Experimental report

17/12/2021

Proposal:	**1-01-184**	**Council:**	10/2020

Title: Characterising Irradiation InducedMn-Ni-Si Precipitates in Low Cu Steels for Generation III Reactor Pressure Vessel Steels

Research area: Materials

This proposal is a new proposal

Main proposer: **Alexandra DICKINSON LOMAS**

Experimental team: Robert CUBITT

Local contacts: Nina-Juliane STEINKE
Robert CUBITT

Samples: RPV steel (see proposal text)
Fe-1.35wt%Mn-0.75wt%Ni
Fe-1.35wt%Mn-0.20wt%Si
Fe-0.75wt%Ni -0.20wt%Si
Fe-1.35wt%Mn-0.75wt%Ni -0.20wt%Si

Instrument	Requested days	Allocated days	From	To
D33	3	3	25/06/2021	28/06/2021
D22	3	0		
D11	3	0		

Abstract:

This experiment will provide insight into the mechanisms and microstructural effects of Mn, Ni and Si in nuclear reactor pressure vessel steels. Analysis of irradiation induced precipitate and cluster formation using SANS will enable the deduction of the mechanisms of microstructure evolution and their contribution to radiation hardening and embrittlement. Scattering data will provide information on precipitate size and distribution information. Together with laboratory characterisation (TEM, hardness (nanoindentation)) this will contribute to existing gaps in steel property databases for reactor pressure vessel steels. This can directly applied to predicting structural integrity of reactor pressure vessels and extending their operating lifetime.

Characterising Irradiation Induced Mn-Ni-Si Precipitates in Low Cu Steels for Generation III Reactor Pressure Vessel Steels- Preliminary Report 17/12/2021

Background

An understanding of the radiation effects on mechanical property evolution of materials used in Generation II, III fission and fusion reactors is essential to their safe operation, appropriate life extension decisions and design of future power plants. Over the design lifetime of an RPV, ~40 years (for generation II reactors) the damage accumulated due to neutron irradiation is generally of the order of 0.1dpa (displacements per atom).[1] Displacements of atoms in the lattice by neutron bombardment increase the rate of diffusion in the steel enabling the formation of various microstructural defects such as vacancy clusters, dislocation loops, solute clusters and the segregation of solute atoms to grain boundaries and dislocations.[2] These results in a reduction in fracture toughness and strength and an increase in hardening and embrittlement. [3] As the wt.% of copper has decreased in RPV steels the effect of Mn-Ni-Si rich precipitate evolution on mechanical properties has gained interest. Compared with copper rich precipitates, Mn-Ni-Si precipitates show smaller average sizes and narrower size distribution.[4] Previous work in the field disagrees on whether Mn-Ni-Si precipitates are stable thermodynamic phases. Some studies claim they form stable G and Γ_2 Mn-Ni-Si phases after high fluence irradiation.[5] Others claim the precipitates nucleate at low fluence, are metastable and grow at high fluences. [6]

Experimental Summary

10 samples, two of each composition, were measured using the D33 Small angle scattering diffractometer.

The compositions of the samples were: 4 model steels of varying Mn, Ni and Si content (Figure 3) and an A508-3 RPV steel (wt.%C-0.17, Cu-0.03, Ni-0.74, Mn-1.35, Si-0.19, Mo-0.48, P-0.005, S-0.002, Cr-0.09, V-0.007).

Sample	Mn	Ni	Si
1	1.35	0.75	-
2	1.35	-	0.20
3	-	0.75	0.20
4	1.35	0.75	0.20

Figure 1- Table showing the compositions in wt.% of the Fe-Mn-Ni-Si model alloys to be measured in this project.

One sample of each composition had been irradiated at 300˚C, 15µA, with 2.9 MeV protons to 0.1 dpa in the plateau region. The other was heat treated to simulate the thermal history of the irradiated sample.

Scattering was measured over a q range between 0.1 Å^{-1} and 0.001 Å^{-1} to obtain scattering intensities across a range of scattering particle sizes across the nm and um scales.

Cadmium, air scattering and transmission measurements were also taken. A horizontal saturating magnetic field (0.75 T) was applied perpendicular to the neutron beam to separate magnetic and nuclear scattering components of the sample.

Data was reduced using GRASansP software from the ILL[7] and model fitting has been carried out using NCNR SANS Fitting Macros, in IgorPro. [8]

Preliminary Results and Findings

There was a clear increase in scattering intensity in irradiated alloys compared with unirradiated alloys in the q range around 0.02 Å^{-1} indicating an evolution of microstructure due to radiation damage. (Figure 3) Clear anisotropy of scattering was observed when a magnetic field was applied to proton irradiated versus unirradiated indicating a difference in magnetic behaviour of scatterers compared with the matrix. (Figure 2)

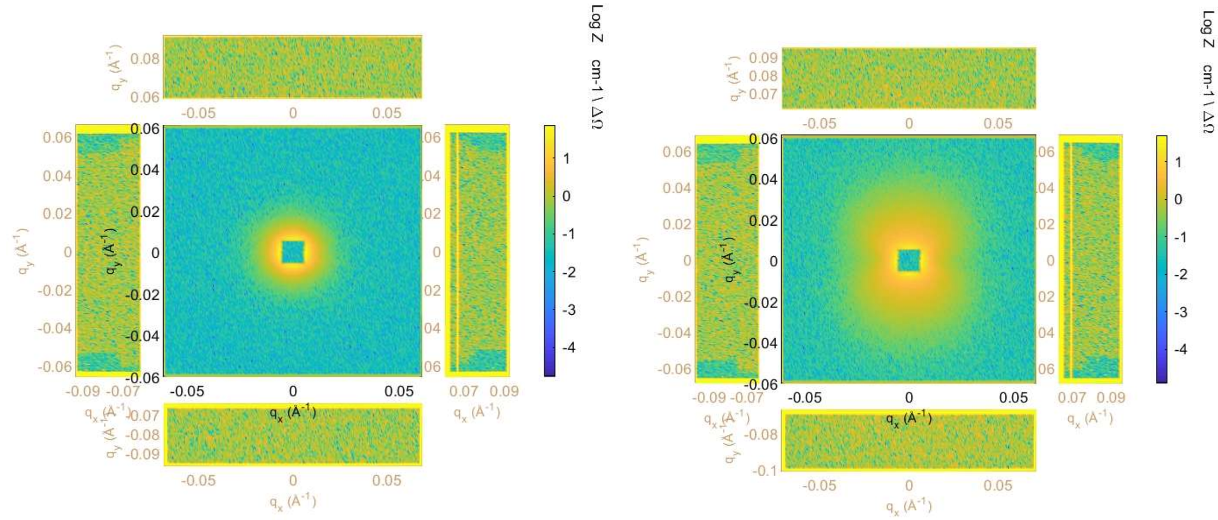

Figure 2: Detector intensity images showing isotropic scattering of the heat-treated sample (left hand side) and the anisotropy of scattering in the proton irradiated sample (right hand side).

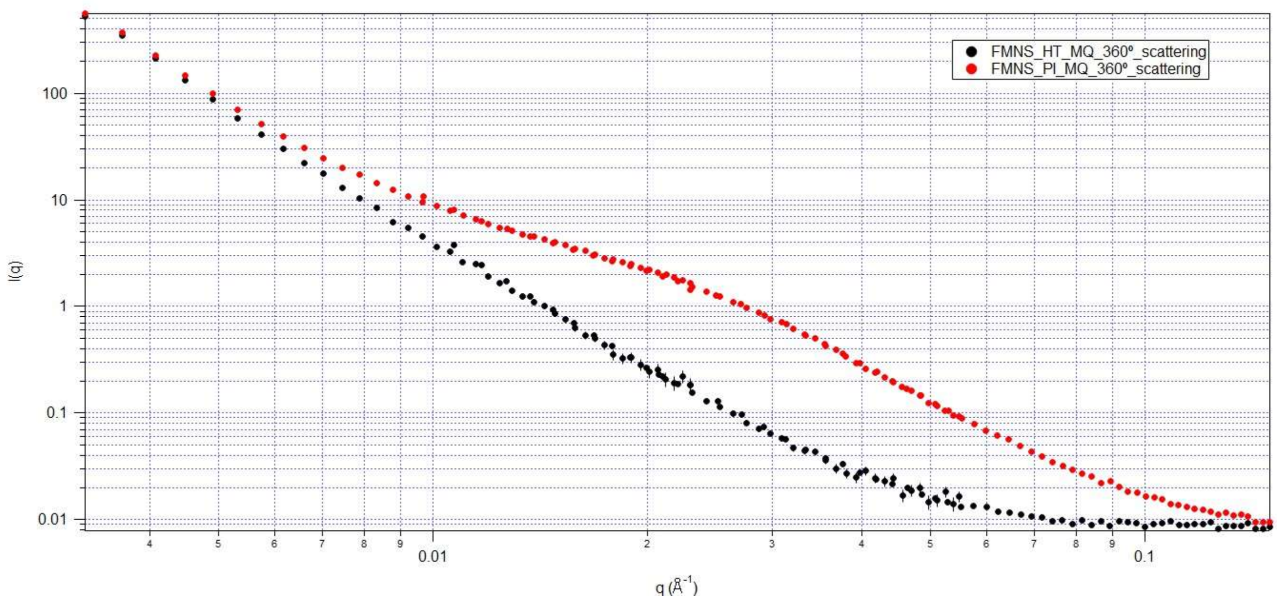

Figure 3: Total scattering intensity comparison of heat treated (black) sample data and proton irradiated data (red) for the FeMnNiSi alloy composition.

Sectors were taken vertically (total scattering) and horizontally (nuclear scattering) to extract the magnetic scattering component via a subtraction and then the heat treated data was subtracted from the proton irradiated data to provide a background subtraction accounting for thermal effects. (Figure 4)

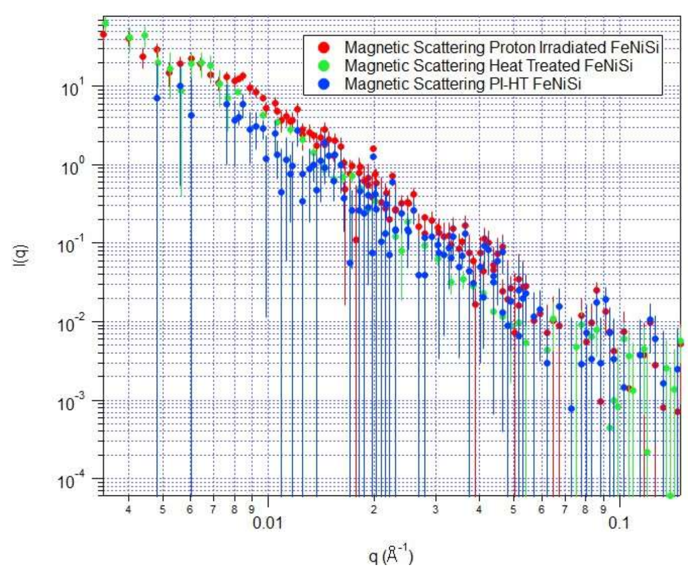

Figure 4: Plot showing the magnetic scattering vs q for proton irradiated, heat treated and proton-irradiated minus heat treated FeNiSi alloy.

Initially two size distributions have been assumed for the scattering solute clusters: a Log Normal Sphere dsitribution and a Gaussian sphere distribution. The fits for the FeMnSi alloy can be seen in Figure 5.

The volume fractions calculated for Mn containing alloys are around 5 times larger than those for the FeNiSi alloys suggesting that

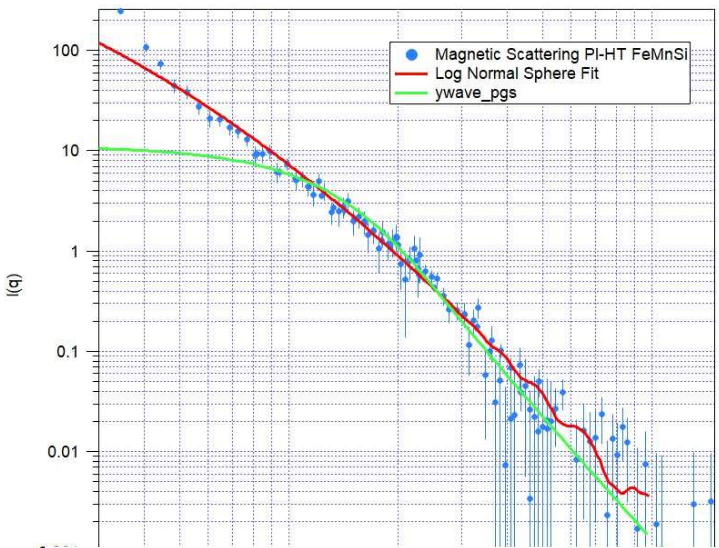

Figure 5: Plot of I versus q for the magnetic scattering of FMNS sample with a log normal (red) and gaussian sphere particle size distribution model (green) fitted to the data.

Mn diffusion dominates clustering. A wide variation in cluster size is calculated using different models but all values are around the 1-5nm scale. This will need confirmation in further studies using other techniques.

Scans across three of the irradiated samples showed an increase in scattering in the irradiated areas compared with the unirradiated areas which indicates that radiation damage is resulting in a change in microstrcuture along with the effects of elevated temperature.

Further work on the same materials using atom probe tomography in 2022 will provide compositional inforation on the clusters which can be used to better estimate their magnetic scattering contrasts and create improved fits to the SANS data.

This data will provide insight into the effects of long-term radiation damage and how microstructural evolution depends on the composition of solute elements in low alloy steels.

[1] J. C. van Duysen and G. Meric de Bellefon, "60th Anniversary of electricity production from light water reactors: Historical review of the contribution of materials science to the safety of the pressure vessel," *Journal of Nuclear Materials*, vol. 484. Elsevier B.V., pp. 209–227, Feb. 01, 2017. doi: 10.1016/j.jnucmat.2016.11.013.

[2] W. J. Phythian and C. A. English, "Microstructural evolution in reactor pressure vessel steels," *Journal of Nuclear Materials*, vol. 205, no. C, pp. 162–177, Oct. 1993, doi: 10.1016/0022-3115(93)90079-E.

[3] N. Almirall *et al.*, "Precipitation and hardening in irradiated low alloy steels with a wide range of Ni and Mn compositions," *Acta Materialia*, vol. 179, pp. 119–128, Oct. 2019, doi: 10.1016/j.actamat.2019.08.027.

[4] M. Hernández-Mayoral and D. Gómez-Briceño, "Transmission electron microscopy study on neutron irradiated pure iron and RPV model alloys," *Journal of Nuclear Materials*, vol. 399, no. 2–3, pp. 146–153, Apr. 2010, doi: 10.1016/j.jnucmat.2009.11.013.

[5] P. B. Wells *et al.*, "Evolution of manganese-nickel-silicon-dominated phases in highly irradiated reactor pressure vessel steels," *Acta Materialia*, vol. 80, pp. 205–219, 2014, doi: 10.1016/j.actamat.2014.07.040.

[6] R. Ngayam-Happy, C. S. Becquart, C. Domain, and L. Malerba, "Formation and evolution of MnNi clusters in neutron irradiated dilute Fe alloys modelled by a first principle-based AKMC method," *Journal of Nuclear Materials*, vol. 426, no. 1–3, pp. 198–207, Jul. 2012, doi: 10.1016/j.jnucmat.2012.03.033.

[7] "Grasp - ILL Neutrons for Society." https://www.ill.eu/users/support-labs-infrastructure/software-scientific-tools/grasp (accessed Jul. 05, 2021).

[8] "SANS and USANS Data Reduction and Analysis Software | NIST." https://www.nist.gov/ncnr/data-reduction-analysis/sans-software (accessed Jul. 05, 2021).