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A sensitivity study using maximum entropy to interpret SANS data from the Ringhals Unit 3 NPP

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Abstract

SANS experiments were performed on a high Ni weld surveillance sample from the Ringhals NPP and the Maximum Entropy method was applied to determine the most probable size distribution of irradiation-induced scattering features. The results were shown to be consistent with atom probe observations. The sensitivity of the data analyses with respect to constraints such as the limited experimentally available Q range was explored. The calculated volume fraction and the mean volume-weighted diameter of the precipitates were found to be relatively insensitive to Q_{max} (the maximum scattering vector) greater than ~0.40 Å⁻¹. However, use of a lower Q_{max} results in a shift of the size distribution to larger diameters and a reduced particle number density. Simulations demonstrated that the experimentally observed decrease in the A-ratio at higher Q values is consistent with the presence of vacancies or higher Mn contents in smaller features. Importantly, features which are experimentally unresolvable do not add to the apparent volume fraction of the features which are resolved.

Keywords: Reactor pressure vessel steels; Small angle neutron scattering; Maximum entropy

1 Introduction

Ringhals Units 3 and 4 LWR have weld metal in the beltline region of the reactor pressure vessel (RPV) with high Ni and Mn. Charpy V-notched impact samples from the surveillance capsules were tested [1] and the ΔT_{NDT} values for fluences of 3.6 × 10¹⁹ncm⁻² and above were significantly greater than the Reg. Guide 1.99 revision 2 predicted values. It was predicted that the additional radiation damage observed was associated with the higher Ni contents of these welds than those materials used to develop Reg. Guide 1.99 and this has subsequently been substantiated by Atom Probe Tomography (APT) which demonstrated that the higher Ni levels led to the formation of a higher number density of irradiation-induced solute clusters [2,3]. These clusters impede dislocation movement leading to hardening and potentially embrittlement [4-6]. Odette recognised the potential for rapid precipitation of Mn and Ni in the early 1990s [7] which could potentially limit the lifetime of RPVs (particularly those with high Ni contents). Subsequently several major international research programmes were initiated to determine whether or not such effects need to be explicitly included in safety assessments of RPVs [8-11]. The majority of studies used a range of microstructural techniques. Insight gained from APT has proved particularly valuable but interpretation is not trivial and it can be extremely difficult to compare results from different research groups [12]. Small Angle Neutron Scattering (SANS) has a number of advantages when compared with APT. In particular, scattering data can be obtained from

a significant volume (i.e. a few mm³) rather than <1 micron³ which, in principle, means that there can be greater confidence that the data are truly representative of the material.

Analysis of SANS data involves interpretation of the scattering intensity from elastically scattered neutrons. Absolute precipitate size distributions can be determined provided that the nature (composition and magnetic properties) and shape of the scattering features are known. The analysis must take into account the size distribution of the clusters. This can be achieved by making an assumption about the shape of the size distribution (e.g. Gaussian, Log-normal or Schulz) and then using a least squares refinement to obtain the best fit [13]. Alternatively an indirect Fourier transform can be used but this can result in un-physical negative size distributions [14,15]. These limitations can be avoided by adopting a Monte Carlo approach as developed by Martelli et al. [16] and applied by Wagner et al. [17]. Alternatively, the Maximum Entropy technique can be used to identify the most probable distribution of cluster sizes that is consistent with the observed scattering [18-20]. However, even when adopting the most appropriate analysis methodology, it is important to understand the inherent uncertainties. In this research, a combination of experimental SANS data obtained from a surveillance sample of Ringhals R3 NPP and modelling has been used to examine:-

- The effect of the *Q*-range over which SANS data are fitted using the maximum entropy program on the resulting particle size distribution.
- The effect of assuming the presence of two different types of cluster.
- The effect of “unresolvable” (typically assumed to be sub-10 Å diameter) features on the size distribution returned by maximum entropy fitting.

The results are compared with previously published APT data [3] on the same material condition.

2 Experimental details

2.1 SANS experiments

Small angle neutron scattering (SANS) measurements were made using the D11 instrument at the Institut Laue-Langevin (ILL) in Grenoble. The experiment focused on an irradiated weld sample from Ringhals unit 3 (R3U) and a corresponding unirradiated reference sample. The irradiation history and nominal compositions of the weld metals are given in Table 1.

Table 1 Nominal compositions of Ringhals 3 high Ni welds (at.%).

alt-text: Table 1

Weld	Post-Weld Stress-Relief Treatment	Irradiation Conditions (n/cm ²) E > 1 MeV	C	Ni	Mn	Cr	Si	Cu	Mo	Al	P	S	N
Unit 3	575 °C for 26 h; 630 °C for 30 h; furnace cooled	5.6 × 10 ^{19a} (T _{irr} = 284 °C)	0.24	1.50	1.48	0.08	0.42	0.07	0.29	0.06	0.02	0.01	0.05

^a The highest dose in the capsule is 6.26 × 10¹⁹ncm^{−2}, but the sample used in this work was extracted from the centre layer which had an estimated fluence of 5.6 × 10¹⁹ncm^{−2}.

The differential scattering cross section was measured in zero field, and with the scattering vector, Q, both parallel and perpendicular to an applied magnetic field of >1 T for both the irradiated and unirradiated sample. The scattering vector $Q = \frac{4\pi}{\lambda} \sin\left(\frac{\varphi}{2}\right)$ where φ is the scattering angle and λ is the neutron wavelength. The data were normalised to determine the absolute differential scattering cross-sections. Subtraction then yielded the irradiation-induced component of the differential scattering cross section, $\left(\frac{d\sigma}{d\Omega}\right)_S$.

The macroscopic coherent neutron scattering cross-section (cm^{−1} steradian^{−1}) for a dilute concentration of a distribution of spherical precipitates with varying diameters is given by:

$$\left(\frac{d\Sigma}{d\Omega}\right)_S = N_S \left(\frac{d\sigma}{d\Omega}\right)_S = [N + \gamma M] \int C(D) \cdot V(D) \cdot |F_D(Q)|^2 dD$$

where $(d\sigma/d\Omega)_S$ is in units of barns/steradian/Fe atom and N_S is the number of sample (Fe) atoms per unit volume. N, the ‘nuclear contrast factor’, reflects the difference in mean nuclear scattering length density of the precipitate and matrix. Similarly, M is the ‘magnetic contrast factor’, which reflects the difference in mean magnetic scattering density of the precipitate and matrix. The scattering length densities are determined from published tables [21]. [N + γ M] is the overall ‘contrast factor’ for the precipitates where: (Please see attached file for formatting this section. The carriage return needs removing, the next 3 lines indenting, the equation involving C(D) aligning with the text and the equation involving Vf aligning with the text.)

γ is the contribution from the magnetic contrast:

- $\gamma = 2/3$ for scattering in zero field (assuming the magnetic moments are randomly orientated),

- $\gamma = 0$ for scattering measured with Q parallel to the magnetic field,

- $\gamma = 1$ for scattering measured with Q perpendicular to the magnetic field.

$\mathcal{C}(D)\delta D$ is the volume-weighted particle size number distribution, of spheres with diameter between D and $D+\delta D$. Hence, if $V(D)$ is the volume of spheres of size D , $V(D) = \pi D^3/6$, and: (Please remove the carriage return)

$$C(D) = V(D) \cdot N(D)$$

where $N(D)\delta D$ is the particle size number distribution, of spheres with diameter between D and $D+\delta D$.

$|F_D(Q)|$ is the form factor for spherical precipitates of diameter D , which is given by:

$$|F_D(Q)| = \left[\frac{24 \left[\sin(QD/2) - (QD/2) \cos(QD/2) \right]}{Q^3 D^3} \right]$$

The total volume fraction of precipitates in the sample is given by: (Please remove the carriage return and add a full stop after the equation for V_f)

$$V_f = \int C(D) dD$$

The A-ratio, defined as $\frac{(d\sigma/d\Omega)_{\perp}}{(d\sigma/d\Omega)_{\parallel}} = \frac{N+M}{N}$ provides information on the chemical composition of the scattering features.

2.2 Simulated data

Simulated SANS data may be generated by calculating scattering cross-sections as a function of Q for a given size distribution of particles of known contrast by numerically integrating the expression

$$[N + \gamma M] \int C(D) \cdot V(D) \cdot |F_D(Q)|^2 dD$$

Numerical integrations were performed, using the assumption that the nuclear and magnetic contrast originate from the same scatterers:

- For each value of Q , the form factor $|F_D(Q)|$ was calculated for discrete values of particle diameter at 2.5 Å intervals.
- The scattering cross-section at each discrete particle diameter D was then calculated as $[N + \gamma M] \cdot V_{f(D)} \left(\pi D^3/6 \right) \cdot |F_D(Q)|^2$, where $V_{f(D)}$ is the absolute volume fraction of particles in the size bin centred at D .
- The total scattering cross-section at each value of Q was then determined by summing the cross-sections across all particle diameters.

This enabled simulated data to be produced over Q ranges which extend beyond experimental measurements. Maximum entropy fitting of the simulated SANS data then allows the fitted size distribution to be compared with the original modelled distribution. Since experimental data extend over a higher Q -range for Zero Field (ZF) measurements than for field-on measurement with Q perpendicular or parallel to the applied magnetic field direction, data simulations have generally been made for ZF ($\gamma = 2/3$) conditions.

2.3 Maximum entropy analyses

Assuming that the particles are approximately spherical (as suggested by APT) and that their number density is sufficiently small/low that multiple scattering effects are negligible, the volume-weighted size distribution, $\mathcal{C}(D)$ versus D , is determined by fitting the theoretical expression for the coherent cross-section to the experimental data using the maximum entropy algorithm. The maximum entropy program is generally used to fit subtracted (irradiated sample - unirradiated control sample) data files corresponding to the irradiation-induced coherent scattering component.

During the initial stage of data fitting using the maximum entropy procedure, a fixed ‘nominal’ contrast factor of $1 \times 10^{28} \text{ m}^{-4}$ is generally used to fit the SANS data. This gives rise to a ‘relative’ volume fraction of particles

which is related to the absolute volume fraction and the true contrast factor by:

Rel. $V_f \times$ (nominal contrast factor) = Absolute $V_f \times$ (true contrast factor)

Since the simulated data covers a wider Q range than is obtainable experimentally, it is possible to study the effect of varying the Q-range on the resulting precipitate size distribution returned by the maximum entropy fitting program. Two approaches were adopted.

- by curtailing experimental data
- by using simulated data to extend the Q-range beyond that for which experimental measurements have been made

3 Results

3.1 Analysis of SANS data

The irradiation-induced (sample minus unirradiated reference) macroscopic cross-section data for R3U (individual points) and the maximum entropy fits (lines) are shown in Fig. 1a. As expected, the maximum entropy fits closely follow the experimental data. The corresponding volume-weighted size distributions derived from the maximum entropy fits using a nominal contrast factor, and the resulting nominal particle number density distributions, are given in Fig. 1b.

Fig. 1 (a) Maximum entropy fits to the macroscopic irradiation-induced scattering intensity data for sample R3U and (b) Relative volume-weighted size distributions, C(D), of irradiation-induced features in sample R3U (based on Q perpendicular to H data).

alt-text: Fig. 1

The results, Table 2, show good consistency between the zero field and in-field data. For each, the mean volume weighted diameter $\overline{d}_v$ was ~ 37 Å and the simple averaged (number density weighted) mean diameter was ~ 22 Å. The volume weighted results are consistent with previously reported APT data [3] on this material condition (35 ± 1 Å).

Table 2 Relative volume fractions, mean volume and number density weighted diameters, and mean A-ratios of irradiation-induced features and previously reported APT data.

alt-text: Table 2

Sample	SANS Data										APT data [3]	
	Zero Field			Q perp H			Q parallel H			A-ratio		
	V_f^{Rel}	$\overline{d}_v$ (Å)	$\overline{d}_n$ (Å)	V_f^{Rel}	$\overline{d}_v$ (Å)	$\overline{d}_n$ (Å)	V_f^{Rel}	$\overline{d}_v$ (Å)	$\overline{d}_n$ (Å)		$\overline{d}_v$ (Å)	V_f^{Abs}
R3U	0.249	37	22	0.297	36	25	0.143	37	22	2.1	35 ± 1	0.014

The composition, density (vacancy content) and magnetic properties of the scattering features must be known before the relative volume fraction data can be converted to absolute volume fraction data. Despite APT providing information on the ratio of solute elements, and the A-ratio providing a further constraint, assumptions still need to be made regarding the vacancy and magnetic properties [22] and solutions found are not unique. For instance, using the mean cluster composition observed by APT (58Fe, 22Ni, 14Mn-5Si-1Cu at.% [3]) coupled with an assumed magnetic moment of 0.7 for Fe in the scattering features and a small contribution from vacancies results in $V_f^{Abs} = 0.015$. Although this is consistent with the APT data ($V_f^{Abs} = 0.014$), it does not prove that the assumptions are correct. In particular, there is increasing evidence that in APT data, a significant fraction of Fe results from trajectory aberrations associated with the local magnification effect [22,23] and the uncertainties on the volume fractions from atom probe data are high because the precise methodology used to identify clusters will impact the results [24(Please change to

reference 12. Reference 12 and 24 are the same and so reference 24 needs removing.]. If it is assumed that the scattering features do not contain any Fe, the calculated $V_f^{Abs} \approx 0.005$ (from SANS) and ≈ 0.006 (from APT). Clearly there are multiple assumptions regarding the composition of the scattering features that appear consistent with both the SANS and APT data and therefore this paper focuses on analysing the data in a self-consistent manner to give relative volume fractions.

4 Discussion

4.1 Effect of the Q-range

Zero field data for the irradiated Ringhals sample R3U were obtained over a usable Q -range of $0.046\text{--}0.434\text{ \AA}^{-1}$. The irradiation-induced component of the coherent scattering data for R3U was obtained by subtracting measured data for the corresponding unirradiated reference sample. The effect of curtailing the maximum fitted Q value on the resulting relative volume fraction and number density of irradiation-induced features for R3U is shown in Fig. 2 and is summarised in Table 3.

Fig. 2 Effect of curtailing $Q_{\max}$ on relative volume-weighted size distribution, $C(D)$ of irradiation-induced features in Ringhals sample R3U.

alt-text: Fig. 2

Table 3 Effect of curtailing $Q_{\max}$ on relative volume fraction and number density of irradiation-induced features in Ringhals sample R3U.

alt-text: Table 3

$Q_{\max}$ ($\text{\AA}^{-1}$)	Rel. Vf (ZF contrast)	$\bar{D}_V$ ($\text{\AA}$)	Rel. N_d^a (10^{24} m^{-3})	$\bar{D}_N$ ($\text{\AA}$)
0.434	0.2488	36.7	21.9	22.2
0.397	0.2480	36.8	20.5	23.1
0.359	0.2465	37.0	18.2	24.6
0.322	0.2447	37.2	16.3	26.1
0.284	0.2427	37.4	14.5	27.7

^a Rel. N_d is the total number density summed over each size bin and is based on the nominal SANS contrast factor of $1 \times 10^{-28}\text{ m}^{-4}$ used in the maximum entropy fitting program.

The results indicate that the overall volume fraction and volume-weighted mean diameter ($\bar{D}_V$) of irradiation-induced features for R3U are relatively insensitive to the fitted Q -range (although there is a consistent trend of slightly decreasing relative volume fraction and increasing $\bar{D}_V$ with decreasing $Q_{\max}$). However it is clear from Fig. 2 that the positions of the peaks in the distributions are shifted to larger sizes as $Q_{\max}$ is reduced, resulting in a significant reduction in the apparent feature number density (Rel. N_d - for which the nominal units are 10^{24} m^{-3}) and a corresponding increase in the number-weighted mean diameter ($\bar{D}_N$).

In order to extend the Q-range beyond that for which experimental measurements have been made, simulated data were produced using the contrast factors for the 1Cu-14Mn-22Ni-5Si-58Fe features as observed by APT [3] in this material condition. The simulated data were based on a bimodal Gaussian size distribution which was chosen to be similar to experimental data (with the two peaks corresponding to the experimentally observed distribution). The modelled size distribution is compared with the maximum entropy fit to the Zero Field data for R3U (for experimental $Q_{\text{max}} = 0.434 \text{ \AA}^{-1}$) in Fig. 3a, and the simulated scattering data are compared with the experimental data for zero field and for Q both perpendicular (N + M contrast) and parallel (N contrast) to the applied magnetic field H in Fig. 3b. The simulated zero field scattering data were extended to $0.55 \text{ \AA}^{-1}$ to further examine the effect of Q-range on the real space size distribution returned by the maximum entropy fitting procedure.

Fig. 3 Comparison of (a) relative volume-weighted size distribution, $C(D)$ and (b) simulated macroscopic scattering intensity data for sample R3U and modelled distribution.

alt-text: Fig. 3

The effect of Q_{max} on the relative volume-weighted size distribution $C(D)$, and the relative precipitate number density N_d , determined by maximum entropy fitting of the simulated data set, is shown in Table 4 and Fig. 4. The results indicate that the volume fraction and volume-weighted diameter of the precipitates are reasonably well determined irrespective of Q_{max} , though the apparent size distribution is shifted to larger diameters (resulting in a reduction in number density and an increase in the number-weighted mean diameter) as Q_{max} is reduced below $\sim 0.40 \text{ \AA}^{-1}$.

Table 4 Effect of curtailing Q_{max} on relative volume fraction, mean diameter and number density of features in simulated zero field data set.

alt-text: Table 4

$Q_{\text{max}} \text{ (\AA}^{-1}\text{)}$	Rel. Vf (ZF contrast)	$\bar{D}_V \text{ (\AA)}$	Rel. $N_d^a \text{ (10}^{24} \text{ m}^{-3}\text{)}$	$\bar{D}_N \text{ (\AA)}$
0.55	0.2738	34.5	31.2	21.2
0.50	0.2738	34.5	31.1	21.3
0.45	0.2740	34.5	31.3	21.2
0.40	0.2738	34.5	30.8	21.3
0.35	0.2719	34.8	26.7	22.9
0.30	0.2671	35.3	20.9	25.7
Modelled Distribution*	0.2756	34.5	31.8	21.0

^a Rel. N_d is the total number density summed over each size bin and is based on the nominal SANS contrast factor of $1 \times 10^{-28} \text{ m}^{-4}$ used in the maximum entropy fitting program.

Fig. 4 Effect of curtailing Q_{max} on the relative volume-weighted size distribution, $C(D)$, and number density, N_{ϕ} , of features in simulated zero field data set.

alt-text: Fig. 4

The experimental data for sample R3U and the associated simulation represent a case of a bimodal size distribution where the smaller features are in a size range of ~10–20 Å diameter, which would generally be expected to be resolved by SANS measurements. An implicit assumption has been made that all of the features in the distribution exhibit the same SANS contrast; this may not necessarily be true and the potential effect of compositional variation on the apparent size distribution is considered next.

4.2 Effect of assuming the presence of two different types of cluster

It is frequently assumed that the irradiation-induced features are all of one type, i.e. exhibit the same contrast factor. The composition of irradiation-induced features is often based on Atom Probe measurements, with the density (which effectively is dependent on the vacancy content of the features) and the magnetic moment of Fe in the features then being adjusted to give an A -ratio which is reasonably consistent with the experimental data. The assumption that all the features are of one type implies that the A -ratio (given by $[(N + M)/N]$) should be independent of the scattering vector Q , though in practice some variation in the A -ratio is often observed experimentally. The two component model was adapted so that the composition of each of the binomial distributions could be assigned independently. As shown in Table 5 and comparing Fig. 5 with Fig. 4, the scattering data can be interpreted as resulting from a lower number density (or volume fraction) of higher contrast features or a higher number density of lower contrast features.

Table 5 SANS contrast factors of different types of Cu-Mn-Ni-Si-Fe precipitates.

alt-text: Table 5

Ppt type	Solute Content (at.%)					Magnetic moment of Fe in feature	Density (g cm ⁻³)	A -ratio	Zero Field Contrast, (N+2 M/3) (1028 m-4)
	Cu	Mn	Ni	Si	Fe				
A (main peak at ~35–40 Å)	1	14	22	5	58	0.7	6.0	2.17	15.3
D (minor peak at ~15 Å)	5	26	22	5	42	0.7	5.0	1.65	31.4

Fig. 5 Volume-weighted size distributions, $C(D)$ of bimodal model data which exhibit the same total coherent, zero field, neutron scattering cross-sections as shown in Fig. 1a.

alt-text: Fig. 5

The dependencies of the calculated A -ratios with Q are compared with experimental data for sample R3U (determined from the ratio of the scattering cross-sections for Q perpendicular and parallel to the applied magnetic field) in Fig. 6. It is clear that the presence of different types of feature in the modelled size distribution gives rise to variations in the A -ratio. In practice it would be difficult to simulate the experimental variation in the A -ratio precisely, because such variation could arise not only from differences in composition (and/or vacancy content and magnetic properties) between features of different size, but also from compositional gradients within the features themselves (e.g., if the core of the feature differs from the outer shell). However, the results shown in Fig. 6 suggest that the experimentally observed decrease in the A -ratio in sample R3U at the higher Q values could feasibly be due to a higher vacancy and Mn content in the smaller features detected by SANS.

Fig. 6 Comparison of calculated A -ratios for modelled bimodal size distributions containing different types of feature with experimentally determined A -ratio for Ringhals sample R3U.

alt-text: Fig. 6

4.3 Effect of “unresolvable” (sub-10 Å diameter) features on the size distribution returned by maximum entropy fitting

To examine the potential effect of unresolved (below ~ 10 Å diameter) features, artificial data sets were produced to simulate zero field results. Simulations were carried out for $Q_{\max}$ of 0.55 Å^{-1} for a bimodal distribution with a small peak, centred at 7 Å (Rel. $V_f = 0.0257$), overlapping a larger peak centred at 14 Å (Rel. $V_f = 0.2573$) (Fig. 7). In this case the smaller peak was not resolved by the maximum entropy fit.

Fig. 7 Comparison of relative volume-weighted size distributions, $C(D)$, of features derived from maximum entropy fitting with modelled bimodal distributions.

alt-text: Fig. 7

The data were re-examined using different values of $Q_{\max}$ for maximum entropy fitting, including reduced values of 0.35 and $0.45 \text{ \AA}^{-1}$ and extended values up to $1.10 \text{ \AA}^{-1}$. The results, shown in Fig. 8 and Table 6, demonstrate that the true modelled size distribution including the smaller diameter particles is only returned accurately by maximum entropy fitting when the data extend to very high Q values of $0.90\text{--}1.10 \text{ \AA}^{-1}$. (This is consistent with the nominal resolution limit for SANS of $D = 2\pi/Q_{\max}$, which derives from Bragg's law, $\lambda = 2D \sin\theta$, and the definition of the scattering vector, $Q = 4\pi \sin\theta/\lambda$ [25].) However, it is evident that the contribution from the smaller particles to the overall fitted volume fraction and number density increases with increasing $Q_{\max}$. At the lower $Q_{\max}$ values more typically attained in experimental measurements it is evident that sub-10 Å diameter features are not resolved.

Fig. 8 Effect of $Q_{\max}$ on the relative volume-weighted size distribution, $C(D)$ of features in simulated zero field data.

alt-text: Fig. 8

Table 6 Effect of $Q_{\max}$ on relative volume fraction, mean diameter and number density of features determined from maximum entropy fitting of simulated zero field data set with peaks in the size distribution at $D_V = 7$ & $14 \text{ \AA}$ (with corresponding Rel. $V_f = 0.0257$ & 0.2573).

alt-text: Table 6

$Q_{\max} (\text{\AA}^{-1})$	Rel. V_f (ZF contrast)	$\bar{D}_V (\text{\AA})$	Rel. $N_d^a (10^{24} \text{ m}^{-3})$	$\bar{D}_N (\text{\AA})$	Smaller features resolved?
0.35	0.2539	14.2	180	13.7	No
0.45	0.2654	13.9	211	13.1	No

0.55	0.2692	13.8	232	12.5	No
0.70	0.2774	13.5	283	11.0	Partially
0.90	0.2809	13.4	335	10.7	Yes
1.10	0.2816	13.4	367	10.1	Yes
Modelled Distribution ^a	0.2830	13.4	358	10.3	

^a Rel. N_d is the total number density summed over each size bin and is based on the nominal SANS contrast factor of $1 \times 10^{-28} \text{ m}^{-4}$ which is used in the maximum entropy fitting program.

It is of note that the volume-weighted mean diameter and relative volume fraction of features returned by maximum entropy fitting with Q_{max} restricted to 0.35 and 0.45 Å⁻¹ are close to the values expected for the second peak in the modelled distribution on its own ($\bar{D}_V = 14 \text{ Å}$, and relative $V_f = 0.257$), indicating that the fitting procedure is not unduly affected by the presence of smaller unresolved features – an important point for the interpretation of experimental data.

5 Conclusions

SANS data, analysed using a Maximum Entropy method to determine the most probable size distribution of scattering features, from a high Ni surveillance weld from Ringhals Unit 3 are consistent with previously reported APT observations. Simulations, encompassing a wider Q range than obtainable experimentally, have been used to explore the limitations of the SANS measurements. Scattering cross-sections were generated for a given size distribution of particles of known (or assumed) contrast. Maximum entropy fitting of this simulated SANS data then allowed the fitted size distributions to be compared with the original modelled distributions. Three particular aspects relating to the interpretation of experimental SANS measurements have been examined:

- The effect of varying the Q -range on the resulting precipitate size distribution has been examined. The results obtained from fitting the simulated and experimental data sets were found to show similar trends, with the overall volume fraction and the mean volume-weighted diameter of the precipitates being relatively insensitive to Q_{max} , but with the size distribution tending to shift to larger diameters (resulting in a reduction in particle number density and an increase in the number-weighted mean diameter) as the fitted Q_{max} is reduced below $\sim 0.40 \text{ Å}^{-1}$.
- The consequences of having different types of feature in a bimodal size distribution have been examined by changing the composition (and, hence, the SANS contrast factor) of the smaller features while keeping the composition of the larger features fixed. Any change in composition and/or vacancy content that increases the SANS contrast factor of the smaller features requires a smaller true volume fraction to produce the same overall relative volume fraction and scattering curve. Comparison of data for the irradiated sample with simulated data sets suggests that the experimentally observed decrease in the A -ratio at higher Q values could be due to a high vacancy or Mn content in the smaller features detected by SANS.
- The effect of sub-10 Å diameter features on the resulting size distributions returned by maximum entropy fitting has been examined by fitting simulated SANS data for a bimodal distribution with peaks centred at 7 and 14 Å. The results indicate that much higher values of Q_{max} , of the order of $\sim 1 \text{ Å}^{-1}$, than are obtainable experimentally are required to resolve sub-10 Å sized features. Importantly, detailed examination of the results indicates that features which are experimentally unresolvable do not add to the apparent volume fraction of the features which are resolved.

Data availability

The raw/processed data required to reproduce these findings cannot be shared at this time as the data also forms part of an ongoing study.

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