Neutron scattering techniques as a tool for non-destructive testing

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Abstract – Neutron scattering techniques, such as neutron diffraction, small angle neutron scattering and neutron reflectometry consist very powerful tools for the non-destructive study of materials. At the Greek Research Reactor, GRR-1, a number of neutron scattering methods are employed and developed. In the present contribution, a short description of the methods and facilities is given, as well as some demonstrative examples of their use in non-destructive materials testing.

Keywords: Neutron scattering, neutron diffraction, neutron reflectometry, small angle neutron scattering.

1 Introduction

Neutron scattering techniques constitute a powerful tool for non-destructive testing of materials. Neutron has a high penetration depth into matter and this allows the study of volume effects and bulky components. Furthermore, in-situ time-dependent experiments may be performed within complex sample environments such as furnaces, cryostats, pressure vessels or chemical reactor vessels.

Compared to the commonly used X-ray techniques, neutrons have the additional advantage of distinguishing among different metals and are sensitive to light elements. Another unique advantage of neutrons relies on the fact that the neutron is scattered differently by the various isotopes of an element. This gives the ability to vary the contrast of a defect, for example, by changing the isotopes in the defect or its surrounding matrix. The strong interaction of neutrons with hydrogen gives a unique ability to investigate through scattering techniques, non-destructively, the concentrations, locations and movements of hydrogen in solids, which is useful in the development of hydrogen storage alloys and fuell cells, and in studies of hydrogen embrittlement of structural metals.

Neutron diffraction technique is the most useful method for studies of structural changes of the crystal lattice particularly in the presence of heavy elements. The structure and volume fraction of minority phase can be measured at levels appreciably below that possible by X-ray diffraction. A rapidly growing field is the measurement of internal stresses in engineering components through the shifts in lattice spacing, the diagnosis of the modification of irradiated materials, etc [1]. Neutron diffraction is unique in being able to measure the full strain tensor from a specified volume within a bulk specimen.

Small Angle Neutron Scattering (SANS) and Ultra Small Angle Neutron Scattering (USANS) are actually scattering methods for detecting heterogeneities in the range from 1 to 2000 nm embedded within a matrix of different neutron scattering power. SANS can be used for predicting failure in nickel-base superalloy turbine blades, to study porosity in cements and other porous materials, to reveal the kinetics of precipitation in alloys or to study the debonding of fibers in carbon-carbon fiber composites.

Neutron Reflectometry (NREF) is a relatively new technique that has widespread applications as a powerful tool to analyze interfacial structure and composition of thin films, multilayers, surfaces, solid/liquid interfaces, membranes and other « two-dimensional » structures. Its application extends in the new emerging research areas in the fields of Material Science, Biology, Chemistry and Nanotechnology [2].

In this paper the neutron scattering facilities at the Greek Research Reactor GRR-1 installed at the National Centre for Scientific Research « Demokritos » are described in connection with some examples of non-destructive applications in Materials Science.

2 Neutron Scattering Facilities at GRR-1

The Greek Research Reactor GRR-1 is an open pool-type reactor of 5 MW thermal power, cooled and moderated by light water, and employing beryllium reflectors at two opposing sides of the core. Its neutron scattering facilities consist of a two-axis neutron diffractometer, which is in operation since 2001, a Time-of-Flight (TOF) Reflectometer which is under installation, a SANS and a USANS instrument which are under development.

2.1 Neutron Diffraction

Neutron diffraction refers to the phenomenon associated with the interference processes which occur when neutrons are scattered by the atoms within solids, liquids, and gases. The use of neutron diffraction requires high thermal-neutron fluxes which can be obtained only from nuclear reactors. These diffraction investigations are

possible because thermal neutrons have energies with equivalent wavelengths near 0.1 nm and are therefore ideally suited for interatomic interference studies.
Diffraction line positions and intensities give structural information on the material under investigation. Line widths give information on the size, strain and defect nature of the crystallites responsible for the diffraction.
A perfectly ordered crystalline material gives only Bragg scattering together with a small contribution from temperature dependent thermal diffuse scattering. Any disorder gives diffuse scattering between Bragg peaks. The magnitude of diffuse scattering may therefore be used as a test of the perfection of crystalline materials.
The neutron powder diffractometer NEDI, installed at GRR-1, was designed so as to enhance the quality of diffraction data which can be obtained from a reactor of modest thermal power. Its applications are mainly focused on the study of crystalline and magnetic structures, phase transitions induced, e.g., by temperature or pressure changes, residual stress measurements and kinetics of chemical reactions. A schematic layout of the instrument is given in figure 1. Table 1 summarizes the most important instrument parameters.

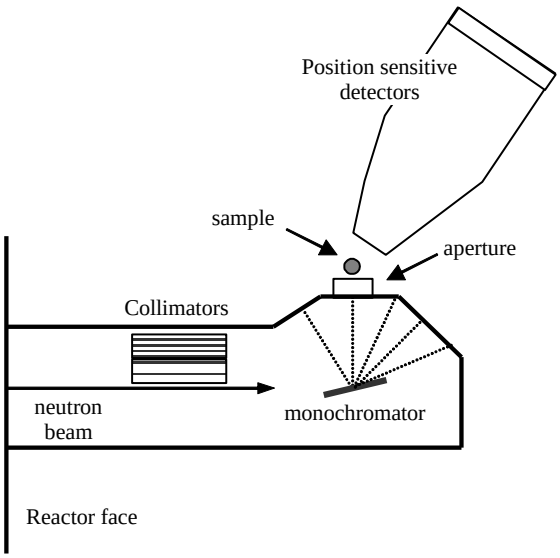

Figure 1 : Schematic Layout of the Neutron Diffractometer at GRR-1.

An in-pile main beam collimator of 1.87 m length is installed in the beam tube ending at a rotating drum which enables opening and closing of the beam. A sapphire filter of 75 mm thickness is used in the primary beam to reduce the fast neutron flux [3]. The neutron diffractometer radiation shielding was optimized by Monte Carlo neutron and photon transport code calculations (MCNP) [4]. Figure 1 shows the schematic layout of the instrument. Two alternative multi-crystal focusing monochromators are available: germanium (Ge/hkk) and pyrolytic graphite (PG/002), having mosaicities of 15′ and 30′, respectively. The Ge monochromator provides wavelengths in the range 0.41 – 2.95 Å. The pyrolytic graphite uses the 002 reflection and can provide wavelengths from 1.28 – 5.81 Å. It is used in combination with a graphite filter for the suppression of high order harmonics.
Two Soller collimators placed before the monochromator, made from mylar-foil covered with gadolinium oxide, offer a collimation of 15′ and 30′ with transmission of 93% and 96%, respectively.

Table 1. Characteristics of the 2-axis neutron diffractometer NEDI at the GRR-1 reactor.

Collimators (Soller)	Automatic exchange α_1=15', 30'
Monochromator	Automatic exchange of germanium (Ge/hkk) and pyrolytic graphite (PG/002) with vertical focusing on the sample
Graphite Filter	Elimination of 2nd order Bragg reflections
Take-off angles	$2\theta_M$=22°, 45°, 60°, 90° and 120°
Wavelength range	Ge:0.41 – 2.95 Å PG: 1.28– 5.81 Å
Angular range	5° <= 2θ <=120°
Neutron flux	5×10^8 n/cm²/s (at the monochromator)
Sample-detector distance	0.5 to 1.6 m
Sample environment	10 – 300 K (closed cycle refrigerator) 300 – 1800 K (furnace)

The collimator and monochromator units are housed within the main instrument shield, which is constructed as to allow easy access to both units. The main monochromator shield provides take-off angles at 22, 45, 60, 90 and 120°.
A sample stage sustained on air cushion pads allows easy movements to the different beam exit tubes. Different sample environments can be easily assembled allowing convenient sample mounting. The incident to the sample neutron beam is defined by an adjustable in height and width aperture. The incident beam may be focused on a minimum sample height of 2 cm giving an intensity gain of a factor of about three.
The detection system consists of seven linear position sensitive ^{3}He detectors placed horizontally one on top of the other. The whole detector assembly can be moved on rails in and out towards the sample position and the acceptance angle of the detectors system can vary from 18° to 56° in Bragg angle 2θ. This offers the flexibility to choose between high resolution or high counting rate needed for a dynamic experiment.
More details about the instrument design can be found in Ref. [5].
An example of the application of neutron diffraction technique refers to the structural characterization of irradiated SiC_f/SiC composite materials with fusion applications [6]. Composite specimens of Nicalon-S fibres in a SiC matrix, types N3 and N4, were subjected to neutron irradiation at GRR-1. Different dose levels were studied with irradiation times ranging from 200 to 1600 hours at a neutron flux of 7.5×10^{13} n/cm²/s and a temperature of 40ºC. The maximum fast neutron fluence was up to 4.3×10^{24} n/m² which corresponds to about

0.43 dpa (displacement per atom).
The irradiated samples were measured by neutron diffraction at the NEDI neutron diffractometer of GRR-1. Structural changes are observed to occur mainly after 800 hours of irradiation. These show lattice contraction and indicate the existence of defect accumulation. There is no clear indication of amorphization caused by fast neutron displacements up to 0.43 dpa.

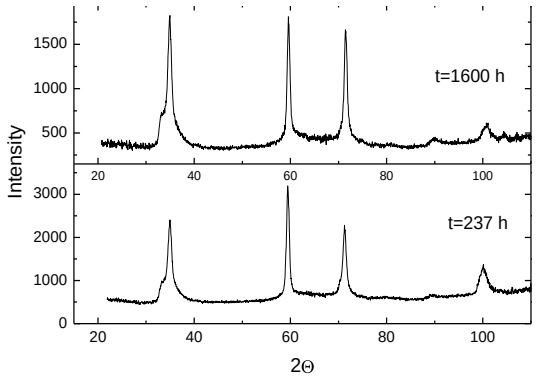

Figure 2 : Neutron diffraction spectra of irradiated SiC_f/SiC-N3 for 237 and 1600 hours.

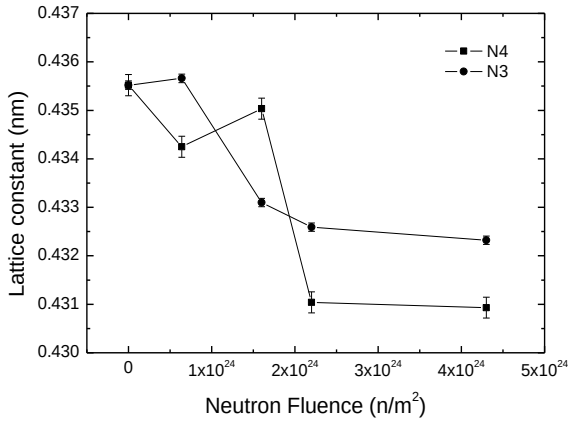

Figure 3 : The lattice constant of irradiated N3 and N4 type of SiC_f/SiC versus the neutron fluence.

In Figure 2 it is shown the neutron diffraction spectra from SiC_f/SiC-N3 composites for irradiation times of 237 and 1600 hours. Lattice contraction takes place for both N3 and N4 types after irradiation at a neutron fluence of 1.6×10^{24} n/cm^2 (about 0.16 dpa) and 0.6×10^{24} n/cm^2 for N4 and N3 correspondingly. The lattice contraction is more intense (of about 1%) for N4 type than for N3 type. In Figure 3 it is shown the lattice constant for both types N3 and N4 of SiC_f/SiC composites versus the fast neutron fluence.

2.2 Neutron Reflectometry

Neutron reflectometry is an established technique for the study of thin films, multilayers, and surfaces. In its simplest form, neutron reflectometry consists of measuring the specular reflection of neutrons from the sample surface at grazing angles of incidence. The reflected neutron intensity is recorded as a function of the scattering wave-vector, $Q = 4\pi \sin\theta / \lambda$, where θ is the angle of incidence and λ the neutron wavelength.

Up to an angle of incidence θ_c, called the critical angle, neutrons of a given wavelength undergo total external reflection. Beyond θ_c the beam penetrates the stratification and gets reflected at the interfaces. The information that can be obtained through the use of this technique are the thickness, density and roughness of each layer of the thin film sample, as well as phenomena like inter-diffusion can be revealed. If one measures reflectivity outside the specular condition, i.e. angle of reflection different than that of incidence, then the reflected beam provides information on the in-plane height-height correlation function at various interfaces in the thin film and on lateral structures.

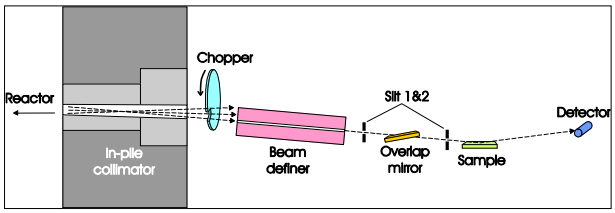

Figure 4 : Schematic layout of the neutron reflectometer.

The reflectometer at GRR-1 has been designed so as to satisfy different experimental requirements such a) the ability to measure both solid and liquid samples, b) utilization of the short neutron wavelengths, c) availability of different wavelength ranges, d) ability to vary the resolution, e) easily maintainable detection system and electronics, f) reliable and easy alignment of samples and g) ability for the extension of the instrument for polarized neutrons. Taking into account the absence of a cold neutron source at the GRR-1 reactor, the reflectometer has been designed to operate in Time-Of-Flight (TOF) mode.

Table 2. Characteristics of the time-of-flight reflectometer at the GRR-1 reactor.

Scattering Geometry	Vertical
Incidence Angle	
Liquids	0 – 1.5°
Solids	0 – 5°
Beam Size	Max. 10mm(V)×50mm(H)
Angular Resolution $\delta\theta$	0.05° – 0.5°
Pulse repetition rate	20 – 200 Hz
Chopper–Detector distance	6 – 10 m
Wavelength Range	1 – 10 Å
Wavelength Resolution $\delta\lambda$	0.1 – 0.5 Å

In the TOF technique a white neutron beam is pulsed by means of a chopper, a rotating disk of neutron absorbing material with suitable cut-out windows for shaping the neutron pulse. Neutrons of different wavelengths are

discriminated based on the time they need to reach the detector. The TOF mode has the advantage that a large Q range can be covered in one geometrical setting and a wide wavelength range can be utilized. In addition, the complexity and the time lost in a continuous change of θ, 2θ angles can be avoided and lower Q values can be obtained.

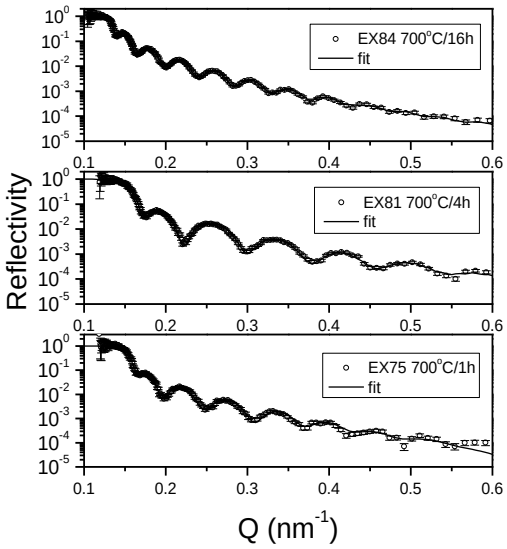

Figure 5 : Neutron reflectivity data from SiC films oxidised at 700°C for 1, 4 and 16 hours. The solid line is a least square fit to the data.

For measuring liquid samples the in-pile collimation system has been designed so that the neutron beam emerging from the reactor is inclined with respect to the horizontal axis. Thus the desired reflection angle θ is achieved by simply moving the apertures and the sample vertically. With this method a maximum value of θ = 1.6º may be achieved for liquid samples. Different wavelength ranges are available and all of them have the same minimum wavelength of 0.15 nm utilizing, thus, the short wavelengths where the GRR-1 reactor offers the maximum flux [7][7]. The instrument resolution is also a variable parameter, determined by the chopper rotation speed and the chopper-detector distance. Thus, the user may choose between high resolution and high counting rate settings, according to the experimental needs. Pulse overlap is avoided by the employment of a supermirror. Since there are no guides and it is essential to reduce the background, a sapphire filter is used for the removal of the fast neutron component. For achieving an efficient removal of the fast neutrons without considerable reduction of the neutron flux in the wavelength range from 0.15 to 1.0 nm the optimum sapphire thickness has been calculated by MCNP code. Further, towards a low background instrument, optimization of the shielding has been made by MCNP calculations.

The layout of the neutron reflectometer is presented in Figure 4 while the instrument parameters are given in Table 2.

An example of the application of the neutron reflectometry technique refers to the oxidization properties of SiC coatings. SiC films of 128 nm nominal thickness were grown on Si wafers by the rf sputtering technique. The samples were subsequently oxidized in air at different temperatures and for various oxidation times. Figure 5 presents the reflectivity data from the SiC coatings oxidized at 700°C for 1, 4 and 16 hours. The solid lines are least square fits to the data. The information that was

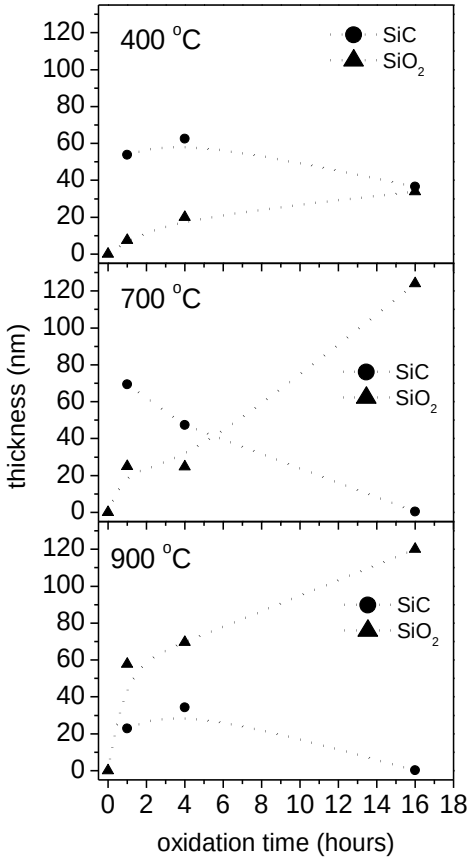

Figure 6 : The thickness of the SiC (● solid circles) and SiO_2 (▲ solid triangles) layer versus the oxidation time at 400, 700 and 900°C. The dotted lines are guide to the eye.

obtained from the oxidized films includes the thickness of a SiO_2 layer that forms during oxidation, the thickness of the remanent SiC layer, as well as the density and interfacial roughness of both layers. The evolution of the thickness as a function of oxidation time, both for the SiO_2 and the SiC layer, is depicted in Figure 6. The dotted lines in Figure 6 are guides to the eye.

2.3 Small Angle Neutron Scattering (SANS)

SANS allows the study of inhomogeneities in a matrix. In a typical SANS experiment the measured quantity is the macroscopic scattering cross-section of the inhomogeneities,

$$\frac{d\sigma}{d\Omega} = \left|F\ C(\mathbf{r})\right|^2 \approx n_p \left\langle V_p^2 \right\rangle \left\langle \Delta\rho \right\rangle^2 S^2(QR) I\ QD \qquad (1)$$

as a function of the scattering wave-vector Q. In the above equation $F\{C(\mathbf{r})\}$ represents the Fourier transform of the neutron scattering length density $C(\mathbf{r})$, n_p is the number density of the precipitates, V_p is the volume of one precipitate, and $\Delta\rho$ is the contrast, which depends generally on the atomic volume in the matrix and in the precipitates and on the neutron scattering lengths of the atoms involved. $S(QR)$ is the Fourier transform of the shape of the precipitate and R is the size of the precipitate. $I(QD)$ describes the interparticle scattering, where D is a characteristic distance between the precipitates.

At GRR-1 a SANS and a USANS instrument are under design. The two instruments will be installed at the same beam tube. SANS will be used for the investigation of large size inhomogeneities (0.05 – 5 μm) and USANS for smaller size inhomogeneities (0.002 – 0.1 μm).

The double crystal USANS instrument will consist of two elastically bent Si crystals, the monochromator and the analyzer (Figure 7). The instrument wave-vector resolution δQ may be tuned in the range from 10^{-4} to 10^{-3} Å^{-1} by adjusting the curvatures of the two crystals, according to the expected size of investigated inhomogeneities. The fully asymmetric diffraction geometry on the elastically bent Si analyzer is employed to transfer the angular distribution of the scattered neutrons to the spatial distribution and to analyze the whole scattering curve by a one-dimensional position sensitive detector.

The SANS instrument (Figure 7) will utilize the beam transmitted through the Si(111) bent crystal. A beryllium crystal cooled at cryogenic temperature will act as a filter, cutting out wavelengths smaller than 3.95 Å. A mirror after the sample may be used to deflect the long wavelength neutrons (for example above 5.5 Å). The scattered intensity will be monitored on an image plate.

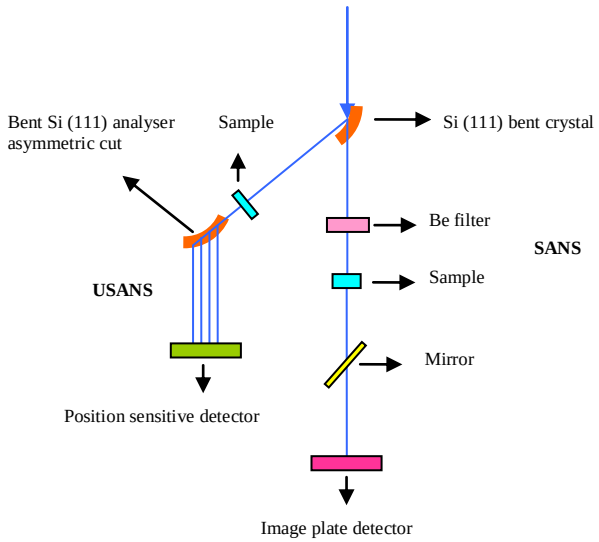

Figure 7: Schematic layout of the SANS and USANS instruments.

The SANS technique can be used in obtaining information about the precipitation in an alloy. Isothermal ageing of AlLi alloys results in the formation of a $L1_2$ ordered metastable δ′ phase of stoichiometric composition Al_3Li The growth of δ′ precipitates in an Al-8.9at%Li alloy was studied by SANS experiments in the temperature range from 90 to 210°C [8]. In Figure 8 is presented the scattering cross section versus the scattering vector Q from an AlLi alloy aged at 210°C for 0.17, 0.50 and 1 hours.

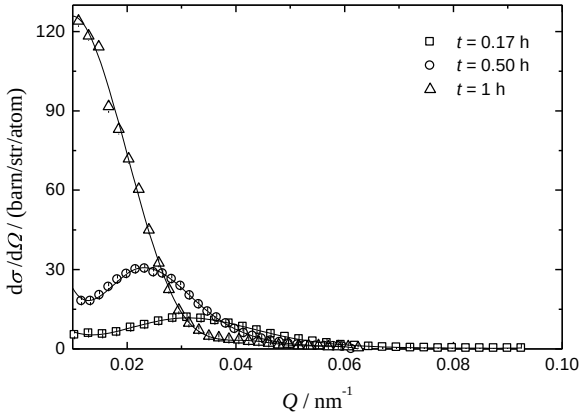

Figure 8: SANS curves from AlLi alloy aged at 210°C for 0.17, 0.50 and 1 hours. The solid lines are guide to the eye.

The radius, number density and volume fraction of the precipitates and the concentration of the solute in the matrix were determined as a function of ageing temperature and time.

The coarsening kinetics predict that the growth of the cube of the size of the precipiates is proportional to ageing time, while the number density of the precipitates varies inversely with time [9]. The proportionality constants are the kinetic constants which depend on the diffusion coefficient and the surface energy. The size of the precipitates obtained from the SANS data agrees very well, for long ageing times, with the predictions of the coarsening theory, as can be seen in Figure 9.

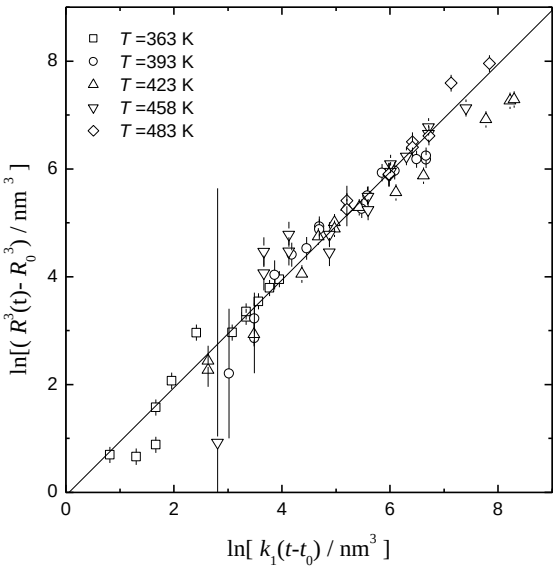

Figure 9: Universal behaviour of the sizes of the precipitates in AlLi alloys according to the coarsening theory.

3 Conclusions

Neutron scattering techniques offer significant advantages for non-destructive testing applications due to the high penetration depth of neutron beams and the unique sensitivity of neutrons to different isotopes and light elements as hydrogen. At the Greek Research Reactor GRR-1 a variety of neutron scattering methods and facilities are employed and developed, offering a wide range of possibilities for non-destructive testing applications. In the present contribution a description of the GRR-1 facilities has been given and some representative application examples have been described. Neutron diffraction has been employed for the study of lattice distortion in SiC_f/SiC composite materials after irradiation. The oxidization properties of SiC coatings has been investigated by means of neutron reflectometry. Finally, small angle neutron scattering has been implemented to determine the kinetics of δ′ phase precipitation in Al-Li alloys.

4 References

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