A New Small-Angle X-ray Scattering Facility Utilizing a Rotating Anode, Pin-Hole Collimation and a Position-Sensitive Proportional Counter*

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A new small-angle X-ray scattering facility which utilizes a 6 kW rotating anode, pin-hole collimation, and a position-sensitive proportional counter has been developed. As presently constructed, the minimum scattering vector \( \kappa = 4\pi \sin \theta / \lambda \) which can be reached with Cu \( K\alpha \) radiation is \( 5 \times 10^{-3} \) Å \(^{-1} \). Under these conditions the flux incident on the specimen has been found to be \( 6 \times 10^6 \) photons s \(^{-1} \). The system has several advantages compared with traditional long-slit geometries; namely, (i) it can quantitatively measure anisotropic scattering distributions, (ii) it avoids large mathematical corrections of the data for slit-smearing effects, and (iii) it minimizes double Bragg scattering in crystalline materials and multiple diffuse scattering in amorphous or liquid materials. To illustrate the performance of this instrument, the scattering curves obtained from four widely different samples are shown. These are: polyethylene, a neutron-irradiated aluminum single crystal containing voids, a dilute suspension of Ludox spheres, and duck tendon collagen. Quantitative comparisons of the performance with a Kratky camera and with the neutron small-angle scattering facility in Jülich are given.

Introduction

Small-angle X-ray scattering (SAXS) experiments are usually performed in a long-slit geometry in which X-rays are appropriately collimated in one direction but are poorly collimated in the other direction. Historically these geometries have developed as a compromise between perfect (point) collimation and insufficient scattering power. Such instruments have limitations and disadvantages, the principal of which are (i) significant collimation corrections must be applied to the observed data to recover the scattering function, and (ii) from a practical point of view, only isotropic scatterers can be investigated. With the advent of position-sensitive X-ray proportional counters it becomes reasonable to collimate the incident beam in both directions and to recover the loss in intensity by simultaneously recording data at many scattering angles. Although two-dimensional detectors have been developed (Borkowski & Kopp, 1970, 1972, 1974), to date only one-dimensional detectors have been used for SAXS (Dupont, Gabriel, Chabre, Gulik-Krzywicki & Schecter, 1972; Barrington-Leigh, Holmes & Rosenbaum, 1973; Barrington-Leigh & Rosenbaum, 1974).

In this laboratory, small-angle scattering is being used to study the nature of voids in neutron-irradiated metals and alloys (Epperson, Hendricks & Farrell, 1974; Hendricks, Schelten & Schmatz, 1974; Hendricks, Schelten & Lippmann, 1974; Mook, 1974). Since such voids are always faceted rather than spherical and are oriented with respect to the crystalline directions of the metal, the small-angle scattering from irradiated single crystals is highly anisotropic. Because of this anisotropy, and because of the experience of Epperson et al. (1974) which indicated that even with single crystals double Bragg scattering could not be avoided in a conventional long-slit geometry, a new small-angle X-ray scattering instrument which circumvents these problems by utilizing pin-hole collimation and a position-sensitive proportional counter has been developed.

It is the purpose of this paper to describe the new facility, to illustrate its performance with four examples from the fields of polymer science, biology, chemistry, and materials science, and to discuss its advantages and shortcomings.

Description of facility

The X-ray source is a Rigaku-Denki RU-3V 6 kW rotating-anode generator. Both Mo and Cu targets have been used operating at 48 kV or 40 kV and 100 mA, respectively. The incident radiation is monochromatized with a compression annealed pyrolytic graphite crystal (Sparks, 1966). A schematic outline of the system is shown in Fig. 1. After monochromatization, the incident beam is monitored with a low-absorption (3%) ion-chamber charge integrator (Hendricks, DeLorenzo, Glass & Zedler, 1973) and is collimated by two apertures separated by 500 mm. The apertures, usually 0.5 or 1.0 mm diameter, are holes in thin lead sheet, 125 \( \mu \)m thick for Mo \( K\alpha \) and 75 \( \mu \)m thick for Cu \( K\alpha \) radiation. These values are sufficient to reduce the incident-beam intensity by \( 10^9 \) for both radiations. Such thin foils are used because it has been found that the parasitic scattering from the apertures is roughly proportional to their thickness, and results

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from both total reflection and diffraction from uneven surfaces. At present, the integrated parasitic scattering lies between $10^{-5}$ and $10^{-6}$ of the incident beam power, and is mainly concentrated near the direct beam. The sample, whose optimum position is directly behind the second aperture, is mounted on a goniometer in a large evacuated chamber (100 $\times$ 100 $\times$ 100 mm). The scattered radiation traverses a long (1000 or 2000 mm) evacuated flight path which is connected to the specimen chamber by a flexible metal bellows, and exits from the system through a thin (0-5 mm) beryllium window of 100 mm diameter. It should be noted that the entire X-ray path is evacuated to about 10$^{-2}$ torr from the first aperture to the exit window. The beam stop and one-dimensional position-sensitive proportional counter are mounted on an x-y-z-$\phi$ positioning device, and are fixed with respect to each other. In front of the counter are two parallel edges whose separation is adjustable. The optimum width of this slit is given by the full width at half maximum intensity of the primary beam at the beam stop (4 mm). The positioning device permits (i) centering the beam stop in the primary beam (x-y motion) (ii) rotating the detector about the primary beam ($\phi$ motion) in order to align it with respect to the specimen, and (iii) placing the beam stop directly against the exit window (z motion) in order to trap scattering of the primary beam from both the beryllium window and from air behind it. To center the beam stop in the primary beam, both in the horizontal and vertical directions, it has been found to be most convenient to symmetrize the very intense isotropic SAXS from a piece of glassy carbon (Perret & Ruland, 1972).

The detector is a Borkowski–Kopp one-dimensional position-sensitive proportional counter, and consists of a xenon-filled (1 atm) beryllium tube (10 mm diameter $\times$ 100 mm long). The efficiency is 21% for Mo $K\alpha$ and 85% for Cu $K\alpha$ radiation. It is operated with a bias of $-1400$ V on the cathode, and the high-resistance graphite-coated SiO$_2$ anode is DC-coupled to two preamplifiers, one at each end. The position of an absorbed photon is determined from the difference in rise times of the two pulses obtained from each end of the detector as described by Borkowski & Kopp (1968, 1970). The electronic components are all commercially available NIM modules, and a block diagram of the circuit is shown in Fig. 2. The energy resolution, measured with the sum-pulse, is 15% for both radiations, and compares well with that of a conventional Xe-filled proportional counter. This is sufficient to discriminate against harmonic wavelengths in the Bremsstrahlung radiation diffracted by the graphite crystal. The spatial resolution of the detector is 0.4 mm FWHM, while at 1% of peak maximum it is 1.6 mm. The electronics were adjusted so that the central 80 mm of the detector were recorded in 100 channels of a multichannel analyzer, thus giving 0.8 mm/channel. This distribution was chosen in order that the effective resolution of the detecting system is essentially determined by the channel width and not by the complex resolution function of the position-sensitive counter. In addition, there is no advantage to have the resolution of the detecting system much better than the width of the incident beam at the beam stop.

**Calibration**

A small-angle scattering experiment performed with this device produces a distribution of photon count rate versus channel number. To interpret such data as an X-ray scattering cross section versus scattering vector $\kappa$, three parameters must be calibrated. These are (i) the end-to-end sensitivity of the detecting system, (ii) the correspondence between the scattering vector and the channel number, and (iii) the power of the incident beam.

The sensitivity of the detector along its length is most easily determined by measuring a source which emits radiation uniformly to the detector. For this purpose, a radioactive $^{35}$Fe source (10 $\mu$Ci) placed approximately 500 mm from the detector is used. The difference in energy of the Mn $K\alpha$ (5.90 keV) radiation from this source and the Mo $K\alpha$ or Cu $K\alpha$ radiation is taken into account by appropriately adjusting the gain of the three pulse-shaping amplifiers. In this

![Fig. 1. Schematic representation of the new small-angle X-ray scattering facility.](image1)

![Fig. 2. Block diagram of the position-sensing electronics. The top and bottom rows of components determine the difference in rise-times of the detector, and thus the position of the incident photon. The middle row, which is the sum pulse, is used for energy discrimination in the usual manner. The components are: $\Sigma$, summing amplifier; SCA, single-channel analyzer; TAC, time-to-amplitude converter: MCA, multichannel analyzer.](image2)
detector, the sensitivity has been found to be uniform within 10%. Variations result from localized fluctuations of resistance along the wire which are caused by uneven deposition of pyrolytic graphite on the quartz fiber. Because we have found these variations to be stable in time, the data from such a calibration are used to correct routinely all scattering patterns to what would be recorded with a detector of uniform sensitivity.

To obtain the correspondence between the channel number and the position of photon absorption, a lead sheet, in which 16 uniformly spaced parallel slits have been cut, is mounted in front of the detector. These slits are 1 x 10 mm and spaced 6-0 mm apart. The detector, with the raster fixed in front of it, is then translated across the incident beam. A typical result is shown in Fig. 3. The calculated centroids of the peaks and the known slit positions are used to determine, by a least-squares fit, the coefficients of a cubic polynomial. It has been found that linear and quadratic polynomials do not adequately describe the required function; however, the deviations from linearity are less than 10% (see Fig. 3). With the cubic polynomial, positions can be determined with a standard deviation of less than 0.1 mm.

The absolute intensity was calibrated using octafluorocyclobutane gas $C_4F_8$ as described by Shaffer & Beeman (1970), Hendricks & Shaffer (1971), and Hendricks (1972). Because of the weak scattering from this gas, a piece of polyethylene (Lupolen) was also calibrated as a secondary standard. This material gives sufficient intensity to rapidly calibrate our machine, even at the 2 m specimen-to-detector length. The material and procedures used in this secondary calibration experiment were identical with those described by Shaffer & Hendricks (1974); however, in the present case the arguments regarding collimation corrections are unnecessary. From the results it was determined that with apertures of 1-0 mm diameter and 0-8 mm diameter spaced 500 mm apart, there are $1 \times 10^7$ Cu K$\alpha$ photons s$^{-1}$ incident upon the sample when the source is operating at 60 kV and 100 mA.

**Comparison with a Kratky camera**

For a comparison with the Kratky camera, a special form of polyethylene (Lupolen) has been examined. Apart from its inherent interest as a polymer, Lupolen has been widely used as an absolute intensity standard (Kratky, 1963) and is thus well known in many laboratories. In addition, for the present comparison purposes, it is especially useful because the lamellar crystals which have a spacing of 200 Å cause a peak in the small-angle scattering function. In contrast to a Gaussian scattering function which remains unchanged in shape on going from a point to a long-slit geometry, the scattering function of polyethylene changes drastically thus giving a more rigorous test of the two instruments. Fig. 4(a) shows the measured scattering from a Lupolen sample in a modified Kratky camera (Hendricks, 1970). The absolute intensity was calibrated with $C_4F_8$ gas, and collimation corrections were performed with the program developed by Schmidt (1965). The resulting scattering cross section is included in Fig. 4(a). The same sample was measured to the same minimum angle in the new facility, and the resulting absolute scattering cross section is shown in Fig. 4(b). Although there is a very large difference between the shape of the scattering curve as measured in the Kratky camera and the true scattering function, it can be seen that this scattering function is directly observed in the new facility. Detailed comparisons of the two results reveal that the collimation corrected function is about 10% narrower than the directly observed function at one-half maximum intensity. Following the discussion in the Appendix, since for this sample $\langle \mu^2 \rangle/e^2$ and $\langle \mu^2 \rangle/e^2$ are both less than 0.01 at the peak maximum, it is easily shown that the application of collimation corrections to our directly observed results has a negligible effect except on the two data points nearest the origin, and thus cannot account for this effect. Three other possible sources of difference
can be envisioned; (i) there were systematic errors resulting from non-simultaneous detection during the measurement with the Kratky camera (e.g., fluctuations in beam power), (ii) there were systematic errors in the desmearing procedure, or, (iii) the sample is slightly non-uniform, and since we measured the scattering from a larger volume in the Kratky camera ($\approx 12 \text{ mm}^3$) than in the new facility ($\approx 1.5 \text{ mm}^3$), the difference may be real. It is not within the scope of this paper to pursue these small differences.

Because this specimen was measured in absolute units on both instruments, it is possible to make a direct comparison of their performance. Our modified Kratky system with its 1400 W sealed Cu $K\alpha$ source is typical of many long-slit small-angle scattering instruments in current use. The scattered power $P(\kappa)$ is [e.g., Hendricks, 1972; equation (4)].

$$P(\kappa) = \varphi F_1 \Delta \Omega_1 t \exp \left( -\mu t \right) \frac{d \Sigma}{d \Omega} (\kappa) \Delta \Omega_2$$

(1)

with

$$\kappa = \frac{4\pi}{\lambda} \sin \left( \varepsilon/2 \right)$$

(2)

and where $\varphi$ is the apparent brilliance of the focal spot, $F_1$ is the area of the entrance slit, $\Delta \Omega_1$ is the solid angle of the incident beam, and $\Delta \Omega_2$ is the solid angle subtended by the detector slit. In the pin-hole geometry $d \Sigma/d \Omega$ is the scattering cross section per unit volume of a specimen of thickness $t$ and linear absorption $\mu$.

In the long-slit geometry, the integrated scattering cross section taking into account the slit-length smearing must be substituted for $d \Sigma/d \Omega$. In Table 1 numerical values are presented for these parameters for each instrument. In the example data were recorded for the same total measuring time. Note that the same number of data points were recorded on one side of the direct beam. However, in the new facility data are recorded on both sides of the beam. This capability has been found to be valuable in aligning the instrument.

This comparison reveals two interesting features of our machines. First, if the apparent brilliance of the special Kratky tube is scaled to the same operating conditions as the rotating anode, one finds

$$80 \text{ mA} \times \frac{2.8 \times 7 \text{ mm}^2}{30 \text{ mA} \times \frac{1 \times 10 \text{ mm}^2}{1 \times 10 \text{ mm}^2}} \times 0.32 \times 10^{12} \text{ photons s}^{-1} \text{ sterad}^{-1} \text{ mm}^{-2}$$

$$= 1.79 \times 10^{12} \text{ photons s}^{-1} \text{ sterad}^{-1} \text{ mm}^{-2}.$$ This result implies that the Cu $K\alpha$ production efficiency is essentially identical in the two sources. Second, the two experimental curves (Fig. 4) have been measured to the same statistical accuracy in the same total measuring time. However, in the new facility the actual scattering cross section $d \Sigma/d \Omega$ has been measured, while with the Kratky camera $d \Sigma/d \Omega$ must be recovered by a desmearing procedure which cannot improve the statistics. This effect has been studied quantitatively by Lin, Von Bastian & Schmidt (1974).

Table 1. Comparison of measurements of polyethylene in the Kratky camera and in the new facility

<table>
<thead>
<tr>
<th>Property</th>
<th>Property</th>
<th>Pin-hole collimation and position-sensitive detector</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kratky camera</td>
<td>Radiation characteristics</td>
<td>Property</td>
</tr>
<tr>
<td>Siemens special Kratky tube</td>
<td>X-ray source</td>
<td>Rigaku-Denki 6 kW rotating anode</td>
</tr>
<tr>
<td>2.8 x 7 (40,30)</td>
<td>Focal spot size ($\text{mm}^2$) (kV, mA)</td>
<td>1 x 10 (40, 80)</td>
</tr>
<tr>
<td>Graphite (diffracted beam)</td>
<td>Monochromator</td>
<td>Graphite (incident beam)</td>
</tr>
<tr>
<td>0.32 x 10^{12}</td>
<td>$\varphi$, apparent brilliance*</td>
<td>1.77 x 10^{12}</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Collimation conditions</th>
<th>$\epsilon_{\text{min}}$, minimum angle to which data are recorded (mrad)</th>
<th>2.76</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.098 x 10 = 0.98</td>
<td>$F_1$ area of entrance slit ($\text{mm}^2$)</td>
<td>$\frac{\pi}{4} \left( 1 \right)^2 = 0.79$</td>
</tr>
<tr>
<td>0.222 x 18 (= 48 \times 10^{-6}) (= \frac{288}{288})</td>
<td>$\Delta \Omega_1$, solid angle of incident beam (sterad)</td>
<td>$\frac{\pi}{4} \left( \frac{0.8}{500} \right)^2 = 2.0 \times 10^{-6}$</td>
</tr>
<tr>
<td>0.633 x 10 (= 94 \times 10^{-6}) (= \frac{259}{259})</td>
<td>$\Delta \Omega_2$, solid angle of receiving slit (sterad)</td>
<td>(0.8 \times 3 (1160)^2 = 1.8 \times 10^{-6})</td>
</tr>
<tr>
<td>15.0 x 10^{6}</td>
<td>$I_0$, power incident on specimen (photons s^{-1})</td>
<td>2.8 x 10^{6}</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Measurement conditions</th>
<th>Accumulated counts at peak of scattering curve</th>
<th>12000</th>
</tr>
</thead>
<tbody>
<tr>
<td>11000</td>
<td>Sample transmission</td>
<td>0.363</td>
</tr>
<tr>
<td>45</td>
<td>Total data points</td>
<td>2 x 45</td>
</tr>
<tr>
<td>60</td>
<td>Total measuring time (min)</td>
<td>60</td>
</tr>
</tbody>
</table>

* This value, computed from absolute intensity calibrations, does not include the effects of Be window absorption, the efficiency of the monochromator (0.1-0.3), or the effect of take-off angle (6'). It is the apparent value at the entrance slit, and not the true brilliance of the focal spot.
Comparison with a neutron facility

In this section the performance of the new X-ray facility is compared with the small-angle neutron scattering facility in Jülich. To do this, new X-ray results are presented for scattering from a neutron-irradiated aluminum single crystal which has also been examined by transmission electron microscopy (TEM) and neutron scattering (Hendricks, Schelten & Schmatz, 1974). It is not the purpose of this paper to argue the general merits of either technique; this has been done recently by Schmatz, Springer, Schelten & Ibel (1974). However, because it has been demonstrated that voids in neutron-irradiated materials can be investigated by both techniques, it is essential to have a quantitative measure of the performance of each in order to extrapolate their applicability to other important areas of radiation-damage research.

Eleven single crystals of high-purity aluminum were irradiated to fluences between 0.3 and 2.0 × 10²¹ n cm⁻² in the Oak Ridge High Flux Isotope Reactor. The results of a neutron small-angle scattering investigation of the void structure developed in these crystals has been reported in detail elsewhere (Hendricks, Schelten & Schmatz, 1974; Hendricks, Schelten & Lippmann, 1974). For the present comparison, we have chosen to examine a thinned piece (0.074 mm thick) of specimen A1–6 for which the void properties have been shown to be almost identical with specimen A1–5. The absolute X-ray scattering cross section for this sample, which was determined in a single run over approximately four decades, is presented in Fig. 5. However, from the Guinier plot one sees that at the best resolution which can currently be achieved, data are recorded only to ½dS(0)/dl² for this sample whose voids have a radius of gyration of 213 Å. From these data, the forward scattering, the volume fraction of voids (swelling), and the specific void surface have also been computed using standard techniques. The results of the present work, based on a data collection time of 60 min, are compared with our neutron data in Table 2, and are seen to be in good agreement.

It is especially important to observe that this X-ray experiment was unaffected by double Bragg scattering. Such was not the case in the earlier results of Epperson, Hendricks & Farrell (1974) which were recorded in a long-slit geometry. In the long-slit geometry, even when single crystals are used, the Ewald sphere is

\begin{table}[h]
\centering
\begin{tabular}{|l|c|c|}
\hline
Property & X-rays & Neutrons \\
\hline
dS(0)/dl², forward scattering* & (2.7 ± 0.2) × 10⁴ × (NZrT)² & (2.60 ± 0.05) × 10⁴ (Nbcoh)² \\
\hline
R₀, radius of gyration (Å) & 213 ± 10 & 215 ± 4 \\
\hline
\frac{dV}{V}, specific void volume (swelling) (%) & 0.72 ± 0.08 & 0.78 ± 0.04 \\
\hline
\frac{S}{V}, specific void surface (m² cm⁻²) & 1.4 ± 0.3 & 1.3 ± 0.1 \\
\hline
\end{tabular}
\caption{Comparison of the small-angle X-ray scattering result for specimen A1–6 with the neutron small-angle scattering result for specimen A1–5}
\end{table}

* N = number density of Al atoms = 6.06 × 10²² cm⁻³; Z = number of electrons per Al atom = 13; rₜ = scattering amplitude of a classical electron = 2.82 × 10⁻¹³ cm; and bcoh = nuclear coherent scattering amplitude of Al = 3.441 × 10⁻¹⁵ cm.

Fig. 4. Comparison of the scattering from polyethylene as measured in the Kratky camera and the new facility. (a) Kratky camera, observed intensity, J(κ) corrected for sample absorption, background, and converted to absolute units; true scattering function dS/d⁴Ω recovered from J(κ) by the method of Schmidt (1965). (b) Pin-hole collimation, directly observed scattering function dX/d⁴Ω; scattering function dZ/d⁴Ω obtained from Kratky camera [same as (a)].

Fig. 5. Small-angle X-ray scattering from voids in a neutron-irradiated aluminum single crystal (fluence= 2.0 × 10²¹ n cm⁻², E > 0.18 MeV). (a) scattering function recorded in a single run; (b) Guinier plot of innermost points.
significantly broadened by the $\pm 10^\circ$ angular divergence of the incident and scattered rays in the slit-length direction. Under such conditions it is almost impossible to orient most single crystals so as to avoid double Bragg scattering. However, in the present system such broadening of the Ewald sphere does not occur and one can almost always find an orientation for which there are no Bragg reflections. Even with polycrystalline specimens, the elimination of the large angular divergence in the length direction has a significant effect in reducing double Bragg scattering. Similar arguments, which will be developed in a forthcoming publication, show that multiple diffuse scattering from amorphous materials [such as encountered by Hendricks, Mardon & Shaffer (1974) in their study of liquid water, and by Bates, Shaffer & Hendricks (1974) in their study of quartz] can be significantly reduced.

It is now possible to compare quantitatively the performance of the new X-ray facility and the small-

| Table 3. Comparison of measurements of voids in neutron-irradiated aluminum single crystals by X-ray and neutron small-angle scattering |

| Neutron small-angle scattering facility at the FRJ-2 reactor in Kernforschungsanlage, Jülich | Property | New small-angle X-ray scattering facility in Oak Ridge National Laboratory |
| Radiation characteristics | Radiation characteristics | |
| 23 MW DIDO reactor with liquid H₂ cold source | Source | Rigaku-Denki 6 kW rotating anode with 1 × 10⁻³ m² focal spot |
| Bent nickel-coated glass neutron guide (40 m) and velocity selector ($\Delta \lambda / \lambda \approx 0.1$) | Monochromatization | Compression-annealed pyrolytic graphite ($\Delta \lambda / \lambda \approx 10⁻³$) |
| 8-0 | $\lambda$, wavelength (Å) | 1.542 |
| $5.6 \times 10^6$ | $\varphi$, apparent source brilliance* (particles s⁻¹ sterad⁻¹ mm⁻²) | $3.8 \times 10^7$ |
| Collimation conditions | Collimation conditions | |
| $20 \times 80 = 1600$ | $F_s$, area of entrance slit (mm²) | $\pi \left( \frac{d}{2} \right)^2 = 0.20$ |
| $10 \times 30$ | $\Delta \theta_s$, solid angle of incident beam (sterad) | $\pi \left( \frac{d}{2} \right)^2 \frac{1}{500^2} = 0.78 \times 10^{-6}$ |
| ($600\Omega$) | $\Delta \theta_s$, solid angle of receiving slit (sterad) | $0.8 \times 5 = 0.95 \times 10^{-6}$ |
| $8 \times 40$ | $I_0$, power incident on specimen (particles s⁻¹) | $5.7 \times 10^8$ |
| ($500\Omega$) | | $2055$ |
| $0.75 \times 10^4$ | Resolution | $5.1 \times 10^{-6}$ |
| $5.1 \times 10^{-6}$ | $\Delta K_i^2$, spread in wave vector of incident beam (Å⁻²)† | $13.0 \times 10^{-6}$ |
| $8.0 \times 10^{-6}$ | $\Delta K_i^2$, spread in wave vector of scattered beam (Å⁻²)‡ | $16.0 \times 10^{-6}$ |
| $4.7 \times 10^{-3}$ | $\kappa_\text{min}$, smallest scattering vector for this geometry (Å⁻¹) | $6.0 \times 10^{-3}$ |
| Specimen parameters | Specimen parameters | |
| $10 \times 30 \times 25 = 7.5 \times 10^3$ | $V$, irradiated volume (mm³) | $\pi \left( \frac{d}{2} \right)^2 0.074 = 1.5 \times 10^{-3}$ |
| 25 | $t$, specimen thickness (mm) | 0.074 |
| 0.8 | $T$, specimen transmission | 0.38 |
| 3.8 | $d\Sigma(k_i)/d\Omega$, cross section per unit volume at $k_i = 0.01$ Å⁻¹ (mm⁻¹) | 510 |
| 76 | $\sum T d\Sigma(k_i)/d\Omega$ | 14 |
| Data recording | Data recording | |
| 50 | $N$, number of data points simultaneously recorded§ | 90 |
| 74 | $P(k_i)$, count rate at 0.01 Å⁻¹ (particles s⁻¹) | 8 |

* This value, computed from the absolute intensity calibrations, does not include the effects of Be window absorption, the efficiency of the monochromator (0.1-0.3), or the effect of take-off angle (6°). It is the apparent value at the entrance slit, and not the true brilliance of the focal spot.

† The apparent brilliance is twice that of Table 1 because of non-uniformities in the incident beam, and better alignment.

‡ $\Delta K_i^2 = (2\pi/k_i)^2 d\Omega_i (i = 1, 2)$.

§ In the neutron machine, there are five parallel one-dimensional position-sensitive detectors, each recording 50 points.
angle neutron scattering facility at the FRJ-2 reactor in Jülich (Schelten, 1972) in manner similar to that used above for comparison with the Kratky camera. Results are presented in Table 3. It is significant to note that the apparent brilliance of the neutron source (for $\lambda=8\,\text{Å}$) is six orders of magnitude smaller than that of the X-ray source (for $\lambda=1.54\,\text{Å}$). Because of the difference in the wavelengths, the collimation of the incident and scattered X-ray beams must be about five times better in both directions to achieve the same resolution. In addition, because the neutrons emanate from a very much larger source area than the X-rays, the neutron collimation is performed with big slits, thus increasing both the power incident on the sample and the number of scatterers illuminated. For these reasons, a difference of six orders of magnitude in apparent brilliance of the sources is reduced to less than one order of magnitude difference in the power incident on the sample. Perhaps the most striking difference between the two experiments is that the volume of the specimen illuminated in the X-ray experiment is $5 \times 10^{-7}$ that illuminated in the neutron experiment. Notwithstanding this difference, it is seen in Table 3 that the scattering probability of the sample [defined as $tT(d\Sigma/d\Omega)\,d\Omega$] is only five times smaller for the X-ray case. Finally, we note that for these two instruments, set up with nearly the same resolution, and starting with radically different apparent brilliances and illuminated sample volumes, the scattered power is not quite ten times smaller with X-rays than with neutrons.

Development of rapid analytical tools for determining properties of voids in accelerator-damaged materials is of crucial importance to the Controlled Thermonuclear Reactor (CTR) and Liquid Metal Fast Breeder Reactor (LMFBR) programs. With current techniques, irradiated specimens are usually of the order of $1\,\text{mm}^2$ in cross section and the damage layer is about $1\,\mu\text{m}$ thick. From the above analysis, it is clear that such samples cannot be investigated with neutrons because their scattering power would be at most only $10^{-7}$ of that in Table 3. However, for X-rays the scattered power will be reduced by only a factor of 100 from that cited in Table 3. Developments necessary to meet this requirement are in progress. A factor of 30 increase in the number of detected photons is expected when the present one-dimensional detector is replaced with a two-dimensional position-sensitive proportional counter which is now under construction, and another factor of 10 to 20 can be anticipated from the use of $60\,\text{kJ}$ or $100\,\text{kJ}$ rotating anode X-ray generators now commercially available.

**Two further examples**

One of the classical applications of small-angle X-ray scattering is to the study of particles in aqueous suspension. To exemplify these systems, the scattering from a dilute Ludox solution ($1\%\,\text{SiO}_2$ spheres in $\text{H}_2\text{O}$) has been measured. Results of a 10 min measurement at 1 m specimen-to-detector distance are shown in Fig. 6. Two features of the scattering curve are immediately apparent: (i) the intensity in the forward direction is reduced significantly from what would be obtained by a Guinier extrapolation, and (ii) oscillations of the scattering curve about the Porod law are clearly visible. Measurements on several samples of differing concentrations have indicated that the first effect is the result of interparticle interference. On the assumption that the data in the angular region $0.017<\kappa<0.030\,\text{Å}^{-1}$ are not seriously affected by interparticle interference effects, we have extrapolated them to zero angle (arrow in Fig. 6) using the Guinier approximation. From these data, and the known electron density of SiO$_2$, the volume fraction of SiO$_2$ is computed to be 1.1\%, the mean particle volume to be $3.0 \times 10^8\,\text{Å}^3$, and the radius of gyration to be 78 Å. The appearance of the oscillations in the tail of the scattering curve suggests that the particle size distribution is nearly monodisperse. Assuming the particles to be spheres of radius $R$, the first minimum in the Rayleigh scattering function which occurs at $\kappa R=4.94$ is computed from the radius of gyration to occur at $\kappa=0.049\,\text{Å}$. This value is shown as $\kappa_1$ in Fig. 6, and is

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seen to agree well with the observed inflection in the data.

As a final example, a small-angle diffraction pattern obtained from tendon collagen is presented. Bartlett, Egelstaff, Holden, Stinson & Sweeny (1973) have studied the structural changes in tendon collagen resulting from nutritionally induced muscular dystrophy in ducks. In that work, moist tendons were examined in a long-slit geometry and, in agreement with earlier investigations, only odd-order reflections from the 670 Å periodicity were observed. The effect of muscular dystrophy was to reduce greatly the peak intensity of these reflections, apparently by increasing the mosaic spread of the collagen fibers. In order to investigate this problem further, and in an attempt to increase the sensitivity of the SAXS experiment, Stinson (private communication) has performed an isomorphic replacement by soaking the tendons in uranyl acetate. He has found that such treatment drastically changes the intensities of the various lines. With this technique, the small-angle X-ray scattering from a tendon taken from a dystrophic 18 day old duck was measured. The pattern obtained with the detector axis parallel to the fiber axis is shown in Fig. 7. Note that the second-, fourth- and sixth-order peaks are clearly visible, while the fifth-order peak has almost completely disappeared. The d spacing obtained from the sixth-order peak is 628 Å. These observations are consistent with those made independently by Stinson on his long-slit machine. However, our value of 628 Å for the d spacing is 3 % lower than his value of 648 Å. Both values are lower than the 673 Å reported by Bartlett et al. (1973), and can be explained because our specimen was vacuum-dried, Stinson’s was air-dried, but Bartlett et al. measured moist tendons. The data in Fig. 7 were recorded in 10 h, as compared to the usual 24 to 48 h measuring time required by Bartlett et al. (1973).

Conclusions

The above examples demonstrate that our new instrument can measure true small-angle X-ray scattering cross sections, dΣ/dΩ, unencumbered by the effects of slit smearing and multiple Bragg scattering, more rapidly and with better precision than a conventional long-slit geometry which records only a complex function of the true scattering dΣ/dΩ. Further advantages are gained in that (i) there are no moving parts, (ii) alignment is simple and rapid, (iii) fluctuations in incident power level do not distort the scattering pattern, and (iv) simultaneous detection and display of data allow early judgment of the quality of an experiment.

Notwithstanding these advantages, the present facility is limited by insufficient resolution and a somewhat high parasitic scattering from slit edges. The former is determined, for the present, by the physical size of the laboratory housing the experiment. Special optically polished slit edges, as developed by Dr A. Franks (private communication), will reduce parasitic scattering by about an order of magnitude.

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Fig. 7. Small-angle X-ray diffraction pattern from tendon collagen. Sample, obtained from a dystrophic 18 day old duck, was stained with uranyl acetate to enhance the scattering from the even-index lines. Without staining, only odd-index lines are discernible. The measurement time was 600 min. The d spacing, determined from the sixth-order reflection, is 628 Å.
APPENDIX

Estimation of collimation corrections

Following the derivation of Hendricks (1972), the measured small-angle scattering \( J(\epsilon) \) and the desmeared intensity function, \( I(\epsilon) \), are related by

\[
J(\epsilon) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} W_\omega(u) W_\phi(u') I[(\epsilon - u)^2 + u'^2] du du' \quad (A1)
\]

where \( W_\omega(u) \) and \( W_\phi(u') \) are slit-width and slit-length weighting functions (Hendricks & Schmidt 1967, 1973). In the present case, \( W_\phi(u') \) has a similar width to \( W_\omega(u) \) in contrast to the more usual long-slit geometry where it may be 100 times wider. Hence, \( u' < \epsilon \) and it is thus possible to expand the scattering function as a Taylor series about the point \( \epsilon \) in terms of \( u/\epsilon \) and \( u'/\epsilon \) as was originally done for only the slit-width correction by Kratky, Porod & Skala (1960) and Taylor & Schmidt (1967). The result is

\[
J(\epsilon) = I(\epsilon) + \frac{\epsilon I'(\epsilon)}{2} \left[ \frac{\langle u_0^2 \rangle}{\epsilon^2} \right] + \frac{\epsilon^2 I''(\epsilon)}{2} \left[ \frac{\langle u_0^4 \rangle}{\epsilon^4} \right] + \ldots, \quad (A2)
\]

where \( \langle u_0^2 \rangle \) and \( \langle u_0^4 \rangle \) are the second moments of the slit-width and slit-length weighting functions. In this derivation, the symmetry of the collimation system insures that the first moments of the weighting functions, \( \langle u_0 \rangle \) and \( \langle u_1 \rangle \), are zero. The recovery of \( I(\epsilon) \) from this equation follows the earlier slit-width procedure in that derivatives of the observed function \( J(\epsilon) \) are used as the starting point. If necessary, iterations can be computed until \( I(\epsilon) \) converges within suitable limits. This method is adaptable to minicomputers as opposed to the usual large core requirements for most desmearing procedures.

To estimate the magnitude of these corrections we note that the right-hand side of equation (A2) may be written as

\[
J(\epsilon) = \frac{1}{2} I(\epsilon) \left[ (\epsilon + \langle u_0^2 \rangle)^{1/2} + \langle u_1^2 \rangle \right] + \frac{1}{2} I(\epsilon) \left[ (\epsilon - \langle u_0^2 \rangle)^{1/2} + \langle u_1^2 \rangle \right] \quad (A3)
\]

In our new facility \( L \langle u^2 \rangle \), which is the variance of the primary beam in either the width or length direction at the detector, is less than 1 mm (\( L \) is the specimen-to-detector distance).

References


