DRACULA – An ILL Diffractometer for Rapid Acquisition

Dracula will be the world's fastest Diffractometer for Rapid Acquisition over Ultra Large Angles, competing favourably with the machines being built on the new American and Japanese pulsed sources. Dracula will be a third generation ILL machine, following the success of D1A/D1B and D2B/D20. The project was rated as top priority by the ILL Instrument Committee in 2003, and approved by the ILL Science Council in 2004. The idea is to take advantage of the very high time-averaged flux that a continuous source can deliver to the sample with modern neutron optics, and then to match the large detectors that have been used so successfully on pulsed sources. Instruments like Dracula will be essential if Europe is to maintain it's lead in neutron diffraction until ESS can finally be built.

Strategy for Dracula

A third generation neutron powder diffractometer will be one of the baseline instruments on the US-SNS, with installation starting in 2006 and operation by September 2007. ILL plans to match this objective.

"…the SNS should immediately begin work on the conceptual design for… a third generation powder diffractometer with a resolution Dd/d of ~1x10^{-3} at 90^{o}." SNS-IOC recommendations, Nov 1998.

"SNS without a world-class powder diffractometer on day one is unthinkable." SNS-EFAC, May 2001.

*"**1.1 Recommendation:** A high level of priority should be assigned to bringing the powder diffractometer (POW-GEN3) into operation as early as possible."* SNS-EFAC recommendations, Dec 2003.

In making their case for a new high flux neutron source, the American Shelter Island workshop compared the relative advantages of pulsed and continuous sources. It was found that in many cases the two sources were competitive, and Shelter Island finally recommended the reactor source as being more flexible. But because of difficulties in building a high flux reactor significantly better than ILL, it was finally decided to build the pulsed source SNS instead. The comparisons made at Shelter Island remain however, valid, and serve to show how instruments on the existing high flux ILL reactor might be further optimised.

Jorgensen et al.[1] concluded that because neutrons are scattered isotropically, the relative merit of a powder diffractometer with a given resolution can be estimated from the product of just three parameters – the time averaged flux on the sample, the solid angle of the detector and of course the volume of the sample, which may be limited by resolution and other requirements. This product rule appears to be supported by the current world's best high flux diffractometers, GEM on the ISIS pulsed source and D20 on the ILL reactor (below), which are roughly of equal merit, even though the time-averaged flux on D20 from the ILL reactor is more than an order of magnitude higher than on GEM.

	D20	GEM	DRACULA	SNS
Time averaged flux	5x10^{7}	~2x10^{6}	~10^{8}	~2.5x10^{7}
Detector solid angle	0.27	4.0	1.5	3.0
Sample volume	1	1	1	1
Merit product	18	8	150	75

This table shows that GEM competes only because of its exceptionally large detector, much bigger than anything that could be built until recently for reactor-based diffractometers. The pulsed source machine GEM is unique in that the detector is not restricted to high resolution backscattering, but covers all scattering angles, with however correspondingly lower resolution.

1. Jorgensen, J.D., Cox, D.E., Hewat, A.W., Yelon, W.B. (1985) Nuc.Inst.Meth. **B12**, 525-561.

Clearly if we could construct a diffractometer with similar design objectives, the high flux reactor machine would easily outstrip the medium flux pulsed source machine and compete with the high flux SNS pulsed source diffractometer.

Indeed, modern focussing monochromators using very large beam tubes on a high flux reactor can deliver more flux to the sample than even the SNS pulsed source; fluxes greater than 10^8 $n.cm^{-2}.sec^{-1}$ have already been achieved on ILL 3-axis machines, and they approach that number for D20, which is almost 20 metres from the reactor! The clear conclusion is that a modern diffractometer (D20 was conceived 20 years ago) could be superior to anything on the American SNS until ESS could finally be built.

The situation is not very different for single crystal machines with large detectors, especially for reactor-based machines such as quasi-Laue image plate detectors, where a white beam of neutrons is used as on a pulsed source, with the important difference that the beam is not pulsed! White beam diffractometers are the top priority of SNS-EFAC, but they will not match ILL's time-averaged flux on the sample.

The Dracula Proposal

We propose to capitalise on recent ILL advances in the construction of large gas detectors and focussing monochromators to build a very high flux third generation diffractometer that could be used for both powders and single crystals. This Dracula machine would be the world's most powerful neutron diffractometer, especially for the study of chemical kinetics, or small samples in special environments such as pressure cells. It would help maintain Europe's lead in neutron diffraction. The objectives are:

- Construct a more compact machine than D20 to maximise intensity on the sample.
- Use a big doubly focussing composite monochromator to deliver a large wavelength band.
- Provide sufficient space for extreme sample environments such as pressure cells to 100 kbar, magnets to 15 Tesla, very high temperature furnaces and neutron polarisers.
- Ensure $\Delta d/d \sim 10^{-3}$ resolution with low background by working at take-off angles around 90^o with radial collimation to eliminate scattering from special sample environments such as pressure cells.
- Use several different wavelengths in this focussing geometry to cover a wide range of d-spacings.
- Adapt a new D19-type 2D position-sensitive detector to cover an exceptionally large solid angle, integrating the diffraction cones as on super-D2B.

The Proposed H9 Beam Tube for Dracula

Dracula calls for a 200mm diameter thermal beam tube position as close as possible to the reactor face in order to maximise divergence and flux from the large focussing monochromators. The H9 (Lohengrin) beam tube would be the best possible choice; this would not interfere with Lohengrin, and it would even be possible for Dracula to co-exist with Tomography. There are in fact two options:

A) Dracula would simply replace the tomography station. The weight of the Dracula shielding would be very similar to the weight of the present tomography shielding, with a similar distribution.

B) The tomography station would be moved ~4m from its present position, giving slightly less flux and slightly better resolution. The drawing shows a tomography casemate 1m longer than the present casemate, to allow more room for larger components. (This would however restrict the passage behind tomography to ~2m). Calculations are being undertaken to determine if the extra weight of ~150T could be supported, but according to reactor personnel it would be relatively easy to add an additional support pillar in level D if this was necessary.

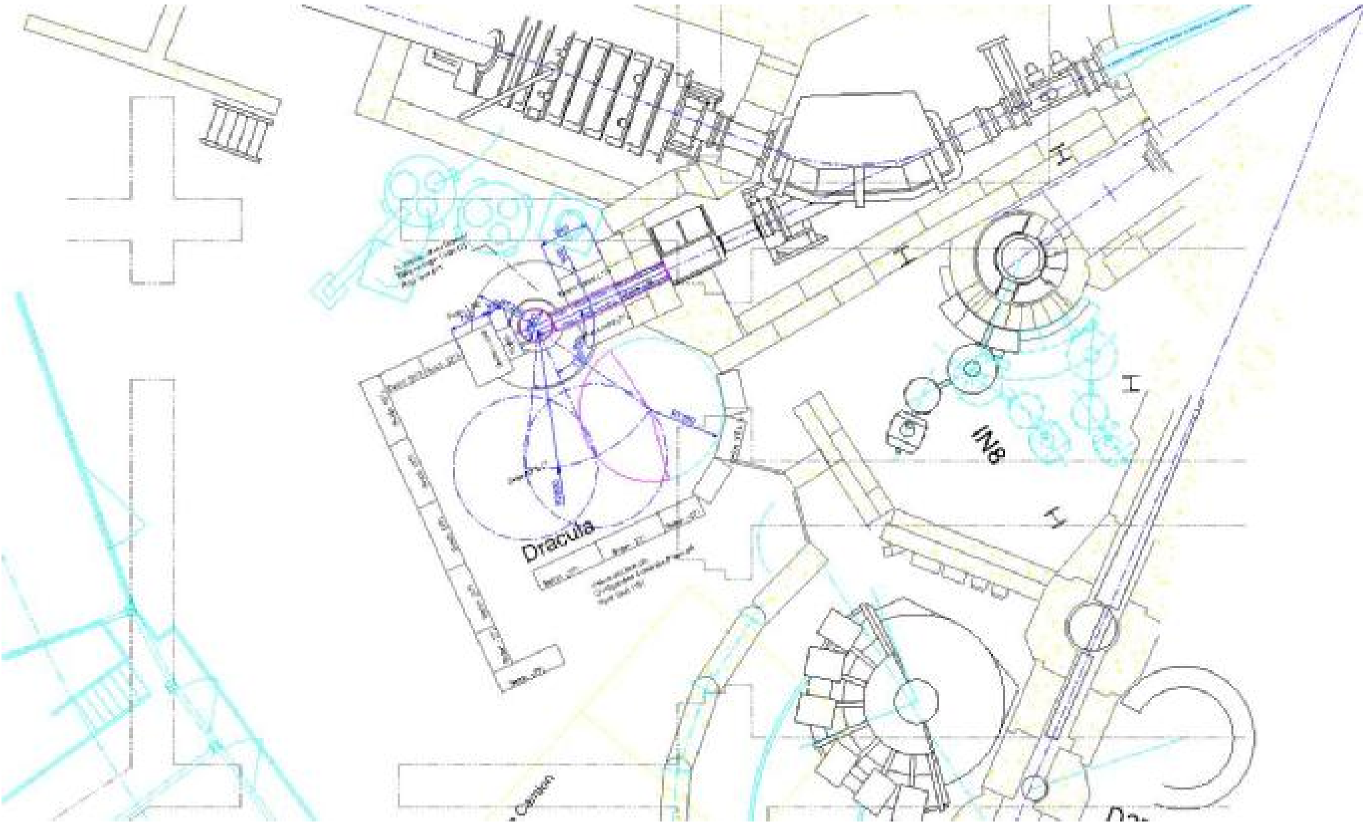


Fig. 1a Option A: Dracula would replace the tomography station on the 200mm diameter high flux H9 Lohengrin beam tube.

Fig. 1b Option B: Dracula would displace the tomography station on the 200mm diameter high flux H9 Lohengrin beam tube.

The H9 beam tube is perhaps the most intense thermal neutron beam anywhere, and if we are to compete with the American pulsed source this position must be used to maximum advantage.

The Dracula Monochromator

As on D2B and D20, very large focussing monochromators would be used, with even larger mosaic spread. As well as focussing in real space, focussing in reciprocal space would mean that a wide band of wavelengths $\Delta\lambda/\lambda$ could be used (below left). When this beam is scattered by the sample back into the direction incident on the monochromator, all of the wavelengths enter the detector with the same incident angle; this means that the width of the Bragg peaks depend only on the size of the sample and detector elements and the divergence of the beam tube. Since very small samples (diameter d) would normally be used, and since a choice of soller collimators would fix the horizontal divergence from the beam tube, high resolution $\Delta d/d \sim 10^{-3}$ would be obtained even with a wavelength spread of ~1%. For angles near 90^{o} with a radial collimator, scattering would be limited to a small volume of diameter D (below right).

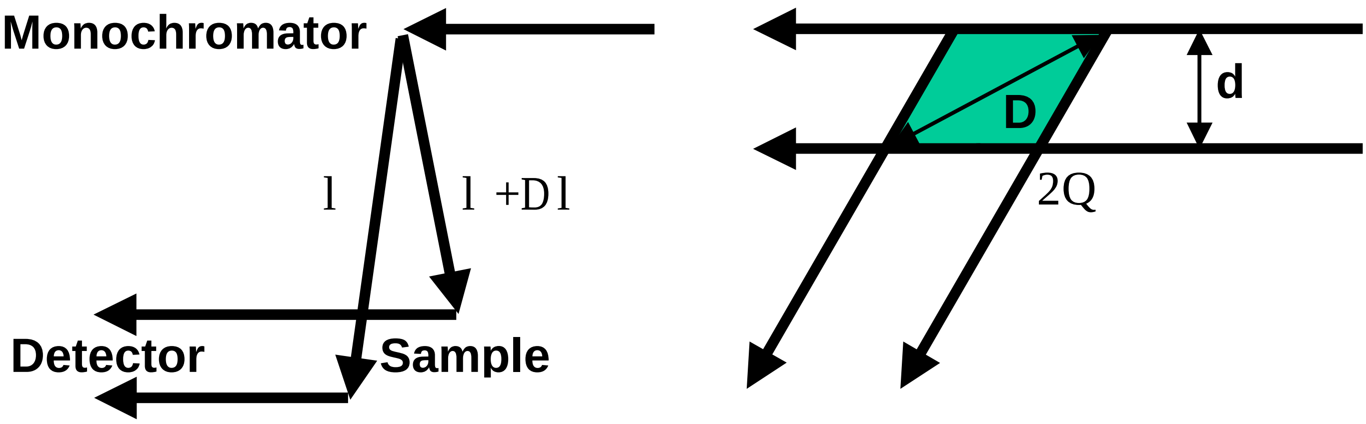


Fig. 2(a) Mosaic monochromator focussing a wide band of wavelengths **2(b)** Small scattering volume with a radial collimator.

A choice of fixed monochromator take-off angles would be available, probably 44^{o}, 60^{o}, 90^{o} and 120^{o}. The standard take-off would be 90^{o}, but a low angle take-off to provide very high flux from a graphite monochromator would also be available, together with a high take-off of 120^{o} for very high resolution. The main monochromators would be Germanium [hhl] giving, for a take-off angle of 90^{o}, the following wavelengths and range of d-spacings for scattering within the range $2\Theta=60^{o}-120^{o}$:

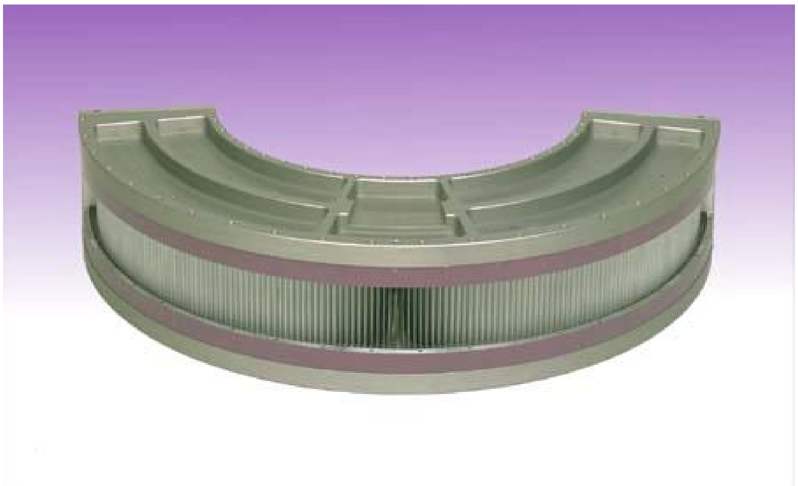
[115] -> 1.54Å		**d= 0.889Å - 1.54Å**
[113] -> 2.44Å	**(graphite filter)**	**d= 1.39Å - 2.44Å**
[111] -> 4.61Å	**(beryllium filter)**	**d= 2.66Å - 4.61Å**

Fig. 3 The large range of d-spacings available using just three wavelengths from a focussing Ge[hhl] monochromator.

Note that even with this limited scattering range of 60^{o}-120^{o}, which is attractive for reducing scattering from the sample environment with a radial collimator, a large range of d-spacings could be covered with just three different wavelengths, which would be obtained by simply rotating the Ge[hhl] monochromator about it's vertical axis as on D1A/D2B. The most useful wavelength of 1.54Å would not require any filter, while a graphite filter would be used for 2.44Å and a beryllium filter for 4.61Å neutrons.

The Dracula Sample Environment

Because of it's very high flux, Dracula will be particularly powerful for measuring very small samples, as found in high pressure cells. A new 100 kbar Paris-Edinburgh cell is currently being constructed at ILL to fit within a special cryostat (N. Kernavanois). This new pressure cell will be particularly suited to Dracula, with its high flux on small samples and radial collimator to eliminate background scattering from the sample environment. The picture shows a radial collimator used on the Swiss high flux powder diffractometer, together with the anvils of the new ILL pressure cell.



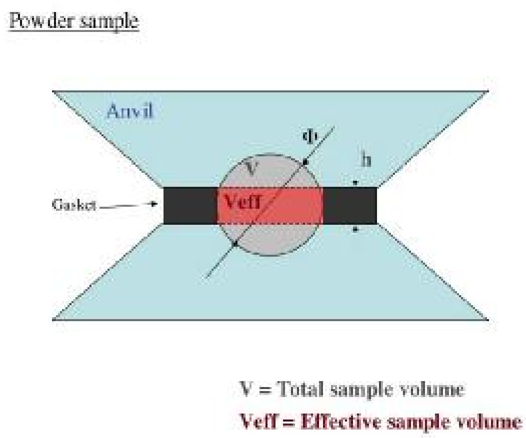


Fig. 4 Radial collimator of the type to be used for Dracula **2(b)** High pressure Paris-Edinburgh cell being constructed for ILL.

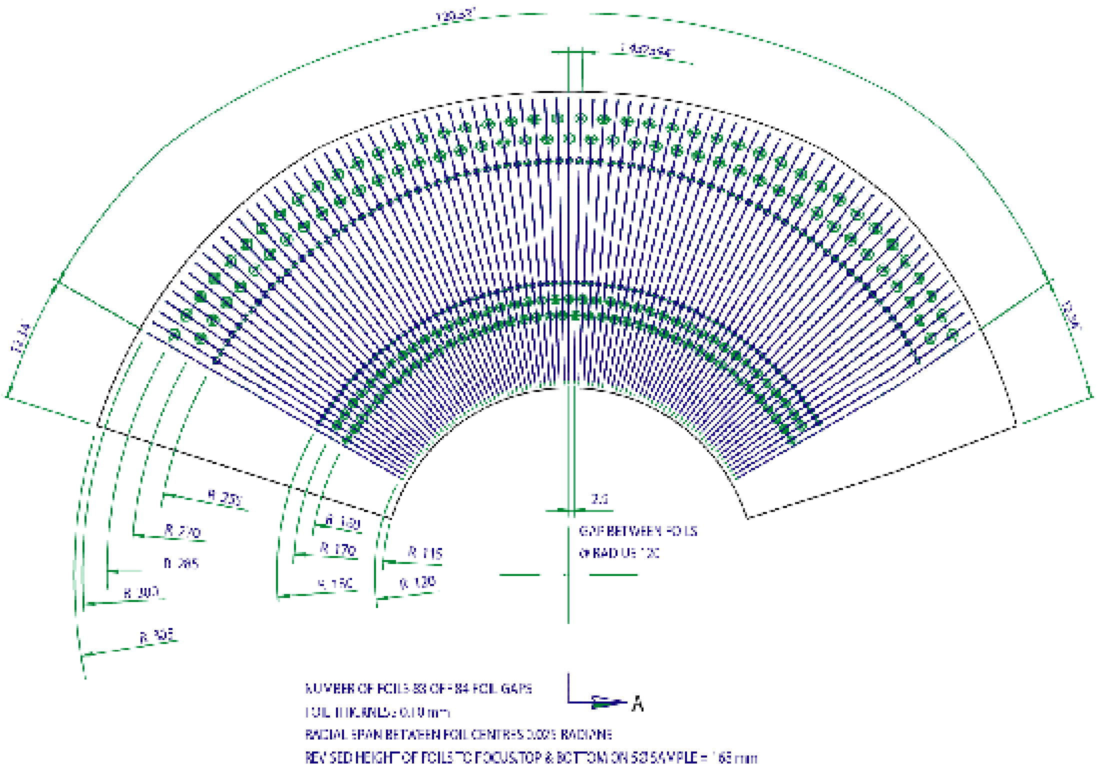


Fig.5. The Dracula radial collimator will cover a very large solid angle (160^o x 30^o vertical) yet restrict scattering to a very small volume of diameter ~5mm around the sample. (EuroCollimators Cheltenham, 2003).

One of the problems with the present ILL powder machines D2B and D20 is that they are covered by the reactor transfer channel, which limits the height of the sample environment. Dracula will not be limited in this way, and will allow even large cryostats to be used, such as the new ILL 15 Tesla magnet, the new 7 Tesla diffraction group cryomagnet, and cryostats equipped with dilution refrigerator inserts.

Very high temperatures can already be obtained on D2B with mirror furnaces and microwave furnaces, which will also be available on Dracula.

The Dracula 2D Detector

This is the key element, and will provide Dracula with a detector x4 times as large as that of D20, which is already the largest, fastest detector on a fixed-wavelength neutron powder diffractometer. Together with the extra flux on the sample, Dracula will then be an order of magnitude faster than D20 at the cost of slightly less resolution than available in D20's high resolution mode. Indeed, the solid angle of the Dracula detector will reach 50% of that of the fastest TOF machines, which unlike fixed-wavelength focussing diffractometers, can also measure at "negative" scattering angles without loss of resolution.

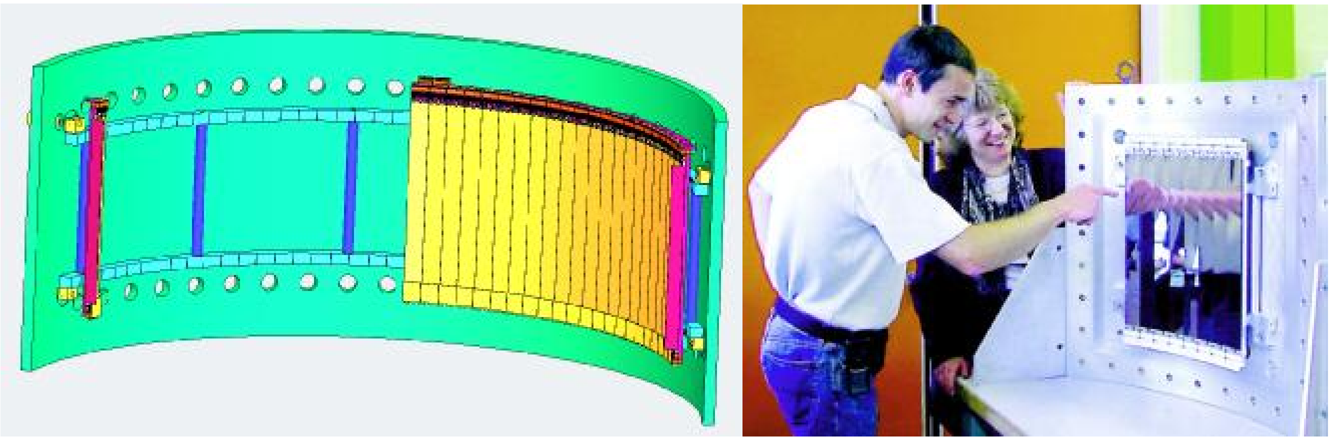


Fig. 6. The new 30ºx120º 2D-PSD for D19. Large detectors match the solid angle of the best pulsed-neutron diffractometers, while benefiting from the very high flux available on the sample at a reactor source. (Guerard et al. 2002).

The Dracula detector will be almost identical to the new D19 detector, except that extra modules will extend coverage to 160º like D20 instead of 120º as on D19. The vertical acceptance angle will remain at 30º, as on the new D19. The principle of using a large 2D detector has already been proved on super-D2B, where it has been shown that the diffraction cones can be integrated around their curved sections without substantial loss of resolution.

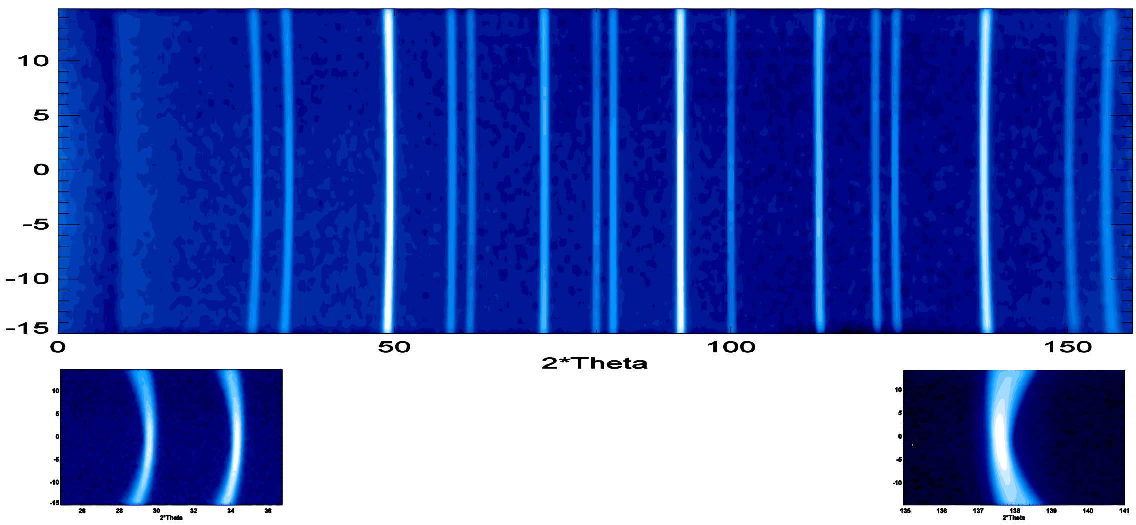


Fig. 7. High resolution powder data from a large 2D detector, showing the curvature of the diffraction cones (Suard et al 2003).

With a horizontal definition of at least 2.5mm, the Dracula detector will be resolution matched to samples as small as 2.5 mm diameter. For even better definition of the powder profile, it will be possible to fine-step the Dracular detector on it's tanzboden, as on D2B.

Expected Performance of Dracula

As we have seen, Dracula will be an order of magnitude faster than D20 for many problems, while providing resolution only a little lower than D1A, or D20 in it's high resolution mode. As such it will be the fastest neutron powder diffractometer for small samples, even compared to the new American and Japanese pulsed source powder machines. As an example of what can be expected, consider the high resolution powder pattern of a relatively complex mineral that can already be obtained from D20 (below).

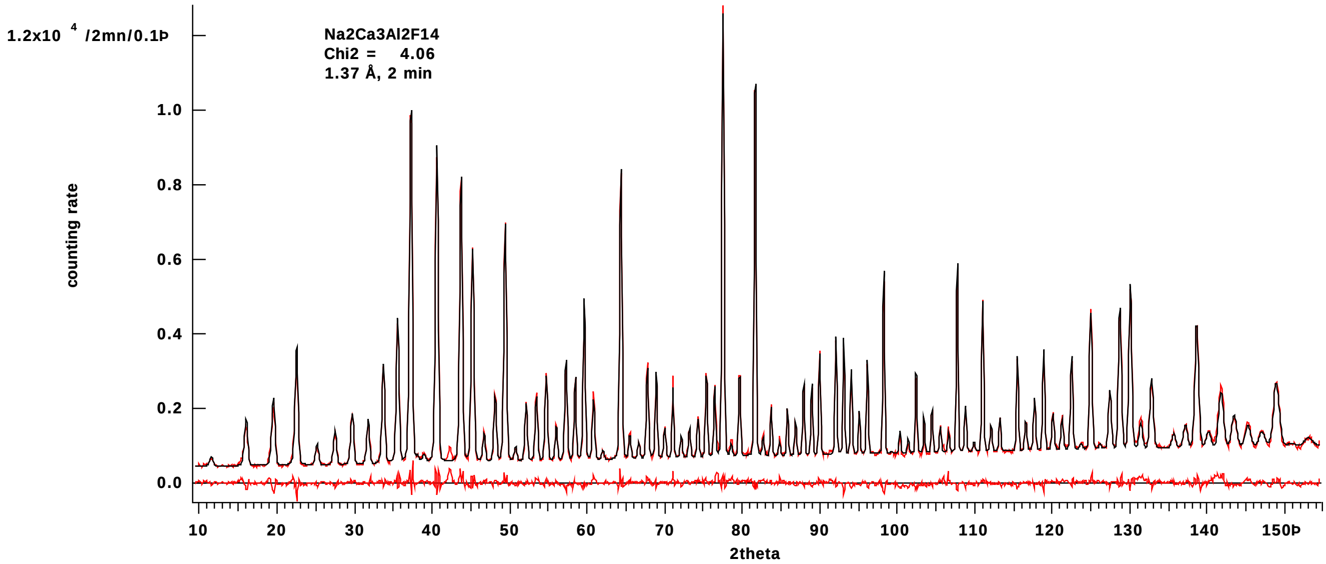


Fig. 8. Complex pattern obtained in only 2 minutes on D20 in high-resolution mode. (Hansen et al. 2003).

The complete neutron pattern, which was obtained in just 2 minutes on D20 can be refined to give a high precision structure, as indicated by the red obs-calc difference pattern in the profile plot. On Dracula we might expect to obtain similar results in a matter of seconds, or a few 10's of milliseconds for large samples to be used for chemical kinetics.

For very small samples, such as those in high-pressure cells, we might expect to collect good quality high-resolution data from samples of a few 10's of milligrams. The sample volume in the new 100 kbar ILL cell, at ~4 mm^3, is large enough to allow counting times of ~1 hour; this would mean that data could be collected as a function of temperature even at very high pressure!

Cost and timeline for Dracula

Costs are indications only at present. The cost of the detector and it's electronics can be estimated quite well from experience with the very similar D19 detector. The cost of the monochromators is mainly ILL manpower (~1 man year for a large Ge[hhl] monochromator), and only the capital cost of the germanium and graphite crystals is estimated here. The radial collimator is available commercially, and again the cost can be estimated quite well. The biggest uncertainty is actually the cost of the casemate and beam-tube protection. Only a rough figure is given, even though this is well-known engineering; a preliminary design study, including costs, should be available in the summer of 2004.

Estimated capital costs in 1000's of Euro (approximate only):

1.	Beam tube protection and casemate	1,000
2.	D19-type 2D PSD detector & electronics	1,000
3.	Monochromator crystals (germanium and graphite)	50
4.	Mechanics for monochromator support	50
5.	Tanzboden floor	50
6.	Mechanics for detector support	100
7.	Radial oscillating collimator (optional)	100
8.	Provisions for extras	50
	Total	**2,400**

Estimated construction timeline:

Estimates are based on experience with the new D2B and D19 detectors, though of course Dracula is a bigger project, since an extension to the H9 beam tube protection, monochromator casemate and secondary diffractometer mechanics must also be constructed. However, these mechanical elements are all relatively straightforward. Apart from money and manpower constraints, the limiting factor is likely to be the large germanium monochromator, so Dracula might first operate with only a graphite monochromator and filter, which can be provided commercially much more quickly.

The nominal project starting date is 2006, but of course detailed planning could start as soon as manpower or design funding became available. The earliest date for start of construction would be with the long reactor shutdown in mid-2005, when construction and testing of the D19 detector will have been finished. The actual start of construction of the detector will naturally depend on manpower. Construction and testing of the Dracula detector and electronics should take ~18 months, based on experience with D19. Much less development time will be needed because the detector will be very similar.

Mid-2005 is also the earliest date when preparation of the H9 site might begin. During the 2005 shutdown it would only be possible to clear the site and prepare the interface between the present H9 beam protection and the Dracula protection, which could then be constructed and installed during reactor operation. Again, construction of the Dracula protection could extend over a period of 12-18 months.

Monochromator and detector tanzboden mechanics would be supplied commercially within 12 months, and the critical factor here would be funding rather than manpower.

With sufficient funding, as was obtained for example with recent EPSRC projects D2B and D19, a working Dracular machine using commercial components apart from the detector, but including a graphite monochromator and filter, might be completed in as little as 2 years. The project would be completed 1 year later with the delivery of the germanium monochromator, but during this time the machine could already contribute to the user programme, and keep ILL one step ahead of the competition.