THE IPEN-CNEN/SP PSD NEUTRON DIFFRACTOMETER

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ABSTRACT

A new IPEN-CNEN/SP neutron powder diffractometer was constructed and installed at the 4.2 MW thermal IEA-R1m research reactor. It is an extensive upgrading of the old IPEN-CNEN/SP multipurpose neutron diffractometer. The old diffractometer was a single-detector instrument with a boron trifluoride (BF_3) detector and a flat copper mosaic single crystal monochromator. The main modification introduced in the old instrument is the installation of a position sensitive detector (PSD). A rotating oscillating collimator, placed at the entrance to the detector shielding, eliminates parasitic scattering from furnace or cryorefrigerator heat shields in the vicinity of the sample, while only reducing the scattered intensity by ca. 10%. The collimator also makes the PSD less sensitive to ambient background leaking in through the shielding entrance. Placed at a distance of 1600 mm from sample, the PSD spans an angular range of 20º of a diffraction pattern, resulting in a quite good resolution for the instrument. An extended powder diffraction pattern can be obtained by moving the detector and collecting the data in 20º segments. In order to increase the neutron beam flux at the sample position, a focusing perfect Si single crystal monochromator will be installed in the instrument. With a take-off angle of 84°, the monochromator can be positioned to produce 4 different wavelengths, namely 1.111, 1.399, 1.667 and 2.191 Å. A beam shutter will protect operator during sample manipulation or installation of any device in the monochromatic beam. In comparison to the former instrument, the new diffractometer will have better resolution and will be ca. 600 times faster in data acquisition.

Key-words: Neutron diffractometer, PSD detector, Focusing monochromator.

INTRODUCTION

A new IPEN-CNEN/SP neutron powder diffractometer was constructed and installed at the 4.2 MW thermal IEA-R1m research reactor. It is an extensive upgrading of the old IPEN-CNEN/SP multipurpose neutron diffractometer. The old diffractometer was a single-detector instrument with a boron trifluoride (BF_3) detector and a flat copper mosaic single crystal monochromator. The new instrument has several new parts. All those parts belonging to the detector system, excepting the detector shield, were imported from Instrumentation Associates Inc. (IA), 8944 Dexter Gables Ln, Dexter, MI 48130, USA, (Info@InstrumentationAssociates.com). Other parts, e.g. the main neutron shield, detector shield, open collimators and beam shutter, were mostly constructed at the IPEN machine shop. Below, a table lists the main modifications introduced in the old instrument followed by a brief description of the main parts of the new diffractometer. Some comments about the instrumentation are also presented.

Modified parts	Old diffractometer	New diffractometer
Detector	A single boron trifluoride (BF_3) neutron detector	A position sensitive detector (PSD)
Monochromator	A Cu flat mosaic single crystal	A nine-blade unit, each blade a Si bent perfect single crystal
Take-off angle ($2\theta_M$)	36°	84°
Wavelength (λ)	1.137 Å	Four different λs available
In-pile collimator	A Soller collimator	An open collimator
Incident-beam collimator	A Soller collimator	A pyramidal open collimator
Scattered-beam collimator	A Soller collimator	A rotating-oscillating collimator
Detector shield	A cylindrical shield	A pyramidal shield
Main neutron shield	A semicylindrical shield	A parallepipedal shield + the old semicylindrical shield
Beam shutter	The beam port door of beam tube no. 6 of the reactor (mainly for gamma radiation)	A shutter formed by two contrarotating drums with peripheral square channels

THE POSITION SENSITIVE DETECTOR

The position sensitive detector (PSD) [1,2] consists of eleven linear detector elements, clamped together at each end to form a rigid plane. A linear detector element is a proportional counter manufactured by Reuter-Stokes Inc. The 25 mm outside diameter stainless steel cylindrical tube of an element have a wall thickness of 0.25 mm and an active detector length of 610 mm. The anode wire is nickel chrome with a diameter of 0.015 mm. The specific resistance of the wire is *ca.* 8000 Ω. The gas fill of the counters is 8 atm of ^3He, for neutron detection, and 4 atm of Ar, for stopping the reaction products (with 0.5% of CO_2 for quenching) [3].

In operation, each end of a detector element is connected to a charge sensitive preamplifier and the detector anode is maintained at a bias of 2000 V. When a thermal neutron strikes the detector element it can be captured by a ^3He atom in the fill gas. The capture creates ^1H and ^3H products with enough kinematical energy to ionize the gas. The well-known nuclear reaction expresses such an interaction:

$$^3He + n \rightarrow {}^3H + {}^1H + 768 \text{ kev}$$

Electrons created in the detector fill gas by the reaction by-products (^1H and ^3H) are drawn to the detector anode, injecting (with gas multiplication) a charge pulse on the detector anode. The positive ions are drawn to the detector wall. This signal propagates to each end of the detector element, is amplified by the preamplifiers and is passed to analog-to-digital converters (dualADC modules). With this configuration, the linear detector element becomes a position sensitive detector. Figure 1 is a schematic drawing for the equivalent circuit representation of the detector-preamplifier assembly, for a single linear position sensitive detector element. It is possible to understand many of the characteristics of these detectors systems using a simple DC analysis. The charge injected at x divides into two currents (I_A and I_B) that flow to the left and right through the anode resistance and the input impedances of the two preamplifiers. Kirchoff’s Law and charge conservation are sufficient to show that:

$$\frac{Q_B}{Q_A + Q_B} = r = \frac{\rho x + Z_A}{\rho L + Z_A + Z_B} \qquad (1)$$

In equation (1), Q_A and Q_B are the charges that pass through the A- and B-side preamplifiers, ρ is the anode resistivity, L the anode (detector) length, Z_A and Z_B are the preamplifier input impedances and x is the neutron capture coordinate measured from the A-end of the detector element. Equation (1) illustrates the linear relationship between the charge ratio $Q_B/(Q_A+Q_B)$ and the neutron capture position, x, and is the basis for the use of these devices and position sensitive detectors [4].

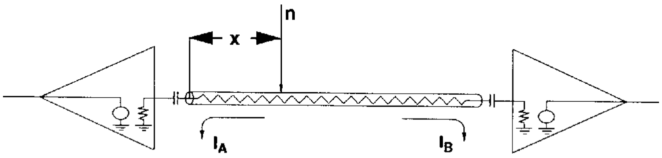


FIGURE 1. Equivalent circuit representation of the detector-preamplifier assembly for a single linear position sensitive detector element.

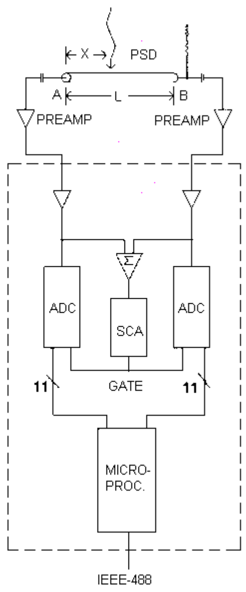


FIGURE 2. Schematic diagram of the position sensitive detector electronics for a single detector element.

Figure 2 is a block diagram of the signal processing electronics for a single linear position sensitive proportional counter detector element, like that depicted in Fig. 1. Signals from the preamplifiers are fed to a single circuit card (dualADC) which contains two nuclear pulse amplifiers, a summing amplifier, a window discriminator and two 11-bit ADCs. Signals that fall within the discriminator window cause the ADCs to simultaneously capture and digitize the pulses from each side of the detector element. The outputs of the ADCs are then used to calculate the neutron capture event position. In general, what is actually recorded is not the charges, Q_A and Q_B, but the digitized representations of the voltages produced by the system ADCs which are proportional to the charge inputs of the preamplifiers. If the ADC output, for a neutron capture event, is given by g.Q=V, where g is the product of the preamplifier-amplifier-ADC charge gain, its possible to show that:

$$\frac{V_B}{V_A + V_B} \approx r + r\left(1 - \frac{g_B}{g_A}\right)(1 - r) \qquad (2)$$

Equation (2) describes how the measured voltage quotient differs from the ratio of charges, $r = Q_B/(Q_A + Q_B)$ by an amount $\varepsilon = r(1 - g_B/g_A)(1 - r)$ which is dependent on the ratio of the electronic gains on the A- and B-sides of the detector. This deviation imposes the requirement that the charge gains of the A- and B-sides of the electronic chains be "matched" and that the gain drifts be minimized [4].

Figure 3 is a schematic diagram showing the configuration of the detector system electronics. This system effectuates control and data acquisition in the PSD neutron powder diffractometer using adequate software for this purpose. The detector, enclosed in the upper box, includes the detector elements and preamplifiers. These are wired directly to a Junction Box that distributes the signals to the dualADC position decoding electronics. The Junction Box also connects the preamplifier power supply, electronic alignment tail pulser and high voltage bias supply to the detector assembly. The peripheral control system, in turn, connects to the motion controls for the monochromator, diffractometer, rotating-oscillating collimator and ancillary equipment (refrigerators, furnaces, etc.). Figure 4 is a photograph of the PSD array constructed for the new diffractometer.

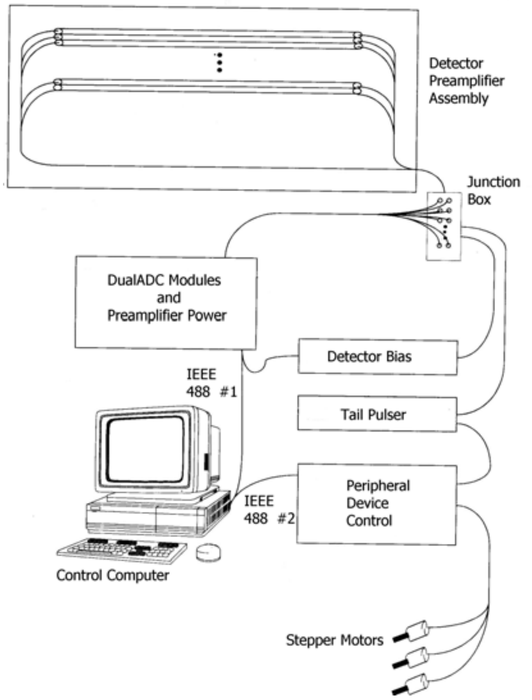


FIGURE 3. Schematic diagram of the signal processing electronics for one linear position sensitive detector element. The dualADC module is the region enclosed in the dashed line.

FIGURE 4. Back view of the PSD array. Preamplifiers are identified by the large yellow capacitors at each end of the detectors. On the left of the detector is the Junction Box.

THE FOCUSING SILICON MONOCHROMATOR

In order to increase the neutron beam flux at the sample position, a focusing silicon monochromator will be installed in the instrument [5]. The unit is made of 9 vertically stacked silicon blades, approximately 5 mm thick, 14 mm high, 190 mm length, mechanically bent in the horizontal plane and quasi-bent by segmentation in the vertical plane. All blades originate from a same plate ensuring their correct relative orientation. At 84° take-off angle the following reflections/wavelengths (Å) can be attained: 533/1.111, 511/1.399, 331/1.667 and 311/2.191. Switching between 533, 511 and 311 reflections only requires rotating the crystal around the vertical $[01\bar{1}]$ zone axis. Switching to 331 requires flipping the monochromator bottom up with a full circle stage [6].

Figure 5 shows the monochromator installed in a goniometer used for its orientation in the neutron beam. The goniometer has three movements: a rocking around a vertical axis, a tilting around a horizontal axis and a displacement along an axis normal to the monochromator surface. All movements are remote-controlled via torque transmitters electrically coupled to torque receivers. In the sixties, when the old diffractometer was built and stepper motors were not yet available, these devices were commonly employed in instrumentation. They were popularly known as ‘selsyns’. The goniometer was used for many years in the old diffractometer. It was slightly modified to support the focusing silicon monochromator.

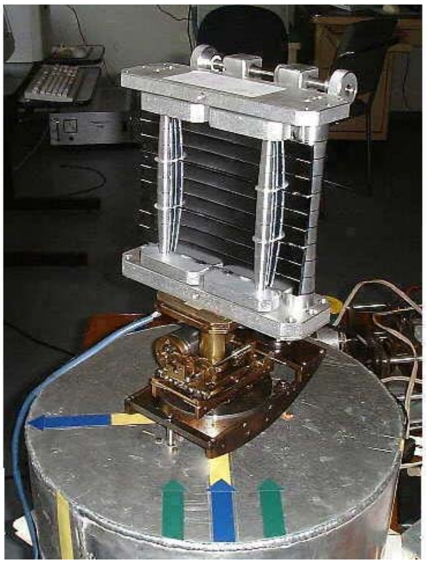


FIGURE 5. Front view of the focusing silicon monochromator. It is installed in the goniometer used for its orientation in the neutron beam.

THE DETECTOR SHIELD

To cut neutrons from the ambient background, a shield for the PSD was designed and constructed at IPEN. This shield has the form of a truncated pyramid coupled with a parallelepipedal box. The shield is made essentially of double-walled high-density polyethylene panels. Neutron trapping is provided by a mixture of paraffin + boric acid filling the panels. Two arms fixed in a large 25 in. dia. rotary table support the shield. The rotary table provides the instrument with the angular movement 2θ for the detector. Placed inside the parallelepipedal box, at a distance of 1600 mm from sample, the PSD spans an angular range of 20º of a diffraction pattern. Depending on the desired extension for the experimental pattern, a complete pattern can be obtained by a 2θ angular scanning performed with only a few steps (maximum 2θ range *ca*. 120°). A smaller rotary table (9 in. dia.), placed underneath and concentric with the larger one, provides the $\omega(\theta)$ movement. Both tables are driven by a computer controlled geared mechanism. The assembly formed by rotary tables and mechanism is the same that was used in the old diffractometer.

Figure 6 shows the detector shield already finished. At the top of the shield, inserted into a narrow opening, an aluminium plate is used for the calibration of the PSD. It is a calibration mask. The mask is covered with a cadmium foil, for neutron absorption, and has several vertical equally spaced apertures to allow the neutrons to reach the detector only in certain positions. These apertures are necessary for the calibration process.

FIGURE 6. The detector shield ready to be installed in the diffractometer

THE ROTATING-OSCILLATING COLLIMATOR

A rotating-oscillating collimator (ROC), placed at the entrance to the detector shield, eliminates parasitic scattering from furnace or cryorefrigerator heat shields in the vicinity of the sample, while only reducing the scattered intensity by *ca.* 10 %. The collimator also makes the PSD less sensitive to ambient background leaking in through the shielding entrance [7]. Both detector shield and ROC are counterbalanced by several lead bricks, placed on a basis fixed in an opposite position in the two supporting arms.

Figure 7 is a photograph of the ROC. The plates were removed in order to protect them before installation. In the upper part of the device, a stepper motor is coupled to a worm gear that makes the central region of the collimator assembly oscillate over a small angular range. Reversion is provoked by two microswitches placed at both ends of the movement. The microswitches are barely seen in the photograph.

FIGURE 7. The rotating-oscillating collimator (ROC).

OTHER COLLIMATORS

Two other collimators were constructed for the instrument: the In-pile and the Incident-beam Collimators. The In-pile Collimator is an open collimator, i. e. it has no plates. It is inserted into the beam tube no. 6 of the reactor and is used to guide neutrons toward the monochromator forming the polychromatic beam. A central square tube guides the neutrons. The Incident-beam Collimator is also an open collimator. An inner duct, with a pyramidal form, has appropriate dimensions to allow focusing of the monochromatic beam on the sample. It is placed after the monochromator, at 84° (take-off angle) from the polychromatic beam.

Figure 8 is a photograph of the In-pile Collimator separated into three parts. The larger tube is its body. It is inserted into the beam tube. On the right of the body, a tube filled with barite concrete serves as a plug to cut the radiation off during maintenance. It is inserted into the body instead of the collimator. The tube with a “cage” in the middle is the collimator itself. It is inserted into the body for normal operation of the instrument. The “cage” will accommodate a (future) sapphire filter to cut fast neutrons off from the polychromatic beam. Figure 9 is a photograph of the Incident-beam Collimator.

FIGURE 8. Parts forming the In-pile Collimator.

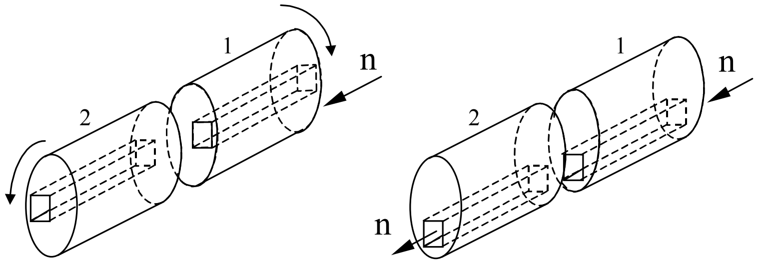
FIGURE 9. The Incident-beam Collimator.

The Incident-beam Collimator is inserted into a square hole in the Main Neutron Shield of the new diffractometer.

THE BEAM SHUTTER

A Beam Shutter was designed, constructed and installed in the Main Neutron Shield. It protects the operator during sample manipulation or installation of any device in the sample position. The beam shutter is formed by two 500 mm diameter x 500 mm length contrarotating drums with 92 x 92 mm^2 peripheral square channels. The drums are filled with barite concrete for neutron shielding and coupled each other by a pair of identical gears. The drums rotate toward opposite directions supported by pairs of ball bearings. When the channels are aligned they are in the right position to allow the neutron beam, coming from the reactor, to pass and reach the neutron monochromator. Owing to the gear coupling of the drums, moving one of them towards a certain direction the other moves to the opposite direction. Consequently, the channels go to opposite positions shutting the passage of neutrons. The shutter is driven by a 180 VDC / 0.11 A electric motor provided with a 1,100:1 reduction gearbox. A metallic gear attached to the drive shaft of the motor is coupled to a rubber toothed strap fixed on the cylindrical surface of one of the drums. They form a sort of rack and pinion coupling. Movement and positioning of the shutter is controlled by an Electronic Control Module, also designed and constructed at IPEN. The frontal panel of the electronic module has three colored push buttons (*red*, *black* and *green*) that command movement and positioning of the beam shutter. The status of the system is indicated by three colored lamps stacked on a pedestal (*red*, *yellow* and *green* for, respectively, *beam on*, *intermediary position* and *beam out*). The pedestal is placed on the top of the main neutron shield of the diffractometer, for maximum visibility.

Figure 10 is a schematic drawing of the Beam Shutter showing it in both *beam out* and *beam on* conditions.



(a) (b)

FIGURE 10. Schematic drawing of the Beam Shutter in two different situations. On the left (a), the channels are in opposite positions corresponding to the *beam out* condition. On the right (b), the channels are aligned corresponding to the *beam on* condition.

THE MAIN NEUTRON SHIELD

In order to avoid creation of a large ambient background in the diffractometer, a massive shield was designed and constructed at IPEN. This Main Neutron Shield also accommodates the Beam Shutter, the Focusing Silicon Monochromator and the Incident-beam Collimator. The main shield of the old diffractometer is now used as an additional shield, together with the new one. Both are supported by movable platforms to allow access to the beam port in the reactor wall.

Figure 11 is a photograph of the new diffractometer installed in the “beam-hole” no. 6 (BH-6) of the reactor. On the top of the shield it is seen the indicator of the Beam Shutter status. The spectrometer is also seen in the photograph, without the associated electronic instrumentation.

FIGURE 11. The IPEN-CNEN/SP PSD Neutron Diffractometer.

FINAL COMMENTS

Additional design computations for upgrading the old neutron diffractometer at IPEN have been made by M. Popovici [8]. The configuration of the old instrument was taken in order to compare its characteristics with the characteristics of a new configuration that included a PSD and a focusing monochromator. The old diffractometer used a flat mosaic Cu(200) monochromator at 36° take-off angle and Soller collimators 20´, 27´ and 71´ horizontal divergences for the in-pile, incident-beam and scattered-beam collimators, respectively. The monochromator-to-sample distance was reduced from 1.5 to 1.17 m, for the new instrument. A sample of 5 mm diameter and 5 cm height was assumed for both instruments. A PSD formed by 9 linear detectors 60 cm long was placed at 1.07 m of the sample, in the new configuration. This sample-to-detector distance results in a 30° span for the PSD. A 4 mm spatial resolution was assumed for the PSD, resulting in 150 intensity points measured simultaneously. Computed intensities at detector represent sums over electronic channels corresponding to the PSD spatial resolution. The gain in rate of data collection is the average peak intensity times the number of points measured simultaneously.

A 61° take-off angle was firstly assumed for the new configuration. At this angle, only the Si(311) reflection is of real interest because it produces neutrons of 1.66 Å which is convenient for stress scanning. The take-off angle was then increased to 84°, the angle actually used in the new instrument. The main advantage of this angle is in the multiple choice of wavelengths. As mentioned before, four different wavelengths can be obtained with a simple rotation of the monochromator (one of them needs an additional tilt). Figure 12 is a comparison between focusing and conventional configurations, i.e., between the new and the old diffractometer, regarding the peak resolution in a powder pattern. Resolution is represented in the figure by the linewidths (fwhm) of the peaks. The 2θ

angular interval assumed is approximately the interval found for the real instrument. It is worth noting that the linewidths for the focusing configuration is less than half that for the old one, in the entire 2θ interval.

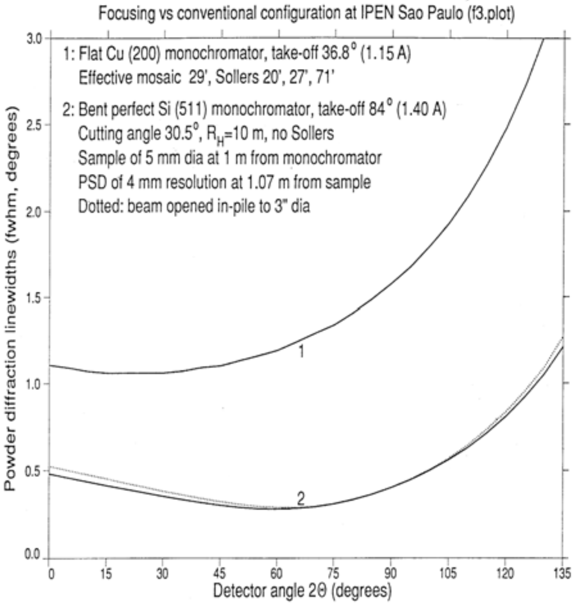


FIGURE 12. Comparison between focusing and conventional configurations, regarding the peak resolution in a powder pattern.

Figure 13 is a similar comparison for the peak intensities for both configurations. The intensity in the case of the focusing configuration is *ca*. two times that of the conventional one.

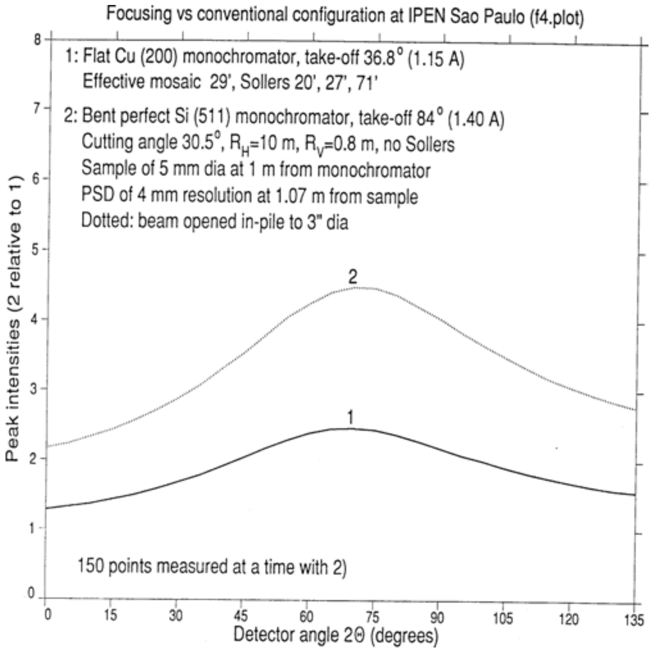


FIGURE 13. Comparison between focusing and conventional configurations, regarding the peak intensities in a powder pattern.

At this point, some other remarks must be done. The first one is that in the computations, for the conventional configuration, the take-off angle was assumed as 36.8° instead of 36.0°, the real one. Probably, this was due to a misunderstanding between brazilian and american participants of the project. Anyway, the small difference between such values does not affect the results, at least in a substantial manner. Another point is that, at the beginning of the project when the calculations were done, a shorter sample-to-detector distance was assumed in order to obtain a 30° span. Afterwards, a 20° span was preferred in order to get a better resolution for the new instrument. Distance, in this case, is 1.60 m. A longer distance certainly causes a diminution in the intensity of the diffracted neutrons, reaching the detector. On the other hand, the PSD array was constructed with 11 detectors instead of the 9 assumed in the calculations. Nevertheless, we believe both Figures 12 and 13 give a good idea about the performance that can be expected for the new diffractometer, regarding resolution and intensity of peaks. In conclusion, the new instrument will have a better resolution and will be around 600 times faster in data acquisition, when compared with the old one.

The IPEN-CNEN/SP PSD neutron diffractometer was designed mainly for crystalline and magnetic structures determination and application of the Rietveld method in multiphase analysis. However, under request, other types of application can be considered. The utilization of this new instrument will be open for the brazilian and latin-american scientific and technological communities.

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