In Defence of ICARE - A High Speed Powder Diffractometer.

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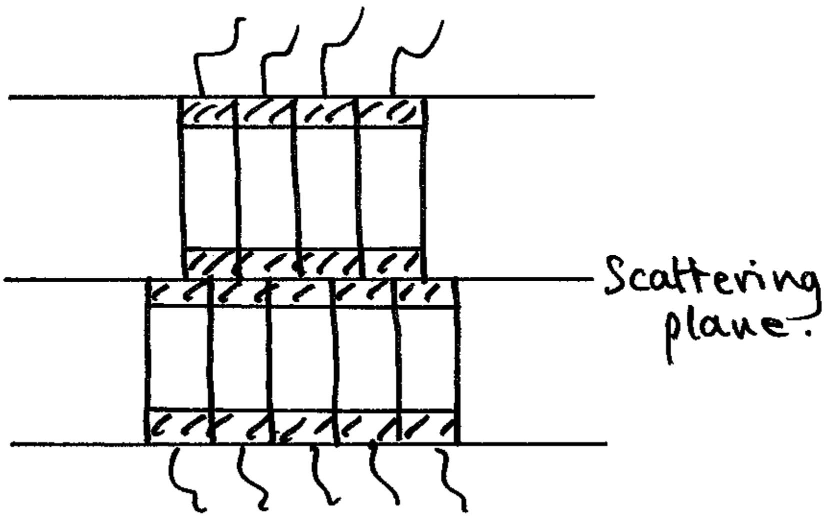
The basic question in constructing a powder diffractometer is whether to put all the wires in the same envelope (D1B type PSD), or in seperate envelopes (ICARE - Independent Counter Array with Replaceable Elements). The answer, for practical purposes, seems obvious! If the probability of each element working is 0.999, then the probability of 400 working (D1B) is something like $(0.999)^{400}$ = 0.67, but only 0.45 for 800 elements and 0.20 for 1600. With ICARE, one simply replaces the faulty element, but with a PSD every minor fault is a disaster.

There is no fundamental reason why either the PSD or ICARE should give higher resolution - or rather higher definition of the diffraction pattern. In this paper, we will describe three alternative methods of increasing the definition of ICARE - that is, the number of points defining a peak.

1) ICARE Divided. (I CARED)

We simply divide ICARE at the equatorial plane and displace the top row of counters by half an element relative to the bottom row (fig. 1).



This doubles the definition of the machine to 0.1^{o} without changing the resolution. ICARED permits a larger vertical aperture and higher count rates than possible with ICARE or a PSD, even though the equatorial plane itself is dead space. Commercially available 6 x 12 mm counters (5NH10/1P) could be used.

2) Very High Resolution ICARE. (VICARE)

It is perfectly feasible to manufacture individual counters of diameter 5mm, squashed to 3 mm wide x 6 mm deep, and arrange them in a staggered array on a circle of diameter 191 cm to provide a definition of 0.1^o 2θ. Competitive efficiencies (eg 80% for 2.5Å neutrons could be achieved with gas pressures of up to 20 atmospheres. Such detectors do not at present exist, but then neither does an 80^o PSD with 0.1^o definition. The developement of a 3 x 6 mm counter from the commercially available 6 x 12 mm prototype is not difficult, and this developement would be done by the manufacturer, and not use scarce ILL resources. This solution was not chosen for the high resolution diffractometer D2B (Hewat, 1975) because very small samples reduce the intensity below what can be obtained by collimating the scattering from large samples.

Clearly, with ICARE, one could start with a small number of counters and simply add more as finance becomes available. The mechanical support for 1600 detectors covering the complete 160^o would be constructed from the beginning.

3) Scanning ICARE. (ISCARE)

The definition can be increased as much as required if scanning is permitted. Only a very small scan angle ($<1^o$) is needed, and since it would be quite automatic, the typical user, for whom the multidetector is in any case a black box, would hardly even be aware that it was happening.

Scanning would only be really necessary if the diffraction pattern contained extremely sharp peaks and profile fitting could not be used. We shall see below that a stationary ICARE can be used if preferred for the great majority of experiments; integrated peak intensities are very insensitive to errors in counter bank positioning, even if the peaks are quite sharp, and even if there are dead spaces between counters.

There appear to be two objections to scanning:

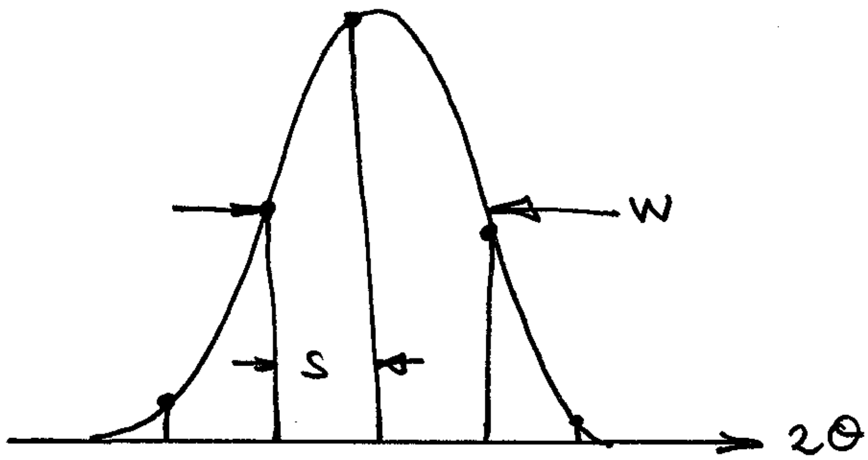
1) Scanning is impossible for extremely short measuring times (<1 sec). These experiments in any case require large samples, and peak widths are not extremely sharp - a stationary ICARE is sufficient.

2) Scanning is impossible for subtractions of long measurements to look for very small differences, since once moved, the counter bank cannot be repositioned in exactly the same place for the next run. This is indeed a problem with low definition, where only a few points fall on a peak and a slight error in the counter bank setting produces big changes in the counts obtained on individual detector elements (but not in the integrated line intensity as we shall see). It is not a problem with high definition or broad peaks, when many counters cover each peak; SCANNING PRODUCES HIGH DEFINITION PATTERNS !

Resolution and Definition of a PSD and ICARE.

Definition, which depends on the step size s (fig. 2) is preferred to resolution, measured by the peak width w, since the latter is determined mainly by the incident beam divergence α_1, and the sample diameter d.

(fig. 2)



Resolution, for focussing geometry, is given by

$$w = \sqrt{\alpha_1^2 + \alpha_3^2} \qquad (1)$$

where $$\alpha_3 = \frac{d+e}{2D} = \frac{e}{D}\left(\frac{1 + d/e}{2}\right) \qquad (2)$$

and $$\delta = \frac{e}{D} \qquad (3)$$

where e is the element spacing in the detector distant D from the sample, diameter d . The intensity is proportional to $\alpha_1 \alpha_3$ and the sample volume $\pi d^2 h$

$$I \approx \alpha_1 \alpha_3 d^2 \qquad (4)$$

Clearly, we should choose $\alpha_1 = \alpha_3$ (according to (1)) and d=e (according to (2)). Then

$$\alpha_1 = \frac{e}{D} = s \qquad (5)$$

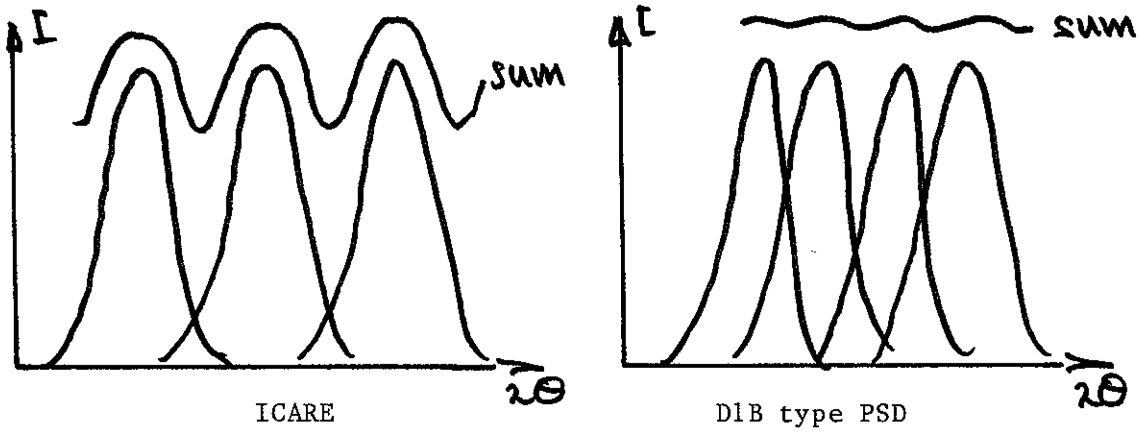
$$w = \frac{e}{D}\sqrt{2} = s\sqrt{2} \qquad (1b)$$

For example, with a counter spacing of 0.2° (D1B) the best possible resolution is 0.28° 2θ. The fact that measured lines are wider than this means that α_1 and d rather than s determine the real resolution.

Errors due to counter position (high resolution case).

Equation 1b means that for the optimum PSD or ICARE, the sharpest peaks are only covered by a small number of elements i.e. that the definition is low. As well, with ICARE, there will be dead spaces between the counters, or at least spaces of lower efficiency (fig. 3). Does not the recorded peak intensity the depend strongly on the exact position of ICARE i.e. on whether the peak is centred on a counter, or between counters ? These considerations lead PSD designers to strive for very uniform detectors.

(fig. 3) RESPONSE OF THE DETECTOR TO A SLIT SOURCE.



It is easy to examine the problem by summing the counts of the group of elements contributing to a peak, and examining the difference obtained if the whole counter bank is displaced by an arbitrary amount. Clearly, the maximum percentage error Δ% in integrated line intensity, will depend on the ratio s/w of sampling interval s (element spacing) to peak width w (fig. 2): if s<<w, there will be no error, and this is the reason why D20 users want high definition. To quantify this argument, we have calculated the following table using equations (1), (2) and (3) and a computer to sum the elements for arbitrary displacements of the counter bank.

d	s/w	$\pm\Delta$%
0	$2/\sqrt{5}$ =0.8944	2.28
e	$1/\sqrt{2}$ =0.7071	0.16
3e/2	$4/\sqrt{41}$ =0.6247	0.02
2e	$2/\sqrt{13}$ =0.5547	0.00

Then even if the peak is very coarsley sampled its integrated intensity is quite insensitive to errors in counter bank positioning. Even for the smallest sample diameter (d=0), the error from positioning error is negligible.

This table was computed for the gaussian resolution (w) of either ICARE or a PSD. In practice, this gaussian will be convoluted with the sample line shape, which is not necessarily a gaussian, or even a single peak: this makes no difference, except that in so far as the measured peak is wider, the error in integrated intensity will be smaller.

Errors due to non-uniform element spacing.

This source of error is much more important. Neither the wires in a PSD nor the individual counters in ICARE can be spaced with sufficient uniformity to achieve the intensity precision demanded by some users of D20. The important parameter is Δs/w: if Δs/w is 1%, then the error in integrated line intensity is also about 1% (Lehmann, 1975). Clearly the problem becomes more important with high resolution (small w). On D1A, it has been solved by calibrating the spacing between counters, as well as the relative counter efficiency, and automatically interpolating to reduce this source of error. The same technique would be used on ICARE, and could also be used on a PSD. This calibration can best be done if the counter bank can be scanned.

In practice, the most precise intensity measurements would be obtained by slowly and continuously scanning the counter bank and recording the counts at regular time intervals (Lehmann, 1975). The angular range covered need be very small; it is hard to see any disadvantage in using such a scanning technique compared with the considerable advantages of increased definition and precision in the diffraction pattern.

The conclusion is that very high resolution with a stationary PSD is a disadantage if very high precision (< 1%) is required in line intensities. A 1% intensity error is an acceptable price to pay for high resolution for structural crystallography, but do D20 users really want high resolution at this price?

Other Objections to ICARE.

1) Non-uniform multidetector response.

It is irrelevent that the response to a sharp line source is periodic and not uniform (fig. 3). Real samples and real diffractometers do not produce line sources; the folding (convolution) of the sample with the detector produces a near perfect Gaussian resolution function.

2) Need for scanning.

It is not necessary to scan ICARE to produce this result. A stationary ICARE gives a periodic sampling of the diffraction pattern, which is all that any PSD gives.

3) What about non-Gaussian lines?

This result also applies to non-Gaussian diffraction line shapes. It is only the resolution function which is necessarily Gaussian; the observed line shape is the sample line shape folded with the gaussian resolution function.

4) Holes between the counters.

The integrated line intensity for ICARE is quite insensitive to errors in the counter bank position even if there are holes (dead space) between the counters, provided that the sampling s (counter spacing) is small compared to the width of the resolution function w i.e. that $s/w < 1/\sqrt{2}$. This is always the case, even for very small samples. Since this result is true for the resolution function itself, it is also true for any measured line (convolution of the resolution function with the sample line shape) whether or not the sample line is Gaussian.

5) Efficiency of ICARE compared to a PSD.

The global efficiency of ICARE is higher than that of a conventional PSD; losses between counters can be reduced to a small fraction by using staggered squashed counters, higher gas pressures can be contained, and larger apertures (up to 300 mm high) are possible. For even the standard squashed counter JNH10/1P (10bars of He3 for a cross-section of 6 mm wide x 12 mm maximum deep) the peak efficiency according to Convert(1975, p14) is competitive.

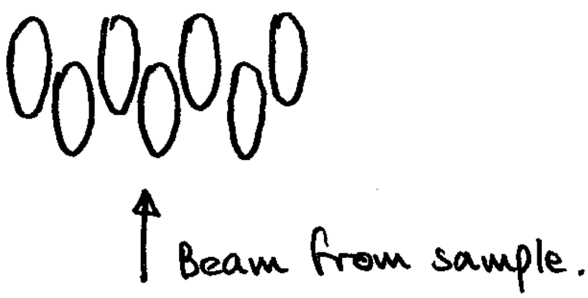
λ	peak efficiency %
2.5	90
1.5	78
1.0	72

To obtain global efficiency, one must multiply by the aperture height and average gas thickness.

6) Efficiency losses at counter junctions.

These losses can be greatly reduced with a staggered configuration of squashed counters.

(fig. 4)



The overlap between counters (fig. 3a) is not important provided all can be made identical by calibration. Element overlap is also used in a D1B type PSD in an effort to obtain uniform efficiency (fig. 3b).

7) Scattering from counter walls.

This problem is even more important for the conventional PSD, where the counter wall must be an order of magnitude thicker to contain the gas pressure over a much larger area.

8) Differences in efficiency between counters.

These differences are not fundamentally larger for individual counters than for individual wires in a single envelope. In practice it is necessary to calibrate the elements in any case - the stability of the elements is more important, and independent counters have an advantage here because elements falling outside the norms can be replaced.

References.

Convert, P. (1975) These, Uni. Grenoble.

Hewat, A.W. (1975) Nucl. Inst. Meth. 127, 361-370.

Lehmann, M.S. (1975) J. Appl. Cryst. 8, 619-622.