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Feasibility Study of a Low-Temperature Irradiation Facility at the German Spallation Neutron Source SNQ

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Abstract

A feasibility study was performed on the possible installation of a low-temperature irradiation (LTI) facility at the projected German spallation neutron source SNQ. The LTI facility should allow the irradiation of samples at temperatures between 4.5K and about 450K with either thermal or fast neutrons. We first outline the scientific motivation and then describe the design features and the performance of the facility, which essentially consists of an on-line He refrigerator and two vertical irradiation tubes providing two irradiation positions I and II. In position I a very clean thermal neutron flux $\emptyset_{th} \approx 1 \cdot 10^{14}$ cm^{-2} s^{-1} can be realized, the γ-background being dominated by the contribution from the LTIF structural material. In position II the fast neutron flux is $\emptyset_{f} \approx 2 \cdot 10^{13}$ cm^{-2} s^{-1} (for E > 0.1MeV) or even more. In both cases the dominant heat production comes from the thermal neutrons. The 100s^{-1} time structure of the SNQ represents a major problem for irradiations at the lowest temperatures. Other minor problems for a LTI facility at the SNQ as compared to a new reactor are the high energy neutrons, the secondary protons and some geometrical constraints. The great advantage of the SNQ lies in the very low γ-background and also in its potential of flux tailoring.

1. Introduction

The present concept of the planned German Spallation Neutron Source (abbreviated SNQ from "Spallationsneutronenquelle") assumes a platelike rotating lead target with a vertical axis. The horizontal proton beam of energy 1100MeV, average current 5 mA and repetition rate 100 s^{-1} (pulse length 500 μs) hits the target wheel radially /1/. A large D_2O moderator tank will be placed directly above the target plane (compare fig. 1) and will lead to high time-averaged thermal neutron fluxes $\emptyset_{th}$.

The Low-Temperature Irradiation (LTI) Facility should allow irradiations in the region of this D_2O tank. The samples could have a maximum size of 25 mm diameter and 100 mm length and the irradiation temperature would be either 4.5K in liquid Helium or 5 ÷ 450K in gaseous Helium. Two extreme irradiation positions I and II are considered: in position I the thermal neutron flux should be high and as clean as possible ($\emptyset_{th} \approx 1 \cdot 10^{14}$ cm^{-2} s^{-1}) and in position II the fast neutron flux should be as high as possible ($\emptyset_f \approx 2 \cdot 10^{13}$ cm^{-2} s^{-1} for neutron energies above 0.1MeV). By changing the irradiation position continuously in between these two extremes mixed neutron spectra can be realized, and smaller values of $\emptyset_{th}$ with reduced background radiation are possible beyond position I if necessary.

In what follows we first give an overview of the scientific applications planned for the LTI facility (Chapter 2). The design features of the facility and its estimated performance are described in Chapter 3. Finally, Chapter 4 summarizes the particular aspects which are relevant for a LTI facility at the SNQ as compared to a similar facility on a new middle-flux research reactor /2/. In this context the advantage of a strongly reduced background of γ-radiation and the disadvantage of the time structure of the SNQ are of major concern.- A somewhat more detailed description of the various aspects of a LTI facility at the SNQ can be found in ref. /3/.

2. Scientific Motivation

2.1 Irradiation of Metals with Thermal Neutrons

The emission of prompt γ-rays after thermal neutron capture corresponds to the transfer of small effective recoil energies to the irradiated nuclei and so leads to the production of single vacancies and self-interstitials in metals, i.e. of Frenkel defects. The radionuclides which are simultaneously produced in most cases give rise to some doping and can often be used as radioactive probes for investigating the samples. In contrast to electron irradiation a statistical distribution of isolated Frenkel defects can be produced also in bulk samples. The fast neutron flux must be sufficiently low (see Chapter 3.2) to avoid the production of defect agglomerates in high-energetic cascade processes. A low irradiation temperature is necessary to immobilize the defects produced.

The technique of low-temperature irradiation in a high thermal neutron flux practically opens a new field of research applications, the investigation of isolated Frenkel defects with neutron scattering methods, since bulk samples can be provided. Inelastic measurements should enable the search for low-frequency resonant vibration modes of isolated dumbbell interstitials under significantly better conditions than before and so help to answer a fundamental open question. Elastic diffuse neutron scattering measurements allow the investigation of the size and symmetry of the displacement fields of the point defects and, after stepwise annealing, of defect agglomerates; they complement analogous measurements using X-ray scattering techniques, where the experimental parameters are different.

The radioactive probes can be used for measurements of the Mößbauer effect and of the perturbed angular correlation. Since the probe nucleus as well as the radiation damage have been produced in the same (n,γ) elementary process they are correlated in space. Investigations of this type can be compared with "uncorrelated" experiments, where the probe atoms have been brought into the samples already before irradiation. In all these cases the trapping of interstitials or vacancies at the probe nuclei becomes detectable via hyperfine interactions.

In addition the following types of measurements are planned:
elastic constants, mechanical and magnetic properties, precise defect production rates and ideal isochrones, and positron annihilation and myon spin rotation.

2.2 Irradiation of Nonmetals with Thermal Neutrons

During low-temperature irradiation of semiconductors, insulators, organic and biological substances with thermal neutrons not only is one interested in the structural defects, which have been discussed in Chapter 2.1 and which now occur in different charge states,but also in the subsequent chemical reactions. Because of the radiolysis problem in nonconducting materials a very pure thermal neutron flux is now particularly essential, i.e. above all the γ dose rate must be sufficiently low (see Chapter 3.2).

There are many important research applications which are specific for nonmetals and which will be realized in addition to those already discussed for metals. We begin with neutron transmutation doping of solids at low temperature. In nonmetals the simultaneously produced correlated point defects and their annealing behaviour can be studied very efficiently with optical methods, ESR and NMR. The spectroscopic properties of the isolated foreign atoms are interesting in many cases, too (matrix isolation technique, e.g. K in the solid rare gas Ar). Positron annihilation in nonmetals often goes along with the formation of a positronium atom which can chemically react with the radiation transmuted foreign atoms (positronium chemistry). The diffusion behaviour of the statistically distributed and radioactive foreign atoms can also be investigated (e.g. Ar in the ionic crystal KCl).

The field of hot atom chemistry in solids deals with the study of the chemical reactions of the recoil atoms. These "hot" atoms are characterized by both high kinetic and electronic energies. A low irradiation temperature is important to suppress competitive thermal reactions. Hot atom reactions can also be used e.g. for the preparation of rare chemical compounds, and in many cases they allow the production of starting material for a rapid labelling of radiopharmaceuticals.

In a similar way one can also investigate and optimize the production and high enrichment of radioisotopes. During the irradiation of an appropriate complex compound the hot isotopes produced are knocked out ot their chemical bonds and can be easily separated after the irradiation. Again unfavourable interfering reactions can be suppressed by a low temperature and a very low γ dose rate.

Activation analysis experiments must be performed at low temperature if the irradiated substances are sensitive to thermal or radiolytical decomposition, or if the radioisotopes produced are volatile.

2.3 Irradiation with Fast Neutrons

The high recoil energies transferred during the collisions of MeV neutrons with sample nuclei lead to the formation of displacement cascades. It is possible again to use bulk samples. The thermal flux ϕ_{th} should be sufficiently low to reduce unfavourable activation and heat production effects (see Chapter 3.2).

Most of the research applications of thermal neutron irradiation as considered in the previous chapters for metals and, to some lesser degree, for nonmetals will also be realized with fast neutrons under otherwise identical experimental conditions. This is on the one side for comparison purposes, since because of the cascade nature of defect production the degree of agglomeration of the Frenkel defects is relatively high already at liquid helium irradiation temperature, and on the other side due to the fact, that the defect production rate is much larger and hence saturation effects can be investigated now.

In addition there are experiments which shall be mainly performed with fast neutrons. A major field of interest will be the investigation of decomposition and segregation phenomena in concentrated alloys. Since up to now fundamental research has concentrated mainly on the irradiation of pure metals and dilute alloys, this will represent an important trend in the future. Small angle scattering measurements will certainly be useful.

The study of the radiation stability of insulators and technical superconductors is a more technological application. In future fusion reactor magnets these materials will be irradiated during regular operation in very much the same way as in the LTI facility, i.e. with fast neutrons at liquid helium temperature; however, the intensity in the fusion reactors will be smaller by some orders of magnitude so that the time scale will be speeded up in these experiments.

A somewhat different application is the immobilization of dislocations in cold worked material under external stress by irradiating the bulk samples with fast neutrons. In this way the dislocations get pinned by the large defect cascades whereas creep is suppressed by the low temperature. After irradiation and warm up the external stress can be relieved and the samples can be cut and thinned into the extremly thin foil samples which are necessary for observation in the transmission electron microscope, where the unperturbed dislocation pattern can be investigated.

3. Design and Performance of the Facility

3.1 Design Features

The concept of a low-temperature irradiation (LTI) facility as presented in this chapter is based on that of the existing LTI facility at the Munich research reactor FRM which has been used now for over 20 years with outstanding success /4/.

The LTI facility at the SNQ would have its own liquid He refrigerator with a cooling capacity of about 400 W at 4.5K, which would be installed in the hall directly above the target. From there the whole LTI facility would be operated and the irradiation experiments would be performed. The He refrigerator would be connected, probably by flexible tubing, to two vertical irradiation tubes B and A for irradiations with either thermal (position I) or fast (position II) neutrons, respectively; see also fig. 1. On top of each irradiation tube a transfer or measuring cryostat could be plugged in and connected to the He system of the facility. In this way cold sample transfers into or out of the facility could be achieved, e.g. for external measurements, but in many cases the measurements could be performed in the measuring cryostat on the LTIF before or after the irradiation or, in simpler cases, even in situ during the irradiation.

A sample can have a maximum size of about 25 mm Ø · 100 mm and will be fastened to the end of a metal capillary being about 10 m long and usually containing electrical measuring leads. By means of this capillary the sample can be moved through an irradiation tube down into the irradiation position, being always cooled in a direct flow of either liquid Helium (4.5K) or gaseous Helium having the desired temperature (5 ÷ 450K). Both irradiation tubes are about 8 m long and are vacuum shielded from cryogenic reasons and doubly bent (about 16° each) for radioprotection purposes. They are installed along with shielding material in vertical channels in the biological shield of the SNQ, so that some limited vertical movement remains possible.

As shown in fig. 1, the lower end of the irradiation tube A is located in a vertical channel through the D_2O moderator tank in the SNQ vacuum. It extends about 30 cm below the bottom of the D_2O tank and there is located by the side of the proton beam and in front of the target wheel. At this point a very good irradiation position II for fast neutrons is provided. However, it will be very much necessary that the high thermal flux ϕ_{th} in the bottom region of the tank can effectively be shielded with respect to position I and even then the large tube length in the tank being exposed to high values of ϕ_{th} remains a major problem (activation of the sample capillary, heat production). Mixed irradiation spectra can be realized by leaving the sample in some distance above position II whereas the irradiation tube A itself has to be lifted by about 30 cm only in case of a necessary dismounting of the target wheel.

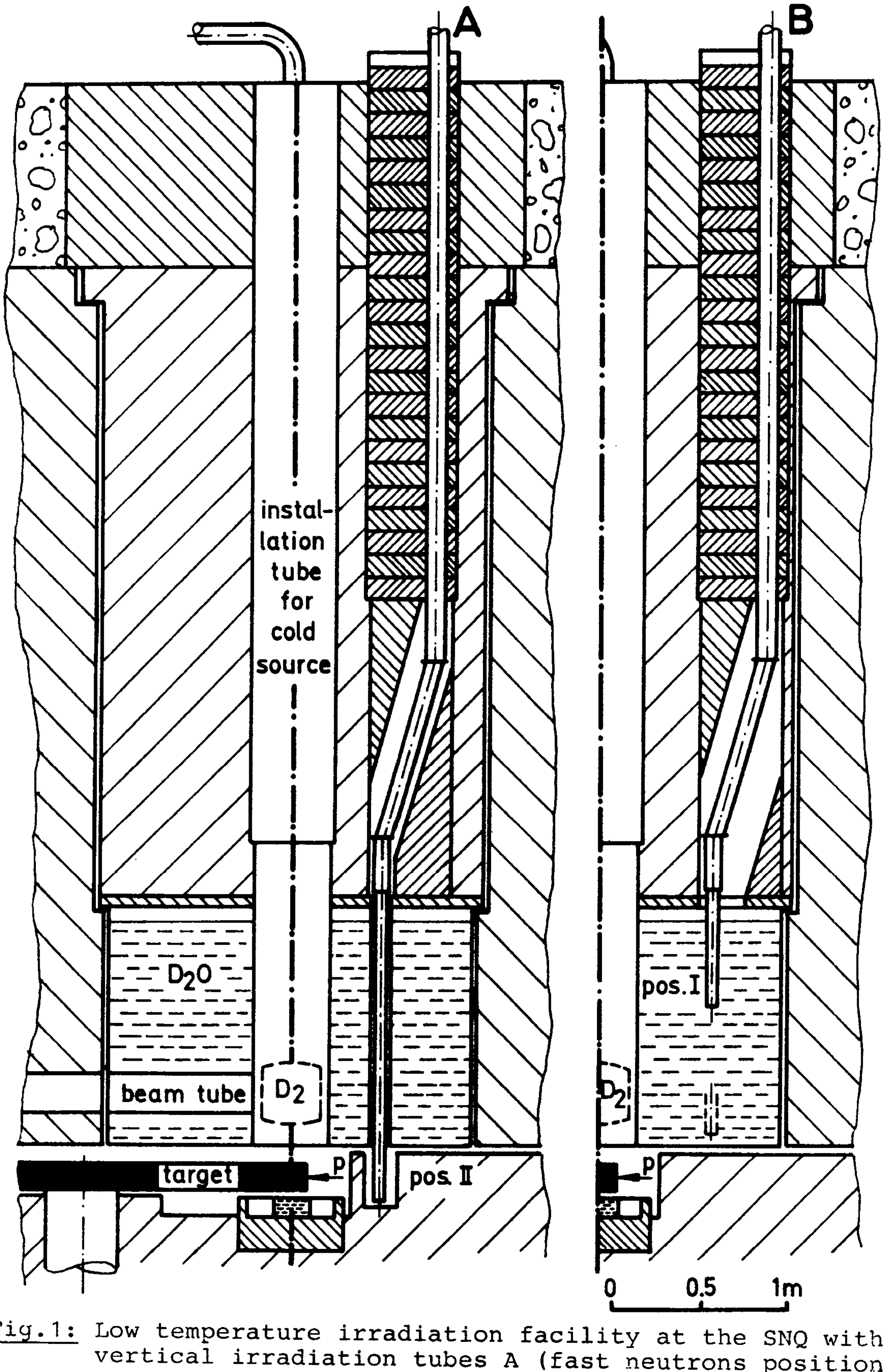

Fig.1: Low temperature irradiation facility at the SNQ with vertical irradiation tubes A (fast neutrons position II) and B (thermal neutrons position I)

The irradiation tube B is immersed directly in the liquid D_2O of the moderator tank and can be vertically moved by about 100 cm. In the position I shown in Fig. 1 the lower tube end finds itself in a practically unperturbed D_2O environment as desired for irradiations in a very pure spectrum of thermal neutrons. Irradiations in a smaller thermal flux $\emptyset_{th}$ with correspondingly reduced background radiation are possible by further lifting the irradiation tube above position I, whereas mixed spectra can again be realized below position I.

3.2 Flux Values and Heat Production

For a LTI facility at an intense neutron source the problem is not only to realize very high thermal or fast neutron fluxes in the irradiation positions, but also (or even more) to achieve sufficiently pure neutron spectra with low background radiation and, for irradiations at the lowest temperatures, to keep the nuclear heat production small enough so that the samples can still be effectively cooled. Taking up this latter argument, we can obtain a conservative estimate by assuming a maximum stable heat flow density of 0.5 W/cm^2 from the sample surface to boiling liquid helium ($T \approx 4.5K$) /3,5/. It follows that for long cylindrical bulk samples of radius r the maximum tolerable rate of nuclear heat production $\dot{q}$ is of the order of 10 W/cm^3 for r = 1 mm and of 1 W/cm^3 for r = 1 cm. The sample surface temperature would be about 5.0K, then, and there would be an additional temperature gradient within the sample as determined by its bulk thermal conductivity. At values of $\dot{q}$ higher than just mentioned an instable transition from bubble to film evaporation sets in and so an irradiation of samples can only be guaranteed at temperatures well above that of liquid helium. Further significant problems arise from the SNQ time structure which is not considered here (compare Chapter 4) since in this chapter we assume a steady state situation /3/.

In the irradiation position I (fig. 1, tube B) the thermal neutron flux $\emptyset_{th}$ should be high and as clean as possible. A value of $\emptyset_{th} \approx 1 \cdot 10^{14}\ cm^{-2}\ s^{-1}$, which is very attractive for the irradiation of metals (Chapter 2.1), can be realized in the D_2O moderator tank in about 1 m distance from the target wheel /1/. Examples of the nuclear heating rate $\dot{q}_{th}$ corresponding to this value of $\emptyset_{th}$ are 0.48 W/cm^3 for an Al-sample and 10.6 W/cm^3 for Cu /3/. All other contributions to nuclear heating as produced by other types of radiation are comparatively small (see also below). For the fast neutron flux $\emptyset_f$ which counts all neutrons with energies above 0.1MeV a ratio of $\emptyset_{th}/\emptyset_f \gtrsim 5 \cdot 10^3$ can well be achieved, which is also very favourable since cascade effects in the samples are very effectively suppressed, then. The applications in the field of nuclear chemistry (Chapter 2.2) desire a value of the γ dose rate D_γ as low as about 10^6 rad/h. Although the direct γ radiation from the SNQ target is smaller by a factor of about 10 as compared to that from an equivalent reactor core, whence this direct contribution is $D_\gamma < 10^6$ rad/h, indeed, the secondary γ (and β) radiation from the structural material of the LTIF irradiation tube is proportional to $\emptyset_{th}$ and yields a contribution of about $D_\gamma \gtrsim 6 \cdot 10^6$ rad/h. In critical experiments the distance of the irradiation position from the target could be further increased in order to reduce $\emptyset_{th}$ and correspondingly $\dot{q}$ and

D_γ . The flux of secondary protons in position I is $\emptyset_p \lessapprox 2 \cdot 10^9$ cm^{-2} s^{-1} which is negligible for both $\dot{q}$ and D_γ .

In the irradiation position II (fig. 1, tube A) the fast neutron flux $\emptyset_f$ should be high. A value of $\emptyset_f \approx 2 \cdot 10^{13}$ cm^{-2} s^{-1} is attractive again for the scientific applications mentioned in Chapter 2.3 and can be realized in about 40 cm horizontal distance from the target wheel /3/. This value of $\emptyset_f$ gives rise to a nuclear heating rate $\dot{q}_f$ of 0.12 W/cm³ for Al and 0.14 W/cm³ for Cu. The main heat input, however, is due to the thermal neutron flux $\emptyset_{th}$ which is expected to be pretty large due to the nearby D_2O tank and which has to be effectively shielded. Assuming $\emptyset_{th} \approx 5 \cdot 10^{13}$ cm^{-2} s^{-1} in position II we obtain values $\dot{q}_{th}$ of 0.24 W/cm³ for Al and 5.3 W/cm³ for Cu. The contribution $\dot{q}_\gamma$ of the direct γ radiation from the target is again relatively small as compared to a reactor and is estimated to be 0.15 W/cm³ for Al and 0.4 W/cm³ for Cu. Finally, the flux of secondary protons is effectively reduced by some additional lead shielding; assuming about 10 cm of Pb as in fig. 1 we estimate $\emptyset_p \approx 3 \cdot 10^{10}$ cm^{-2} s^{-1} and $\dot{q}_p \approx 0.07$ W/cm³ for Al and 0.2 W/cm³ for Cu. Summing up the individual contributions to $\dot{q}$ we obtain 0.6 W/cm³ for Al and 6.1 W/cm³ for Cu /3/.

4. Special Considerations

We will now mention some specific aspects /3/ which are important for a low-temperature irradiation (LTI) facility installed at a spallation neutron source, and in particular at the German SNQ /1/, in comparison with a similar LTI facility say at a new D_2O reflected middle flux reactor /2/.

The 100 s^{-1} time structure of the SNQ implies that all direct radiation from the target (unmoderated neutrons, secondary protons, γ, ...) will be emitted in pulses of 500 µs length and with a peak to average ratio of 20, whereas this ratio is still about 10 for the broadened pulse of thermal neutrons in the D_2O tank /1/. It follows that the time-averaged rates of nuclear heat production $\bar{\dot{q}}$ as discussed in Chapter 3.2 (with $\dot{q} \equiv \bar{\dot{q}}$) have to be multiplied by these factors to obtain the maximum rate of heat production $\hat{\dot{q}}$ in the pulse. To illustrate the problem we will assume for a moment that we have a Cu sample with $\bar{\dot{q}} = 5$ W/cm³ and that sample cooling during the pulse can be neglected compared to cooling between the pulses (or equivalent: δ-pulses); then the energy deposition of $\bar{\dot{q}}/(100 s^{-1})$ induces a jump of the sample temperature from 4.5K to about 12K during the pulse. Higher values of the sample specific heat or of the irradiation temperature or lower values of $\bar{\dot{q}}$ reduce this effect. Although in the real case there will be some sample cooling also during the pulse, the possibility of instabilities in the heat transfer to the flowing liquid helium at too high values of $\hat{\dot{q}}$ (which definitely exist in a static helium bath /5/) make the problem difficult and experimental investigations necessary. The problem does not exist at all on a reactor and also not on a quasi-continuous spallation neutron source (e.g. with a proton-cyclotron) since the time separation of the micropulses is much shorter than the time constants of the heat transfer problem /3/.

The presence of high-energy neutrons with energies between about 10MeV and that of the proton beam (1100MeV at the SNQ) is a unique feature of spallation neutron sources. For a Pb target they represent about 5% of all neutrons, which also means about 5% of the fast neutrons in irradiation position II. The heat which they produce in the samples is negligible. Their contribution to the production of Frenkel defects in metals (Chapter 2.3) is larger by about a factor of three than their contribution to the total fast neutron flux ϕ_f since the recoil energies are much higher. However, due to the formation of subcascades a relatively similar structure of the defects (agglomeration etc) is expected as in the case of reactor neutrons, although the amount of gas production from (n, p) and (n, α) reactions will be larger by one to three orders depending on the sample material.

The spectrum of high-energy secondary protons also extends up to the energy of 1100MeV of the SNQ primary proton beam. In irradiation position II the fast proton flux ϕ_p corresponds to about $1.5 \cdot 10^{-3}$ of the total flux ϕ_f of fast neutrons (Chapter 3.2). First, these protons undergo relatively low energy transfer Rutherford scattering processes and so produce Frenkel defects in metals at a rate which is typically some 10^{-3} of that of the fast neutrons. Second, due to the high proton energies nuclear processes are possible, too, which are roughly equivalent to those of the high-energy neutrons but which contribute only with a few percent of those because of the smaller flux. Third, the proton interaction with the electrons accounts for the main part of the proton heat production in metals, whereas in addition defects are produced in nonmetals.

The heavy biological shielding which is necessary at spallation neutron sources to attenuate the high-energy radiation components (see fig. 1) makes the design and operation of a LTI facility relatively inconvenient. This becomes obvious by comparison with a swimming-pool type research reactor as in ref. /2/ where the LTI facility could easily be moved around or taken out for repair and modification purposes. In particular, in the present design of the German SNQ /1/ the D_2O moderator tank is most favourable of course for providing an excellent irradiation position I for thermal neutrons, see fig. 1, but the problems of the D_2O tank for the fast neutron position II have already been mentioned in Chapter 3.

The main and essential advantage of a spallation neutron source as the SNQ for a LTI facility is its strongly reduced level of direct γ radiation. Although it is difficult to obtain reliable numbers for this behaviour, there seems to be general agreement that the direct γ radiation from a nonfissionable SNQ target is about one order of magnitude smaller than that from the core of an equivalent reactor. In contrast, it is obvious from Chapter 3.2 that in the case of an irradiation position II being realized at an equivalent reactor the direct γ dose rate would produce an essential and often the dominant contribution to the heat production in the samples.

Finally, also a general but perhaps practically not so important advantage is the SNQ potential of flux tailoring. We have emphasized in Chapter 3.2 how essential it is for a LTI facility to provide proper neutron spectra and fluxes as well as acceptable levels of background radiation for the irradiation positions I and II. At a spallation neutron source such an optimization procedure can be generally performed more rigorously than at a reactor since no criticality criteria have to be satisfied.

Acknowledgement

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