Small-Angle Scattering at a Pulsed Neutron Source: Comparison with a Steady-State Reactor

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Abstract
A time-of-flight small-angle diffractometer employing seven tapered collimator elements and a two-dimensional gas proportional counter was successfully utilized to collect small-angle scattering data from a solution sample of the lipid salt cetylpyridinium chloride, C_{21}H_{38}N^+.Cl^-, at the Argonne National Laboratory prototype pulsed spallation neutron source, ZING-P'. Comparison of the small-angle scattering observed from the same compound at the University of Missouri Research Reactor corroborated the ZING-P' results. The results are used to compare the neutron flux available from the ZING-P' source relative to the well characterized University of Missouri source. Calculations based on experimentally determined parameters indicated the time-averaged rate of detected neutrons at the ZING-P' pulsed spallation source to have been at least 33%, higher than the steady-state count rate from the same sample. Differences between time-of-flight techniques and conventional steady-state techniques are discussed.

Introduction
The systematic development of pulsed spallation neutron sources has been underway at Argonne National Laboratory since 1972, and the second-phase prototype source, ZING-P' (Carpenter, Kleb, Kustom & Ostrowski, 1980), recently concluded operation. A time-of-flight (TOF) small-angle instrument was developed at this facility and tested for approximately two months prior to discontinuing ZING-P' operations. The principles of the instrument have previously been described by Carpenter & Faber (1978). The results reported herein are the first experimental demonstration that several unique features of the instrument can be utilized with time-of-flight instrumentation for small-angle studies.

The compound studied was cetylpyridinium chloride (CPC), which is known to aggregate into micellular structures (Luzzatti, Mustacchi, Skoulios & Husson, 1960; Reiss-Husson & Luzzatti, 1964) in solution. As previous neutron scattering results had been obtained for this compound at the University of Missouri Research Reactor (MURR), the results at the pulsed source can be compared directly to those of a well understood conventional steady-state facility. The comparison allows extrapolation of these results to evaluate the potential of the instrument at the Argonne pulsed spallation neutron source, IPNS-I, and forms a basis for comparison with other facilities.

Methods and instrumentation
Time-of-flight technique
Krueger, Meneghetti, Ringo & Winsberg (1949) were the first to detect the small-angle scattering of neutrons, using, in fact, a TOF method. More recently neutron TOF methods have been described by Cser (1976), Mildner (1978), Carpenter (1978), Carpenter & Faber (1978), and Seeger (1981). While TOF and
steady-state instruments appear superficially similar, they operate on somewhat different principles. In fact the software developed to analyze the ZING-P' data was originally a modification of code provided by Hendricks (1978) for reactor data. However, an additional normalization for the incident intensity distribution must be provided for in the analysis of the TOF data.

This requirement can be understood by considering the time-averaged counting rate, $C_{ijk}$, of the TOF instrument in the $i$th spatial element of the detector and in the $k$th wavelength channel, which can be expressed as

$$C_{ijk} = \int d\lambda \int d\alpha \int d\theta \Phi(\lambda) A S \frac{d\Sigma}{d\Omega}(\theta) \eta(\lambda) L^{-2},$$

where $\Phi(\lambda)$ is the time-averaged flux on the sample per unit wavelength in units of neutrons cm$^{-2}$ s$^{-1}$ Å$^{-1}$, $A$ is the sample area in cm$^2$, $s$ is the sample thickness in cm, $d\Sigma/d\Omega(\theta)$ is the probability per unit distance per steradian that a scattering event will yield a wave-vector change of scalar magnitude $Q$ expressed in units of cm$^{-1}$ s$^{-1}$, $\eta(\lambda)$ represents the detector efficiency as a function of wavelength, and $L$ is the distance from the sample to the detector. Expressed in a manner more amenable to numerical analysis this becomes

$$C_{ijk} = \Phi(\lambda_k) A \eta(\lambda_k) A S \frac{d\Sigma}{d\Omega}(\theta_{ijk}) \Delta Q_{ij},$$

where $Q_{ijk}$ is the magnitude of the wave-vector change observed at the center of the $ijk$th detector element of the $k$th time channel, i.e.

$$Q_{ijk} = \frac{2\pi r_{ij}}{L_{\lambda_k}},$$

where $r_{ij}$ is the radial distance of this detector element from the center line of the beam, and

$$\Delta Q_{ij} = \frac{\Delta x_i \Delta y_j}{L^2}.$$

The analogous expression for the counting rate of the steady-state (SS) instrument is

$$C_{ij} = \Phi_0(\lambda_0) \eta(\lambda_0) A S \frac{d\Sigma}{d\Omega}(\theta_i) \Delta Q_{ij},$$

where $\Phi_0(\lambda_0)$ is the total flux of neutrons centered about the wavelength, $\lambda_0$, satisfying the Bragg condition of the monochromator, with bandwidth $\Delta\lambda_0$ in units of neutrons cm$^{-2}$ s$^{-1}$, and the other quantities are defined similarly to those in (1), recognizing that there is only a single wavelength.

A useful characterization of these counting rates is to sum all count rates contributing to the same $Q$ interval and to represent this total in a manner that separates the inherent scattering probability of the sample from the instrumental contribution to the count rate. By separately combining the count rates for each instrument in such a manner, a meaningful comparison may be made between the two. The instrumental contribution can be separated by considering the count rate per unit $Q$, defined as

$$C_d(Q_n)_{TOF} = \sum_{ijk} U_{nijk} C_{ijk}(Q_{ijk})$$

for the TOF instrument, where

$$U_{nijk} = 1 \text{ if } Q_{ijk} \text{ is in } \Delta Q \text{ around } Q_n$$

$$= 0 \text{ otherwise},$$

and

$$C_d(Q_n)_{SS} = \sum_{ij} U_{nij} C_{ij}(Q_{ij})$$

for the steady-state instrument, where

$$U_{nij} = 1 \text{ if } Q_{ij} \text{ is in } \Delta Q \text{ around } Q_n$$

$$= 0 \text{ otherwise}.$$

Substituting (1) and (2) into (3) and (4), respectively, and introducing diffractometer weighting functions, $g_{TOF}$ and $g_{SS}$, allows the count rate per unit $Q$ for each instrument to be expressed as

$$C_d(Q_n)_{TOF} = g_{TOF}(Q_m, \Delta Q) A S \frac{d\Sigma}{d\Omega}(Q_n)$$

and

$$C_d(Q_n)_{SS} = g_{SS}(Q_m, \Delta Q) A S \frac{d\Sigma}{d\Omega}(Q_n),$$

where

$$g_{TOF}(Q_m, \Delta Q) = \sum_{ijk} U_{nijk} \Phi(\lambda_k) \Delta \lambda_k \eta(\lambda_k) \Delta Q_{ij}$$

and

$$g_{SS}(Q_m, \Delta Q) = \sum_{ij} U_{nij} \Phi_0(\lambda_0) \eta(\lambda_0) \Delta Q_{ij}$$

contain the instrumental contribution to the count rates. The steady-state diffractometer weighting function, $g_{SS}$, in (8) can be seen to be a simple linear function corresponding to the increasing area subtended by rings of constant width centered around the beam center line at the detector position. The TOF diffractometer weighting function, $g_{TOF}(Q_m, \Delta Q)$, is more complex and can be thought of as a linear superposition of steady-state diffractometer weighting functions, one for each time channel, each of which has a different slope and subtends different intervals along
the $Q$ axis, as the maximum and minimum $Q$ subtended by the detector in a fixed position varies with wavelength in a TOF instrument.

The count rates as expressed in (1)–(8) are nonetheless dependent upon the geometric configurations of the detectors and the details of their construction. To evaluate quantitatively the relative numbers of neutrons scattered by the sample at each instrument, it is necessary to determine the count per unit steradian, independent of the solid angles subtended by the detectors. By denoting the total solid angle subtended by all detector elements in the $k$th time channel that fall within $\Delta Q$ of $Q_n$ as $\Omega_{nk}$, the count rates per steradian can be expressed as

$$\sum_k \frac{C_n(Q_n)_{TOF}}{\Omega_{nk}} = \sum_k \phi(\lambda_0) \Delta \lambda \eta(\lambda_0) As \frac{d\Sigma}{d\Omega} (Q_n).$$

and

$$\frac{C_n(Q_n)_{SS}}{\Omega_n} = \phi(\lambda_0) \eta(\lambda_0) As \frac{d\Sigma}{d\Omega} (Q_n).$$

The reactor instrument is considered similarly to the TOF instrument, but is viewed only as spanning a single time channel.

The scattering probability per unit distance per unit steradian, $d\Sigma/d\Omega'$, can then be obtained for each instrument,

$$\frac{d\Sigma}{d\Omega} (Q_n)_{TOF} = \sum_k \frac{C_n(Q_n)_{TOF}}{\Omega_{nk}} \frac{1}{As} \sum_k \phi(\lambda_0) \Delta \lambda \eta(\lambda_0)$$

and

$$\frac{d\Sigma}{d\Omega} (Q_n)_{SS} = \frac{C_n(Q_n)_{SS}}{\Omega_n} \frac{1}{\Delta \Omega} \frac{1}{As} \phi(\lambda_0) \eta(\lambda_0).$$

The two instruments should, of course, give the same resultant scattering probabilities for equivalent samples.

**Time-of-flight instrument**

Fig. 1 shows the TOF small-angle instrument developed at ZING-P'. As typically employed at a low-intensity source (Nunes, 1974; Nunes & Zaccai, 1976; Nunes, 1978), a set of multiple focusing tapered collimator tubes was employed to increase the count rate at the detector. The collimation system consisted of a bundle of seven individual tubes, each tube comprised of two sections. The taper converged at the detector 3 m beyond the sample position (Carpenter & Faber, 1978). The physical limitations of the experimental area constrained the actual flight paths to the dimensions shown in Fig. 1. A BF$_3$ monitor counter was inserted at the midpoint of the collimation sections.

The detector was a two-dimensional position-sensitive gas proportional counter (Kopp & Borkowski, 1975; Borkowski & Kopp, 1975), filled with $^3$He and with an active area of approximately 17 x 17 cm, centered on the incident neutron beam. A 2.5 cm diameter aluminum cylinder, 5 cm long, and filled with $^6$Li$_2$CO$_3$, was positioned in front of the detector to act as a beam stop. The detector was partitioned into 64 x 64 cells across the active area. The intrinsic spatial resolution was about 2.5 mm. Temporal resolution could be adjusted by factors of two to provide from 1 to 256 time channels. The number chosen during any experiment depended upon the wavelength interval of interest and the anticipated temporal (i.e. wavelength) resolution requirements.

The ZING-P' accelerator was maintained at a constant frequency, $f$, of thirty pulses per second during the course of these small-angle experiments. The condition for avoiding frame overlap is that

$$(m/h)(L_1 + L_2)(\lambda_{\max} - \lambda_{\min}) < 1/f,$$

where $m$ is the mass of the neutron, $h$ is Planck's constant, and $L_1$ and $L_2$ are the moderator-to-sample and sample-to-detector distances, respectively. Although no chopper was available to define $\lambda_{\min}$ and $\lambda_{\max}$ mechanically, $\lambda_{\min}$ could be electronically selected by gating out earlier arriving neutrons. With a total flight path of 11 m this requires $\lambda_{\max}$ to be less than 11 Å. It is clear from Fig. 2 that relatively few 11 Å neutrons were present in the spectrum and their contribution was therefore negligible.

As the beam line is horizontal, corrections for
variations due to gravity in vertical displacement along the active surface of the detector as a function of the neutron time of arrival have been accounted for before assigning a scattering angle to each event. This correction for vertical displacement is only 2.5 mm for the longest-wavelength neutrons in the incident spectrum or about equivalent to the intrinsic detector resolution.

Neutron scattering results

The small-angle scattering results obtained from CPC in 100% D₂O at similar temperatures at both the prototype pulsed spallation neutron source, ZING-P', and the MURR reactor can be expressed in terms of the description provided above. The TOF data, presented in terms of (9), are shown in Fig. 3. The upper curve represents the scattering observed from the sample in 100% D₂O solution. The data were collected over 32 time channels in the wavelength range 0.7 to 3.5 Å. The lower curve is a similar plot obtained for the solvent background, scaled to the same total monitor count, which includes the scattering from D₂O, the sample cell itself, and all other collective parasitic scattering. Both curves are presented at this point to illustrate the signal-to-noise level achieved in the TOF instrument and will eventually be used as a basis of comparison with the MURR steady-state instrument.

If the detector efficiency and the incident neutron beam intensity per unit wavelength were constant over all time channels included within the data set, the curves in Fig. 3 would be proportional to conventional plots of the normalized scattered neutron count as a function of Q over the Q range common to all time channels. The product of the detector efficiency, \[\eta(\lambda_0)\], and the time averaged flux per unit wavelength, \[\Phi(\lambda_0)\], can be calculated as a function of wavelength by using the experimental \(C_n\) values obtained from a well characterized sample such as H₂O or vanadium with a known \(d\Sigma/d\Omega\) from (11). For the present, we have estimated these terms, on a relative basis, by determining the total number of neutrons incident upon the detector in each time channel as already shown in Fig. 2. Note that these data on H₂O were taken over 128 time channels between 0.7 and 5.5 Å so that the appropriate product terms for the CPC data can be determined from them. Equation (11) can then be utilized to calculate \(d\Sigma(Q) d\Omega_{\text{TOF}}\) for CPC.

Because of the large variation in the incident neutron flux, \(\Phi(\lambda_0)\), as a function of wavelength, a substantial difference in statistical accuracy exists among the various contributions to \(d\Sigma/d\Omega\) in (11). An alternative method can be used to weight the separate contributions appropriately. The TOF data can be considered as being grouped into numerous sets, each of which provides a separate estimate of \(d\Sigma/d\Omega\). In fact, if the data from a single time channel form such a group, (11) can be seen to be identical to (12). Thus the TOF information can be considered as several sets of data analyzed as independent monochromatic sets of data, each at a different wavelength.

If several independent distributions of values of a parameter \(f\) are available, then the best estimate of \(f\) can be found from the relation

\[
f = \frac{\sum f_i \sigma_i^2}{\sum i \sigma_i^2},
\]

where \(f_i\) is the mean of the ith distribution and \(\sigma_i^2\) is the corresponding variance. Fig. 4 is a plot of \(d\Sigma/d\Omega\) obtained at each \(Q\) interval in accordance with (13) by statistically weighting the contributions from all wavelength channels that subtend a \(Q\) range overlapping the interval of interest.

Steady-state CPC data collected at the MURR reactor are shown in Fig. 5. The small-angle neutron scattering instrument used to collect these data has been described by Mildner, Berliner, Pringle & King (1981) (see Berliner, Mildner, Pringle & King, 1981 for a discussion of the MURR detector). As \(I_0(\lambda_0)\) and \(\eta(\lambda_0)\) in (10) are independent of \(Q\), Fig. 5 can be seen to be directly proportional to the scattering probability per unit length, \(d\Sigma/d\Omega_{\text{SS}}\) and no further normalization is required. It is sufficient for our present purpose to notice that \(d\Sigma/d\Omega_{\text{SS}}\) and \(d\Sigma/d\Omega_{\text{TOF}}\) correspond well.
Discussion

The data obtained from the scattering of CPC in solution allow a direct experimental comparison to be made between a pulsed spallation source and a steady-state reactor for small-angle scattering purposes. However, this comparison is somewhat difficult as the most appropriate figure of merit would be to compare the number of scattered neutrons per unit solid angle per second with equivalent resolution at the two instruments. The resolutions, of course, are not identical and, in fact, the resolution of the TOF instrument varies somewhat with wavelength. A comparison can be made, nonetheless, by integrating the count rate per steradian per unit time for each instrument over the range of the interesting peak in CPC after subtracting the contribution of the solvent and compensating for the geometrical differences.

The integration interval for comparison was restricted by the narrower $Q$ range of the steady-state instrument as shown in Fig. 5. It can be seen that the useful interval extends from about 0.01 to 0.1 Å$^{-1}$. On the other hand, the pulsed source data shown in Fig. 4 illustrate the wider $Q$ range available with TOF techniques because of the shorter wavelength neutrons in the incident spectrum. Owing to the restricted $Q$ range of the steady-state instrument and the position of the CPC peak in reciprocal space, some of the available neutrons from the pulsed source are not counted in this analysis. Cser (1976) has described how a compensation effect can improve the scattering in the higher $Q$ region by adjusting the position of the Maxwellian contribution of the spectrum. Because the short-wavelength contribution at a pulsed spallation source is even greater than at a pulsed reactor this effect can be concomitantly greater, and the resultant comparison consequently will establish only a lower limit for the pulsed source flux.

Integration of $C_n/\Omega_n$ in Fig. 5 yields:

$$K_{MURR} = \int_{0.01}^{0.1} \frac{C_n}{\Omega_n} dQ = 67.8 \text{ neutrons sr}^{-1} \text{ s}^{-1} (\text{Å}^{-1})^{-1}.$$  

The same integral for the pulsed source can be evaluated by utilizing Fig. 3,

$$K_{ZING-P} = \int_{0.01}^{0.1} \sum_k \frac{C_{nk}}{\Omega_{nk}} dQ = 14375 \text{ neutrons sr}^{-1} \text{ s}^{-1} (\text{Å}^{-1})^{-1}.$$  

The contribution due to the solvent should be subtracted from each of these results to eliminate differences in background levels. The two values then become $K_{MURR} = 31.8$ and $K_{ZING-P} = 8403$ still in the same ratio, as the signal-to-noise levels are equivalent in the two instruments.

The relative numbers of neutrons emitted at the moderators of each source that are useful for small-angle scattering can then be estimated despite differences in the geometrical resolution of the two instruments. The value of the integral for each instrument is directly proportional to the quantity

$$A_1 A_2 sc/L^2,$$

where $A_1$ and $A_2$ are the entrance and exit operative areas of the collimation system, respectively, $L$ is the distance between apertures, and $c$ is the number of collimator elements. Table 1 contains the information necessary to scale these two integral values to equivalent instrument resolution and sample characteristics,
Table 1. Experimental parameters required to compare scattered intensities of the TOF small-angle scattering diffractometer at the Argonne pulsed spallation neutron source with the steady-state instrument at the University of Missouri Research Reactor

<table>
<thead>
<tr>
<th>Parameter</th>
<th>ZING-P</th>
<th>MURR</th>
</tr>
</thead>
<tbody>
<tr>
<td>Entrance aperture area, ( A_1 ) ( (\text{cm}^2) )</td>
<td>4.85</td>
<td>0.78</td>
</tr>
<tr>
<td>Exit aperture area, ( A_2 ) ( (\text{cm}^2) )</td>
<td>0.62</td>
<td>0.20</td>
</tr>
<tr>
<td>Distance between apertures, ( L ) ( (\text{cm}) )</td>
<td>636</td>
<td>450</td>
</tr>
<tr>
<td>Number of collimator elements, ( c )</td>
<td>7</td>
<td>1</td>
</tr>
<tr>
<td>Thickness of sample, ( t ) ( (\text{mm}) )</td>
<td>6</td>
<td>2</td>
</tr>
</tbody>
</table>

and the values become

\[ K_{\text{ZING-P}} = 42.5 \]

and

\[ K_{\text{MURR}} = 31.8 \]

in relative units. Thus the time-averaged count rate of detected neutrons over the restricted \( Q \) range subtended by the CPC peak is approximately 33% higher at the prototype pulsed spallation source than at the medium flux MURR reactor. This would imply that the time required to obtain similar data on instruments configured to provide equivalent resolution would be 33% shorter at the ZING-P' source than at the MURR facility, assuming equivalent background levels. The source production of neutrons at MURR relative to other, more frequently used facilities, can be determined from Table 2 (Mildner, 1982) and thus also compared with the ZING-P' source.

The present analysis provides a quantitative basis for the evaluation of the planned developmental stages of pulsed spallation sources at Argonne. The intense pulsed neutron source, IPNS-1 (Carpenter, Price & Swanson, 1978), planned as the next stage of development, will have increased proton energy from 300 to 500 MeV and increased beam current from 5 to 8 \( \mu \text{A} \). This increase should provide almost three times the number of neutrons that were available at the ZING-P' source. A further improvement in the production of long-wavelength neutrons is expected by utilizing a cold moderator on the small-angle scattering beam line. Most importantly, these calculations indicate that the time-averaged rate of neutron production at the Argonne source will be of sufficient magnitude to utilize TOF techniques to obtain good quality data for experiments designed to exploit unique characteristics of the source, namely the high neutron count per unit wavelength concentrated in the short-wavelength region of the spectrum. Fig. 3 clearly illustrates the neutron count per unit wavelength to be about forty-fold higher at the shortest wavelengths than at the longer wavelengths. Thus if the time-averaged count rate is comparable to the best steady reactors, the short thermal neutron count rate will exceed those rates available at other sources.

Table 2. The effective beam source flux for various well known steady-state reactor sources \( (\text{neutrons cm}^{-2} \text{s}^{-1}) \)

<table>
<thead>
<tr>
<th>Source</th>
<th>Flux ( 10^{14} ) ( \text{cm}^{-2} \text{s}^{-1} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Institute Laue Langevin - ILL</td>
<td>12 * 10^{14}</td>
</tr>
<tr>
<td>Oak Ridge National Laboratory - HIFR</td>
<td>9 * 10^{14}</td>
</tr>
<tr>
<td>Brookhaven National Laboratory - HFBR</td>
<td>6 * 10^{14}</td>
</tr>
<tr>
<td>University of Missouri Research Reactor</td>
<td>1 * 10^{14}</td>
</tr>
</tbody>
</table>

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References


