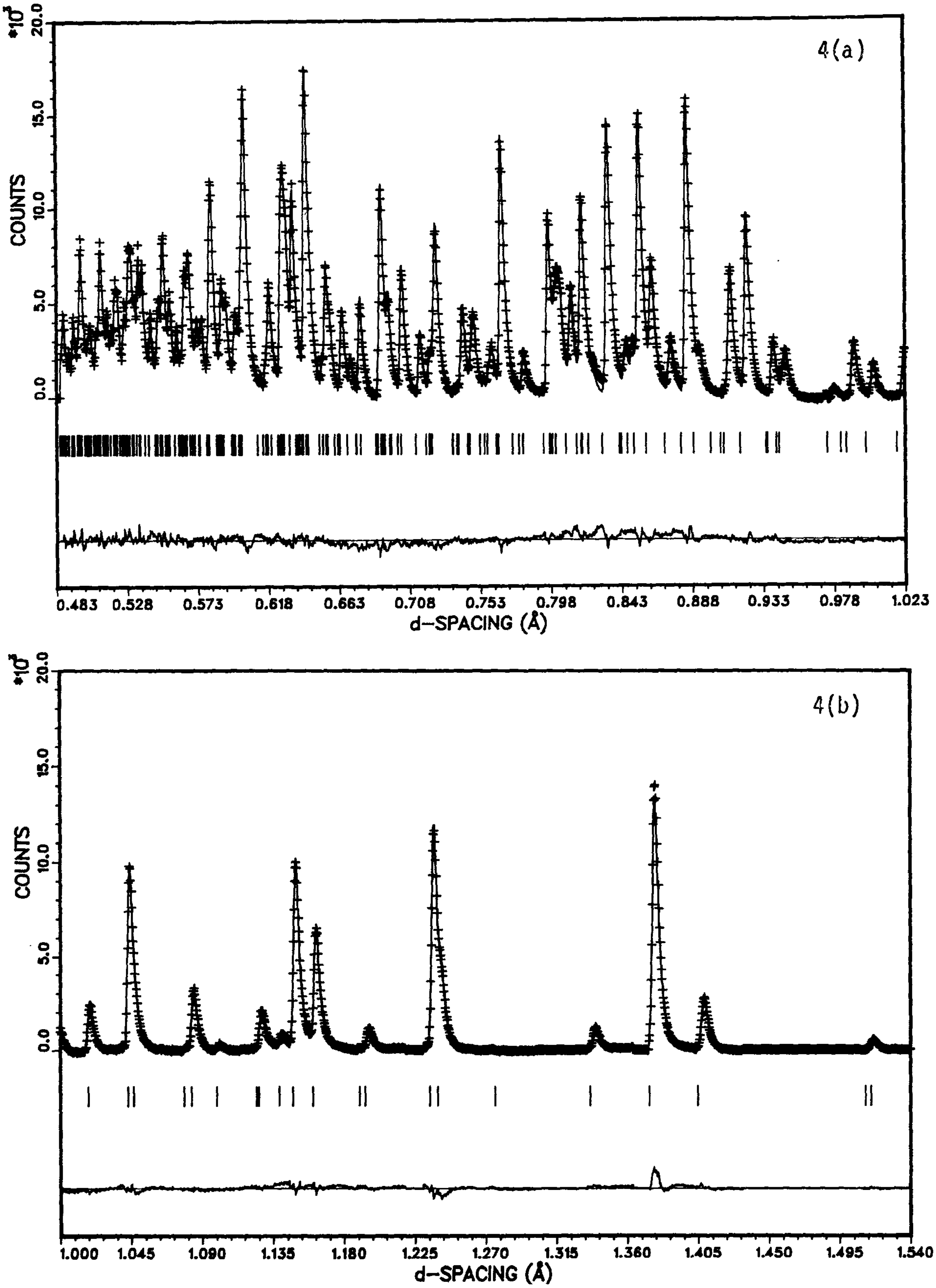
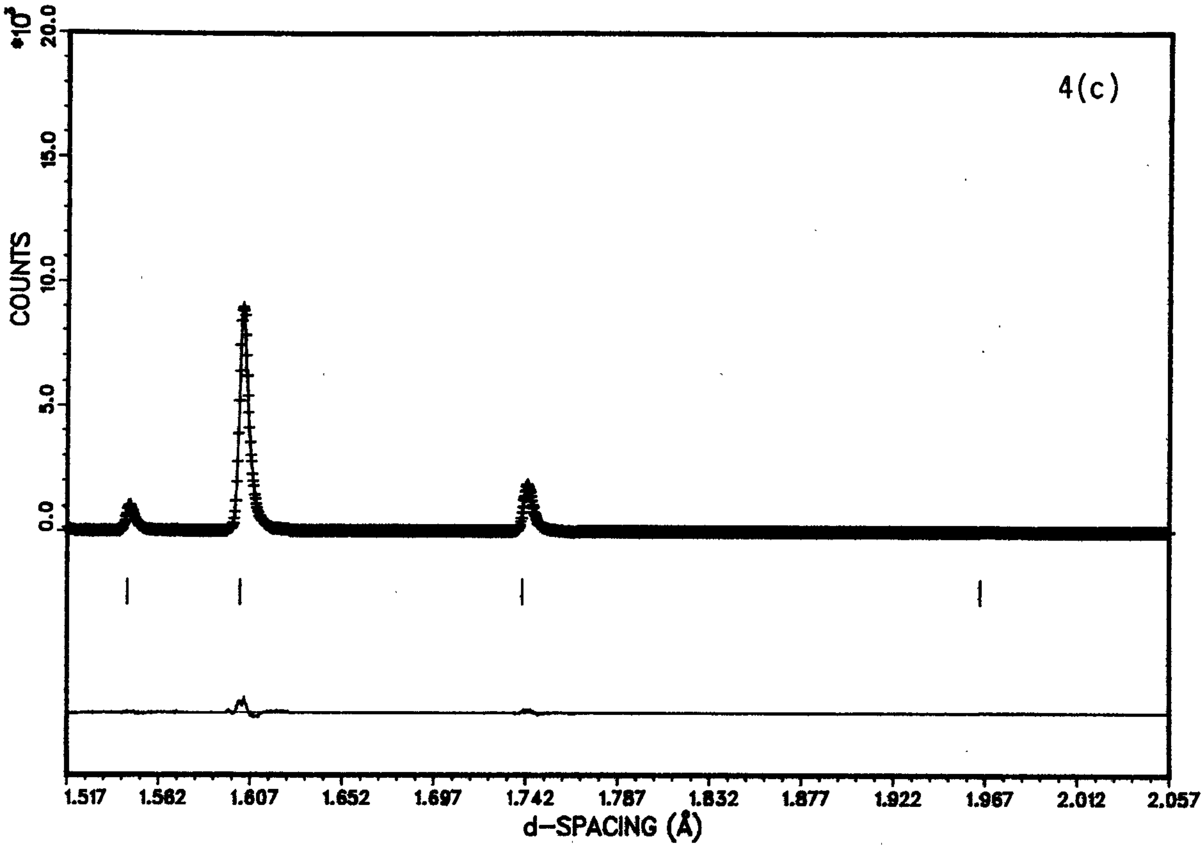
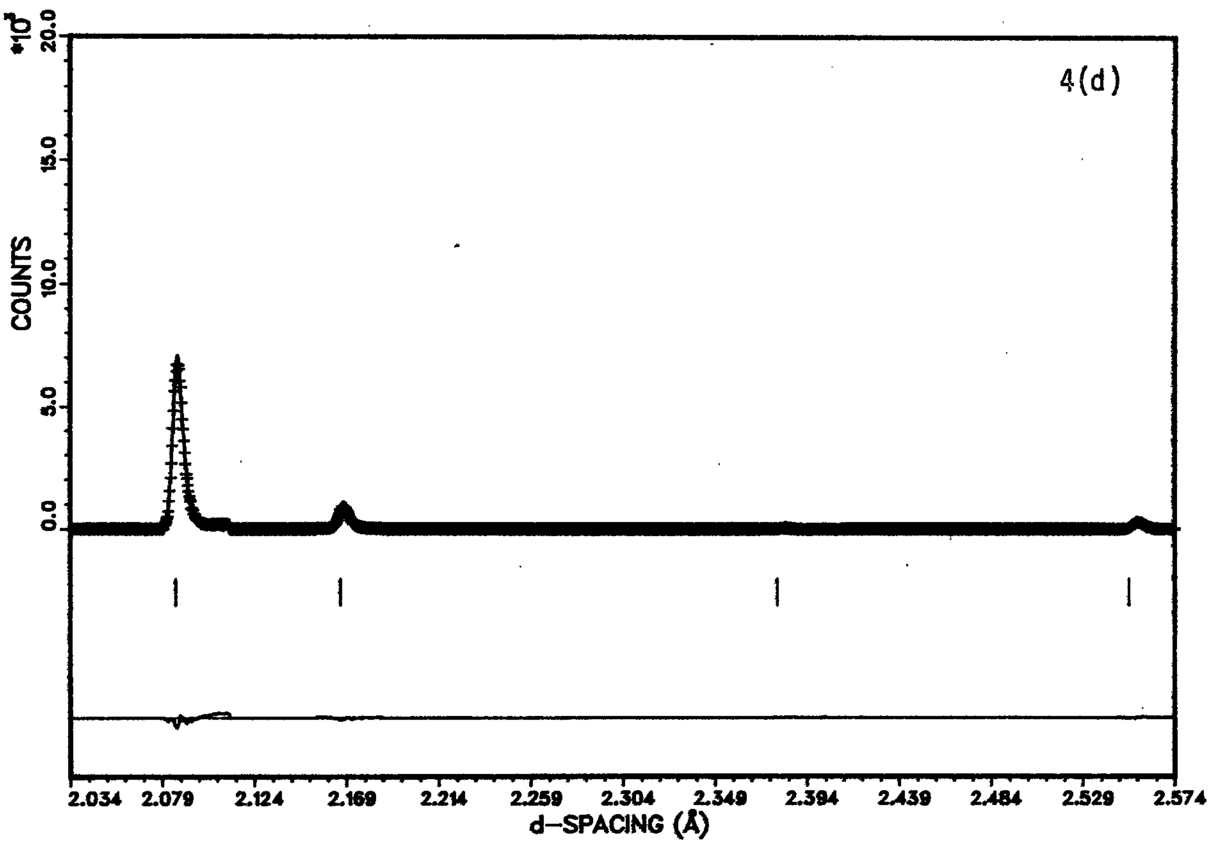


Fig. 4. Raw data (crosses) and calculated Rietveld profile (solid line) for Al_2O_3 taken at $2\theta = 152°$ on the GPPD. Tick marks below the profile indicate positions of all allowed reflections. A difference plot (observed minus calculated) appears at the bottom. Background has been subtracted before plotting.


4(c)
COUNTS
*10^3
0.0
5.0
10.0
15.0
20.0
1.517
1.562
1.607
1.652
1.697
1.742
1.787
1.832
1.877
1.922
1.967
2.012
2.057
d-SPACING (Å)


4(d)
COUNTS
*10^3
0.0
5.0
10.0
15.0
20.0
2.034
2.079
2.124
2.169
2.214
2.259
2.304
2.349
2.394
2.439
2.484
2.529
2.574
d-SPACING (Å)

SUMMARY

The GPPD and SEPD at IPNS-I clearly show that electronic focusing techniques can be used to increase the Q range and flexibility of time-of-flight diffractometers. The two IPNS-I diffractometers do this focusing during data collection with a dedicated microcomputer. This allows high time resolution before focusing and on-line display of the composite histograms.

REFERENCES

1. J. D. Jorgensen and F. J. Rotella, J. Appl. Cryst. 15, 27 (1982).

2. R. K. Crawford, R. T. Daly, J. R. Haumann, R. L. Hitterman, C. B. Morgan, G. E. Ostrowski and T. G. Worlton, IEEE Trans. Nucl. Sci. NS-28, 3692 (1981).