The small angle neutron scattering extension in MCNPX

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Nanomaterials and particle transport

Motivation

- Detonation nanodiamonds provide an intriguing prospect for the development of a VCN source or as a VCN reflector.
  - Quasi-specular reflection from nanodiamonds has been observed.
- Simulation studies require an implementation of SANS (eg. $S(\alpha, \beta)$ or source code).
- The SANS extension in MCNPX was accomplished by adding an additional scattering process, Σ_{SANS}, to MCNPX.
- New SANS implementation via two methods:
  - Analytical polydisperse hard-sphere model, specifying size, polydispersity, and contrast and using the Schulz-Γ function for particle size.[1].
  - User specified input of reduced SANS data table as $I(q)$ vs. q.

[1] W. L. Griffith, R. Triolo, and A. L. Compere, Phys. Rev. A 35, 2200 (1987).

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Modifications to MCNPX

Code modifications

- New input card, `ms`, that modifies materials similar to `mt` card.
- Routine for calculation of analytical cross section given parameters.
- Routine for determining scattering angle from $I(q)$ vs. q tables.
- Additional SANS term added to neutron cross section determination.

Input parameters

- Analytical hard-sphere model

ζ_μ	particle size
p_0, p_{med}	SLD
z	Schulz polydispersity
η	packing fraction

`ms1` $p \quad \zeta_\mu \quad z \quad \eta \quad p_0 \quad p_{\text{med}}$

- User supplied data file
  - Text file with q and $I(q)$ columns.
  - $I(q)$ should be scaled to proper density before use.

```
ms1 sans.dat
```


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The SANS cross section

Cross sections

$\frac{d\Sigma}{d\Omega}(q)$ differential macroscopic cross section (cm^{-1}ster^{-1}), related to $I(q)$.

Analytical hard-sphere model:

$\frac{d\Sigma}{d\Omega}(q) = \rho \int_0^\infty P_i^2(q) f(\zeta_i)\, d\zeta_i + \rho \int_0^\infty \int_0^\infty P_i(q) P_j(q) H_{ij}(q) f(\zeta_i) f(\zeta_j)\, d\zeta_i$

Σ macroscopic cross section (cm^{-1}), used in MCNPX for mean free path.

$$\begin{aligned}\Sigma_{\text{SANS}}(k_{\text{n}}) &= \int_0^{2\pi} d\phi \int_0^{\pi} \frac{d\Sigma}{d\Omega}(q) \sin\theta \, d\theta \\ &= 2\pi k_{\text{n}}^{-2} \int_0^{q_{\max}} \frac{d\Sigma}{d\Omega}(q)\, q\, dq\end{aligned}$$

σ microscopic cross section (barns), used for scaling data to different ρ.

$\sigma_{\text{SANS}}(k_{\text{n}}) = \frac{\Sigma_{\text{SANS}}}{n}$

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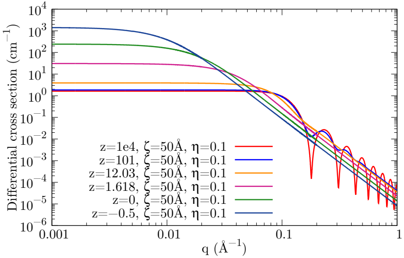
Analytical model - calculated cross sections

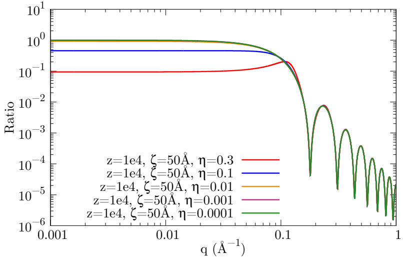
Size distribution: $f(\zeta) = \frac{\zeta^z e^{-\frac{\zeta}{b}}}{b^{z+1}\Gamma(z+1)}$

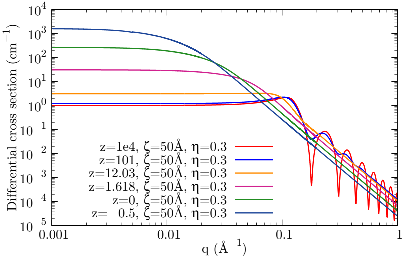
Scattering amplitude:

$P_i(q) = 4\pi p q^{-3}[\sin\left(\frac{q\zeta_i}{2}\right) - \frac{1}{2} q\zeta_i \cos\left(\frac{q\zeta_i}{2}\right)]$

Low-q limit: $I(0) = \eta p^2 V$







The scattering process in MCNPX

Integration

- $\Sigma_{\mathrm{SANS}} = 2\pi k_{\mathrm{n}}^{-2} \int_0^{q_{\max}} \frac{d\Sigma}{d\Omega}(q)\, q\, dq$
- Numerically integrated without k_{n}^{-2} and stored in an array, $C(q_i)$
- A normalized copy on $[0, 1]$:

  $N(q_i) = \frac{C(q_i)}{C(q_f)}$

Cross section determination

- Binary search for $q_{\max} = 2k_{\mathrm{n}}$.
- $\Sigma_{\mathrm{SANS}}(k_{\mathrm{n}}) = k_{\mathrm{n}}^{-2} C(2k_{\mathrm{n}})$

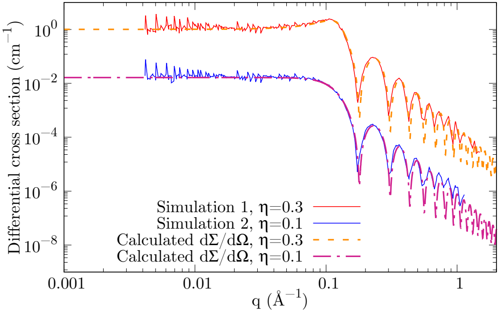
Scattering event

1. Choose uniform random number, c on $[0, \frac{N(2k_{\mathrm{n}})}{N(q_f)}]$.
2. Search for the adjacent bins $(N(l) < N(c) < N(h))$.
3. Interpolate to find fractional location of c: $f = \frac{c - N(q_l)}{N(q_h) - N(q_l)}$.
4. Logarithmic interpolate to find $q_{\mathrm{scatter}} = [q_h]^f\, [q_l]^{1-f}$
5. Find the scattering angle $\theta_{\mathrm{scatter}} = 2 \arcsin \left[\frac{q_{\mathrm{scatter}}}{2k_n} \right]$
6. Rotate by a uniform angle on $[0, 2\pi]$.

Simulation of the analytical hard-sphere model

Analytical model

- Mono-chromatic beam at 2.86 Å, 0.1 mm thick sample, $\zeta = 50$ Å, 2.34×10^{-6} Å^{-2}, $z = 10^4$.
- Surface flux tallied downstream with 10000 cylindrical segments 50 cm, 4 m, and 8 m downstream.
- Simulations were fit using SasView.

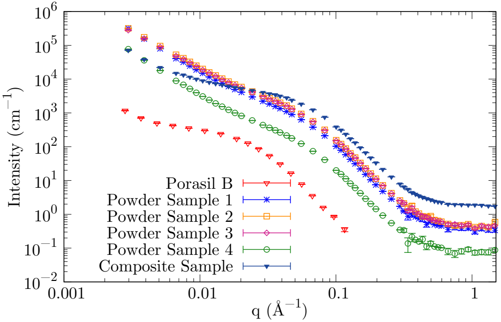


Parameter	Simulation 1	Simulation 2
ζ	50.2 ± 2.2 Å	51.2 ± 6.2 Å
p	$2.42 \pm 0.12 \times 10^{-6}$ Å^{-2}	$2.33 \pm 0.53 \times 10^{-6}$ Å^{-2}
η	0.284 ± 0.023	0.103 ± 0.042

Nanodiamond measurement on EQ-SANS

Samples

- Benchmarking and validation measurement with nanodiamonds on EQ-SANS. using commercial nanodiamonds with size $< 10\text{nm}$.
- Four powder samples, 1 composite aluminum (56.6%)/nanodiamond (43.4%) pellet, porasil calibration standard.
- Initial simulations in MCNPX using this $I(q)$ as $\frac{d\Sigma}{d\Omega}(q)$ resulted in unphysical multiple scattering.



Sample	Thickness (cm)	Density (g cm^{-3})
1	0.076	2.01×10^{-1}
2	0.166	1.60×10^{-1}
3	0.207	1.62×10^{-1}
4	0.132	6.45×10^{-2}
5	0.168	6.00×10^{-1}

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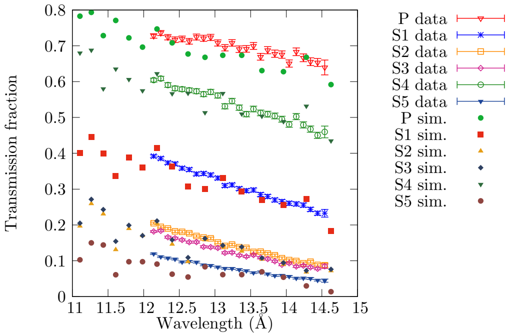
Normalization from transmission

- Measured sample transmission is in disagreement with the mean-free-path as determined from the measured data.
- The integrated $I(q)$ and the measured transmission provide an estimate of the true normalization factor, ≈ 0.11 for sample 4.
- As a check, the transmission can be simulated for a sample of 88% carbon, 1.0% hydrogen, 2.5% nitrogen, and 10% oxygen (all by weight).



- Uncertainty in hydrogen content, sample mass, and sample thickness yields a 20% uncertainty on the SANS cross section for nanodiamonds.

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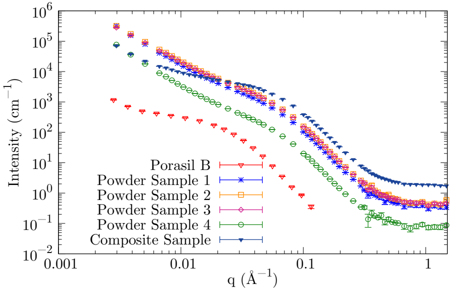
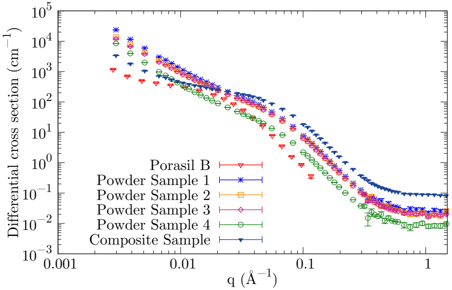
Data after normalization





Input for simulations

- The thinnest sample (4) is fit to a function that is then used to create data files for input in MCNPX.
- Expect that the powder samples could be simulated from sample 4 data with an adjustment for the density.

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Data analysis

Fit function

- Power-law (low-q)

  $I_I(q) = Bq^{-p_{\mathrm{PL}}}$
- Guinier function (mid-q)

  $I_{II}(q) = q^{-s}\exp\left[\frac{-q^2 r_{\mathrm{g}}^2}{3-s}\right]$
- Porod function (high-q)

  $I_{III}(q) = \frac{D}{q^m} + b$

Sample	p_{PL}	s	r_{g}	m
1	2.3613	1.6042	12.791	4.0133
2	2.1231	1.5467	13.285	3.9972
3	2.0781	1.5357	13.372	3.9907
4	2.4894	1.6382	12.551	4.0398
5	2.0521	0.78904	18.019	3.8275

Hard-sphere fit

- Fit sample 4 to polydisperse hard-sphere model in SasView for use in simulations via the HS model in MCNPX.

Parameter	Value
$b(1/\mathrm{cm})$	9.81×10^{-3}
$\rho(\times10^{-6}\,\text{Å}^{-2})$	14.0±0.6
$r_g\,(\text{Å})$	13.2±0.2
η	0.0133±0.001
z	1.064

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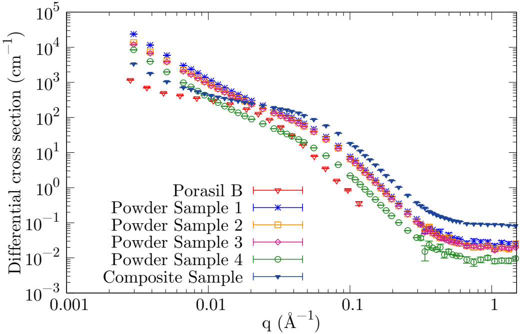
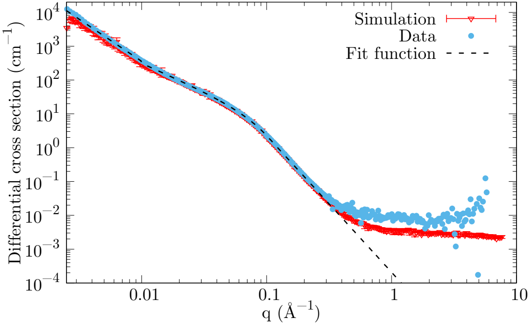
Benchmarking MCNPX to measured data





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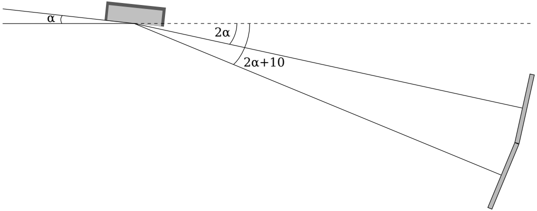
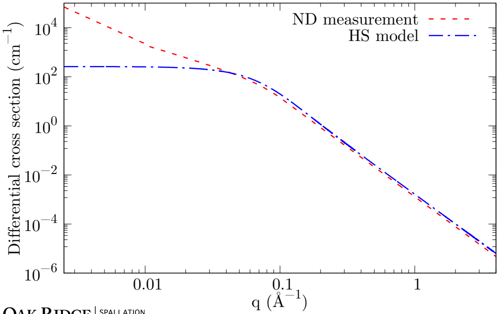
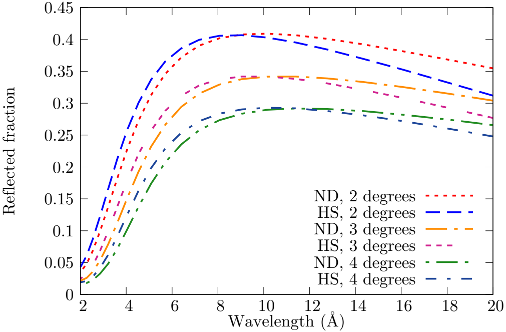
Sample	p_{PL}	s	r_{g}	m
1	2.2162	1.5718	12.932	3.9897
2	2.0343	1.5721	12.427	3.9793
3	1.9973	1.5698	12.359	3.9769
4	2.3147	1.6254	12.783	3.9997
5	1.8607	1.5698	12.004	3.9815

Reflectometer simulation

- Quasi-specular reflection has been observed for nanodiamonds (Cubitt et al., 2010).
- Analytical model and the nanodiamond data used.

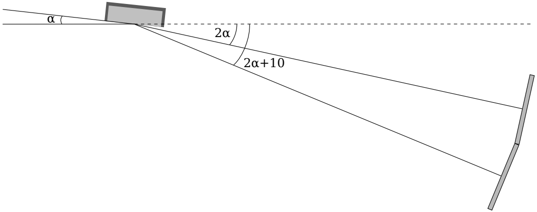


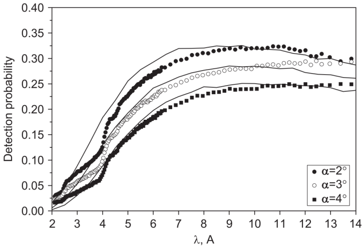


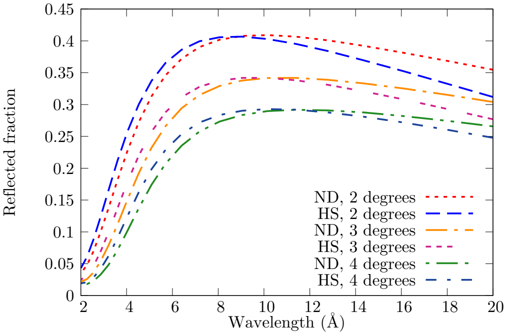


Reflectometer simulation

- Simulation results show reasonable agreement with Cubitt measurement.
- Analytical model shows less long-wavelength reflection, in agreement with Cubitt.







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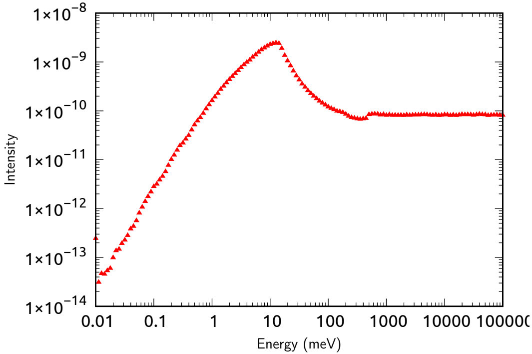
Initial moderator studies

Tube moderator

- Vessel is 0.4 cm thick aluminum.
- Vacuum jacket between vessel and water pre-moderator is 0.5 cm.



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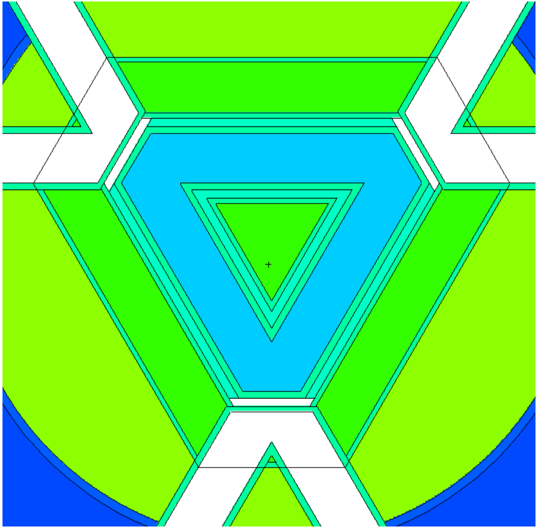
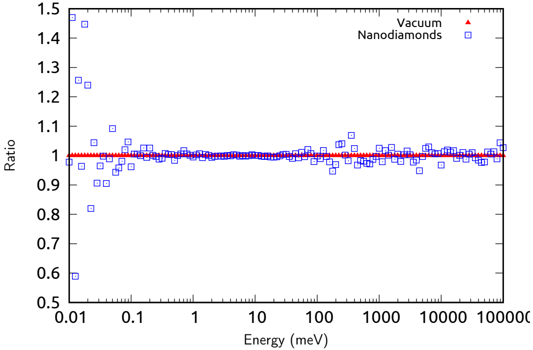


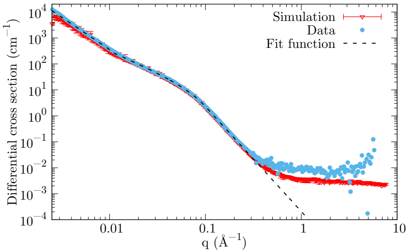
Tube moderator

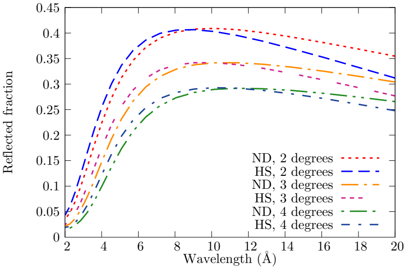
- Simulation assumes that the entire vacuum jacket contains nanodiamonds with density $\approx 0.4\ \mathrm{g\ cm^{-3}}$.



Conclusions

- Demonstrated a powerful new tool for simulating neutron transport with nanoscale materials.
- Excellent agreement between input $\frac{d\Sigma}{d\Omega}(q)$ and simulated SANS profiles.
- Qualitative agreement with Cubitt reflectometer measurements.
- Initial studies of one reflector configuration show negligible effect on spectrum, but still need to verify the SANS code works with variance reduction techniques.





Thank you!