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Design and performance of a novel neutron powder diffractometer: PEARL at TU Delft

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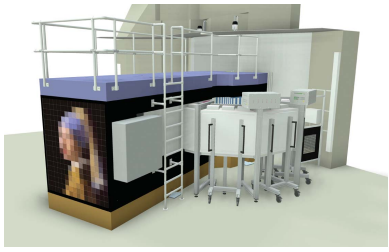
The performance of the new neutron powder diffraction instrument PEARL that is installed at the research reactor of Delft University of Technology is reported. It is based on the optimization concepts developed by Cussen [*Nucl. Instrum. Methods Phys. Res. Sect. A* (2007), **583**, 394–406], which lead to high performance competing with existing constant-wavelength neutron powder diffractometers, despite the relatively low source brightness of the 2 MW reactor of Delft University of Technology.

1. Introduction

Our understanding of the solid state of matter very much depends on the accurate determination of crystal structures at the atomic scale. Powder diffraction is now 100 years old, and together with the Rietveld (1969) method (50 years old), powder diffraction is today the standard technique in refining crystal structures in many laboratories, through X-ray powder diffraction (XRD).

Neutron powder diffraction (ND) has its own *raison d'être*, although it is often considered complementary to XRD, because for neutrons the dominant interaction is with the nucleus of an atom, rather than with the electrons as for X-rays. As a consequence of this particular interaction neutrons are more sensitive to light elements like hydrogen and lithium, and their sensitivity can be altered by isotopic substitution, leaving the physical and chemical properties of the specimen (almost) intact. In addition, elements that are next to each other in the periodic table, *e.g.* Mn and Fe, and thus difficult to distinguish with XRD can be separated with ND. Beside their nucleus-related sensitivity, neutrons can also determine magnetic moments and magnetic structures *via* the dipole–dipole interaction of their magnetic moments with the unpaired electrons in a specimen.

ND, however, is only available at large-scale neutron facilities that operate a neutron source bright enough for such instrumentation. Therefore it is not such a widespread scientific tool as XRD, for which powerful laboratory-based equipment is commercially available. The current number of neutron powder diffractometers is limited to several tens in the world and they are all 'custom-made'. Although these instruments have been conceived to offer high performance, considerable gains can still be achieved by further optimization of the layout, as we have now demonstrated with the new neutron powder diffraction instrument PEARL. This opens up the way for ground-breaking developments at the state-of-the-art and next-generation powerful neutron sources



(JPARC in Japan, SNS in the USA, ISIS in the UK and ESS in Sweden).

PEARL is installed at the 2 MW research reactor of Delft University of Technology and is currently the only neutron powder diffractometer in The Netherlands. It was designed as a medium-resolution general-purpose instrument that may compete with similar instruments across Europe, despite the low thermal power of the reactor.

2. Instrument design

The design of a neutron powder diffractometer mostly concerns the trade-off between the transmission of the beam intensity through the instrument and the instrumental contribution to the width of the diffraction peaks, which determines the extent to which neighboring peaks in a diffraction pattern can be resolved. Since the density of diffraction peaks in a pattern depends on the specimen under investigation, one could imagine building a diffractometer optimized for, for instance, cubic or triclinic crystal structures. When the specimen contains several crystal structures, this optimization criterion does not hold anymore. Being the only neutron powder diffractometer in The Netherlands, PEARL was designed to be 'general purpose', and we chose to follow the approach of focusing on cubic systems as discussed by Hewat (1975) and Cussen (2007, 2016), concerning peak density. This implies that the spacing between Bragg peaks Δd [expressed in Bragg's relation $\lambda = 2d\sin(\Theta)$, where λ is the incident wavelength, d the interplanar distance and Θ the scattering angle] is assumed to be proportional to d^3, and the optimization by Cussen aims to minimize the maximum value of $\Delta d/d^3$ over the whole measurement range Θ_M (Cussen, 2007). Optimizing for a triclinic structure would have required an even larger value for Θ_M which would have posed severe technical problems in shielding design. The optimization was partially done numerically, and an analytical approach was found only recently (Cussen, 2016).

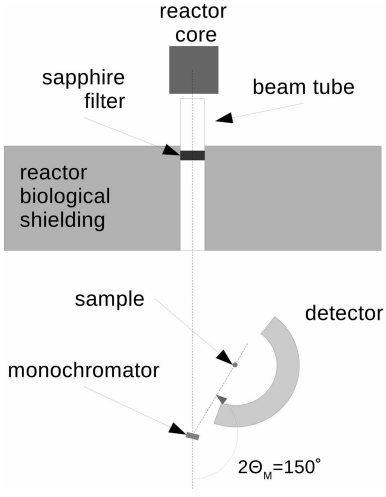


Figure 1
A top-view sketch of the instrument. The monochromator reflects and focuses neutrons from the reactor onto the sample. A multi-pixel one-dimensional detector registers the diffraction pattern of the sample.

The resulting instrument design was then simulated using the *McStas* (Willendrup *et al.*, 2004) Monte Carlo software package, producing the powder diffraction pattern of $Na_2Ca_3Al_2F_{14}$, a cubic crystal with a reasonably large cell parameter of 10.3 Å. Owing to budget restrictions and for shielding purposes, PEARL has only one monochromator take-off angle $2\Theta_M$ (Fig. 1).

The available beamport for the instrument has a diameter of 160 mm (Fig. 1), which has a direct view to the core of the reactor. A focusing Ge monochromator is located at approximately 7 m from the reactor core in backscattering geometry and at a fixed take-off angle of $2\Theta_M = 150°$. By rotating the monochromator around its vertical axis it is possible to select different reflections and associated wavelengths λ, such as the 533 reflection for $\lambda = 1.67$ Å, or the 133 and 733 reflections for $\lambda = 2.51$ and 1.33 Å, respectively. The monochromator consists of 24 crystals (currently 22 installed), each of which is built up out of stacks of 25 hot-pressed wafers (Lebech, 2001) with a rocking curve width of approximately 29′′ FWHM. PEARL has no collimators, and as such the collimation is defined by the beam-port dimensions, the monochromator size (300 mm high, 50 mm wide), the sample size (typically Ø 6, 8 or 9 mm and 50 mm high) and the detector pixel size (200 mm high, 2.1 mm wide). The monochromator–sample distance is 2000 mm and the sample–detector distance 1145 mm.

As a result of the direct view to the reactor core, the shielding of fast and epithermal neutrons was a point of attention in the design of the instrument, in particular because the backscattering monochromator geometry allowed for less than 60 cm of shielding material between the beam emerging from the reactor (core–monochromator line of sight) and the sample. Firstly, the fast neutron background is reduced by an order of magnitude using a 10 cm thick sapphire (Al_2O_3) single crystal, positioned upstream in the beam port. It has a transmission of 0.84 for $\lambda = 1.67$ Å neutrons. The background radiation is further absorbed by a compact shielding consisting of layers of paraffin, steel, boron rubber, polyethylene and heavy concrete. This turns out to be a very efficient solution as 85% of the current background level of 0.35 counts per second per pixel can be shielded if cadmium with a thickness of 1 mm is positioned in front of the detector pixels, proving that fast or epithermal neutrons are not a concern for future improvements.

3. Sample environment

The maximum diameter for the sample environment on PEARL is Ø = 800 mm, defined by the shielding. Currently, experiments are done at room temperature with a vacuum vessel at the sample position. In the near future the instrument will be equipped with a cryostat (T = 1.5–300 K), a cryo-furnace (T = 4–800 K) and pressurized gas loading capabilities.

4. Position sensitive detector

The powder diffraction patterns are collected by a novel ^{6}LiF–ZnS:Ag scintillator detector (developed by authors GJS, EMS and NJR) that covers the scattering angles $10.4 < 2\Theta_s < 158^\circ$ without gaps. It consists of 1408 pixels with a resolution of 2.1 mm and is based on a concept developed at the ISIS neutron scattering facility (http://www.isis.stfc.ac.uk), being developed as a collaboration between the ISIS neutron scattering facility and Delft University of Technology. The detection efficiency is 68% for 1.8 Å neutrons. The scintillation light from neutron absorption is transported to photomultipliers with wavelength-shifting fibers and analyzed by field-programmable gate array (FPGA) electronics that simultaneously decode the fiber encoding. Using fast electronics, it is possible to distinguish neutron- from gamma-generated scintillation light and thus discriminate neutrons from gamma rays with a ratio better than 1 p.p.m. (Sykora *et al.*, 2012). This outstanding performance was tested under several conditions including simultaneous strong neutron and gamma irradiations and paves the way for several other reactor-based applications. Indeed, up to now ^{3}He-based neutron detectors have been preferred over scintillators because of their inherent low sensitivity to the gamma radiation background generated by the neutron source and at the instrument. The high performance and relatively low cost of the PEARL detector marks a breakthrough in this field as it offers high position sensitivity and a competitive low-cost alternative to ^{3}He-based neutron detectors.

5. First results

The performance of PEARL has been assessed by a series of experiments, and Fig. 2 shows a typical diffraction pattern measured for one hour from 1.22 g of Al_2O_3 powder. The sample was loaded in a Ø 6 mm and 50 mm high thin vanadium sample can. The data were analyzed with the *GSAS* Rietveld refinement software package (Toby, 2001) with the pseudo-Voigt resolution profile number 2 (Howard, 1982). The contribution of the vanadium sample can (0.15 mm thickness) is a flat background of 0.15–0.20 counts per second.

The *GSAS* fit also yields the angular resolution of the instrument (Fig. 3). PEARL was designed as a medium-resolution diffractometer ($\delta d/d > 2 \times 10^{-3}$), so that it can collect diffraction data in a time span competitive with the existing diffractometers in Europe. The actual resolution, at best $\delta d/d = 1.5 \times 10^{-3}$, is slightly better than the 'medium-resolution' design criterion.

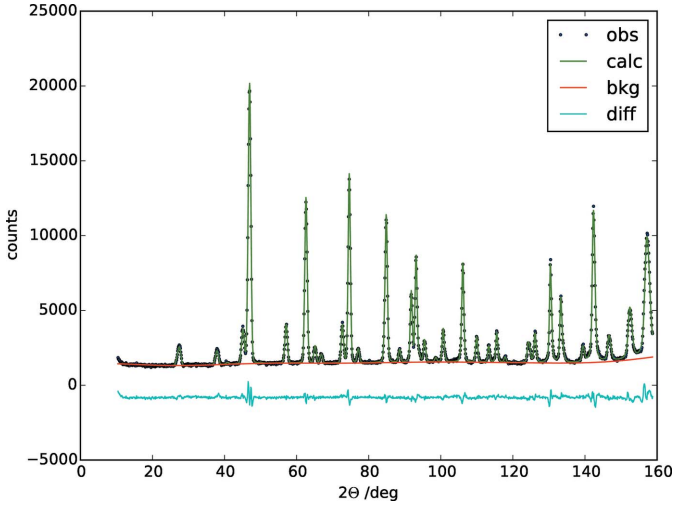


Figure 2
Sapphire powder (1.22 g Al_2O_3, NIST standard) measured for 1 h on PEARL at 1.67 Å. The fit result is shown as a green line, with the background component in red and the difference between observed and calculated in cyan.

6. Comparison with the *McStas* Monte Carlo simulations

The peak width of a diffraction pattern is related to the beam characteristics, dimensions and divergence of the beam, and

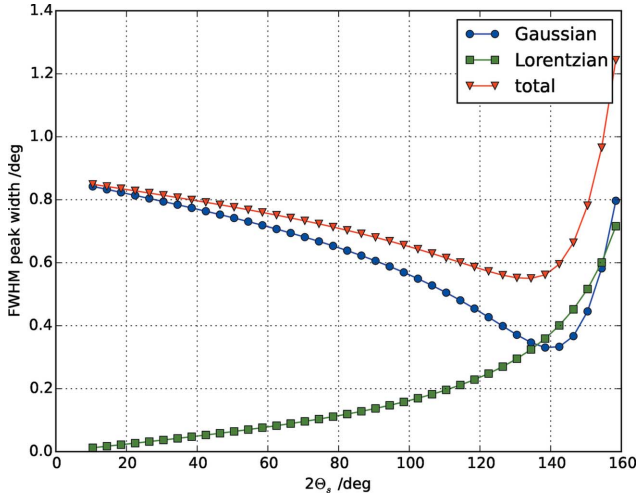


Figure 3
The instrumental resolution as peak width *versus* scattering angle $2\Theta_s$.

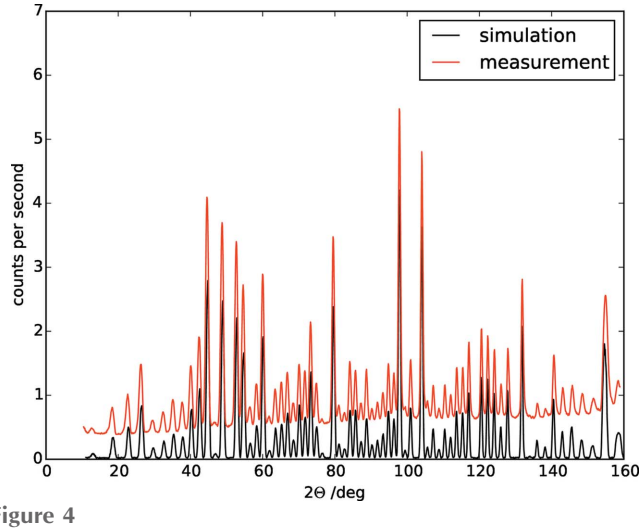


Figure 4
A comparison of experimental data (1 h) with the *McStas*-simulated pattern from the design model. The powder was $Na_2Ca_3Al_2F_{14}$.

both Cussen (2005) and Bobrovskii (2009) extended the Caglioti description of instrumental resolution (Caglioti *et al.*, 1958) by relating the beam dimensions to the divergence. As this relation is implicit in Monte Carlo ray tracing, the instrument design of PEARL was verified with *McStas*, yielding a diffraction pattern of a virtual $Na_2Ca_3Al_2F_{14}$ powder sample. As can be seen in Fig. 4, the agreement in peak width between the simulation and measurement is rather good.

7. Conclusion

PEARL demonstrates that a novel design can result in a very competitive performance at low cost even at a relatively low brilliance neutron source. This opens up new applications not only at the next-generation high-intensity sources but also at small and medium neutron sources worldwide. At Delft University of Technology, PEARL will serve the in-house science program on, for instance, magneto-caloric, battery electrode or electrolyte materials and nuclear materials. Furthermore, its competitiveness should serve to attract external scientists.

Acknowledgements

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References

Bobrovskii, V. I. (2009). *J. Synch. Investig.* **3**, 5–14.
Caglioti, G., Paoletti, A. & Ricci, F. P. (1958). *Nucl. Instrum.* **3**, 223–228.
Cussen, L. D. (2005). *Nucl. Instrum. Methods Phys. Res. Sect. A*, **554**, 406–414.
Cussen, L. D. (2007). *Nucl. Instrum. Methods Phys. Res. Sect. A*, **583**, 394–406.
Cussen, L. D. (2016). *Nucl. Instrum. Methods Phys. Res. Sect. A*, **821**, 122–135.
Hewat, A. W. (1975). *Nucl. Instrum. Methods*, **127**, 361–370.
Howard, C. J. (1982). *J. Appl. Cryst.* **15**, 615–620.
Lebech, B. (2001). *J. Phys. Soc. Jpn. Suppl. A*, **70**, 483–485.
Rietveld, H. M. (1969). *J. Appl. Cryst.* **2**, 65–71.
Sykora, G. J., Schooneveld, E. M., Rhodes, N. J. & van Eijck, L. (2012). *2012 IEEE Nuclear Science Symposium and Medical Imaging Conference Record (NSS/MIC)*, pp. 1567–1571. Piscataway: IEEE.
Toby, B. H. (2001). *J. Appl. Cryst.* **34**, 210–213.
Willendrup, P., Farhi, E. & Lefmann, K. (2004). *Phys. B Condens. Matter*, **350**, E735–E737.