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Irradiation Techniques Employed at the 10 MW_{th} SAPHIR Reactor*

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CONTENTS

Abstract

The 10 MW_{th} open-pool research reactor, SAPHIR, meets the requirements for a range of neutron irradiation applications. These include radioisotope production for research, medicine and industry, radiation damage studies for fission and fusion reactor materials, radiochemistry and neutron activation analysis, industrial irradiations such as silicon transmutation doping and neutron radiography and – last but not least – neutron scattering research using the various beam tube facilities. The current paper describes the corresponding irradiation techniques developed and being applied at the reactor.

Zusammenfassung

Der Forschungsreaktor SAPHIR, ein offener Pool-Reaktor, wird mit 10 MW_{th} betrieben. Er dient gleichzeitig einer Vielzahl von Anwendungen der Neutronenstrahlung. Diese reichen von der Produktion von Radioisotopen, die in der Forschung, der Medizin und der Industrie verwendet werden über Untersuchungen der Strahlenschäden in Materialien der Reaktor- und Fusionstechnologie, die Radiochemie und Neutronenaktivierungsanalyse bis hin zu industriellen Bestrahlungen, wie z.B. die Silizium-Dotierung sowie der Neutronenradiographie. Die Strahlrohre des Reaktors werden vor allem für Neutronenstreuexperimente genutzt. Im vorliegenden Bericht werden jene Verfahren und Techniken beschrieben, die bei Bestrahlungen am Reaktor zur Anwendung kommen.

1 Introduction

Following successive upgrades during earlier years, the Swiss multipurpose research reactor, SAPHIR, commenced operation at its present power level of 10 MW_{th} in January 1984. In 1988, the two federal research institutes, EIR (reactor research) and SIN (nuclear research), were fused into a single unit, the Paul Scherrer Institute. Since then, the formulation of new research policies has led to a modification of priorities for the various types of neutron irradiation applications possible with the SAPHIR reactor. These include radioisotope production for research, medicine and industry, radiation damage studies for fission and fusion reactor materials, radiochemistry and neutron activation analysis, industrial irradiations such as silicon transmutation doping and neutron radiography and – last but by no means least – neutron scattering research using the various beam tube facilities. The current paper describes the corresponding irradiation techniques developed and being employed at the reactor.

2 The SAPHIR Facility

The SAPHIR reactor is basically of the swimming-pool type employing MTR fuel elements [1]. Fig. 1 gives a vertical sectional view of the building and indicates the principal reactor components. The location of some of the experimental devices discussed later is indicated in the simplified plan view of Fig. 2. Fig. 3 shows a typical SAPHIR core configuration along

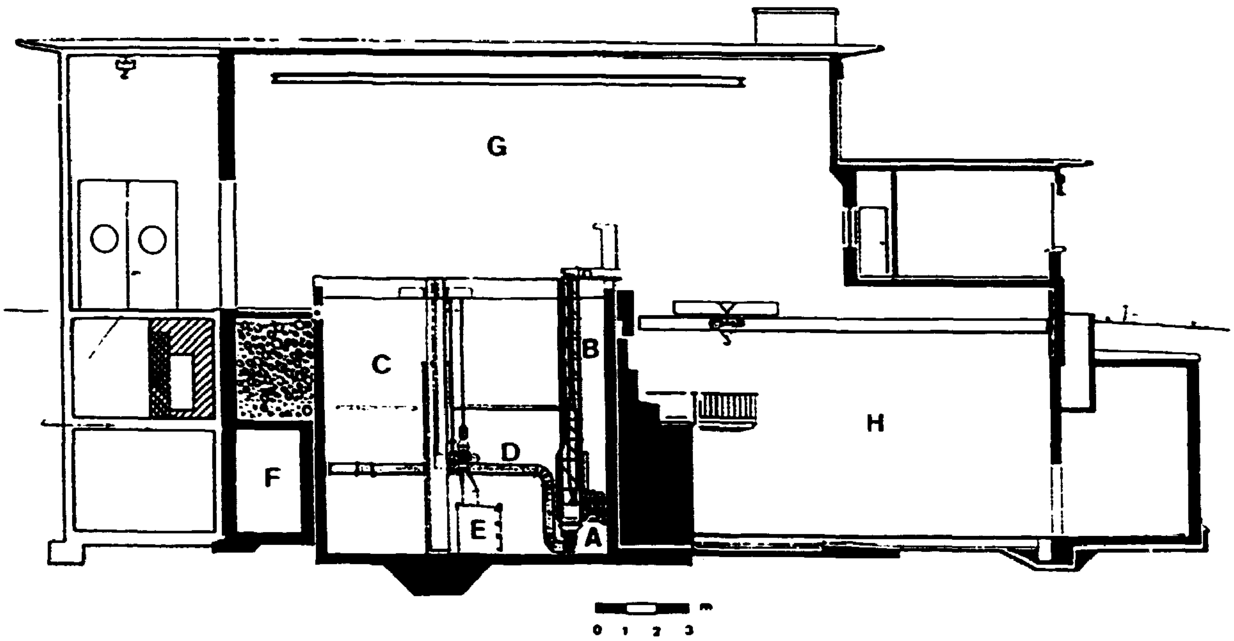

Fig. 1. Vertical sectional view of the SAPHIR reactor building indicating the reactor core (A) and mast structure (B), the reactor pool (C), the primary coolant outlet pipe (D), the diffuser (E), the delay-tank and heat-exchanger room (F), the reactor hall (G) and the experimental hall (H). The 5 m extension of the building (light-shaded, on the left-hand-side) will be constructed in 1993.

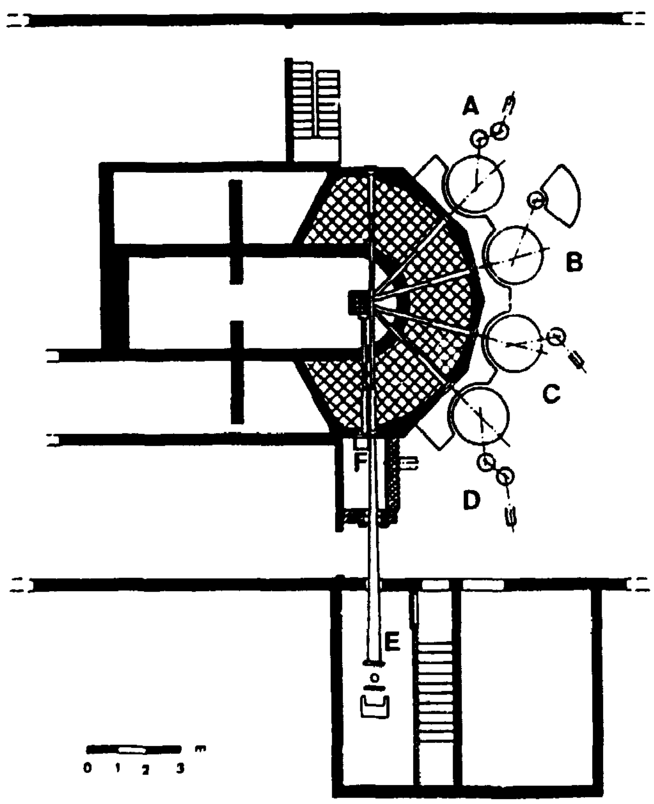

Fig. 2. Simplified plan view of the SAPHIR reactor and experimental hall indicating various experimental devices (A to D: neutron spectrometers, see Table 1 for details; E: neutron radiography (NR), 12 m station; F: gas-jet and 5 m NR-station).

with the beam tube arrangement in the reactor pool and, once again, positions are indicated for some of the different types of irradiation experiments described in subsequent sections.

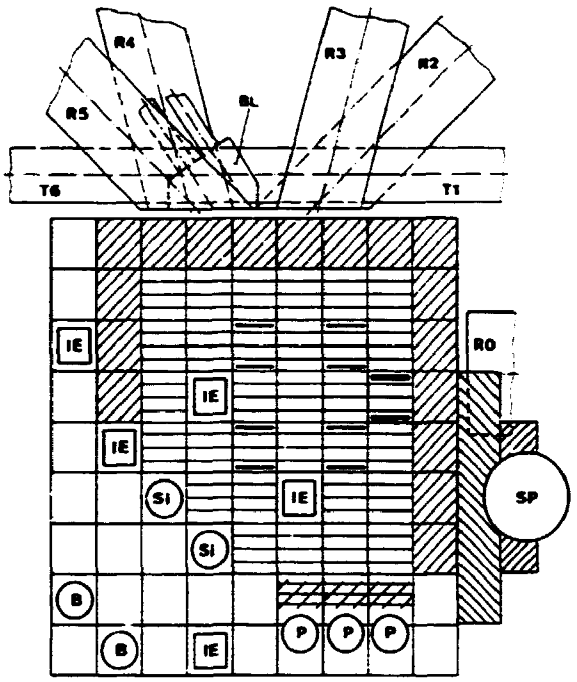

Fig. 3. A SAPHIR reactor core configuration consisting of 26 MTR fuel elements, 5 control elements and 17 beryllium reflector elements. The radial and tangential beam tubes (R0 - R5, T1 and T6) are indicated, as are also typical positions for various types of irradiation experiments (IE: isotope element, B: BWR position for STILO rig, P: PWR STILO-position, SP: SPIRTS facility, BL: BLASIUS irradiations, Si: 62.5 mm-diameter silicon transmutation doping).

A standard core in SAPHIR consists of 30 to 32 MTR elements with beryllium reflector elements usually on two sides in a, broadly speaking, 7 x 8 arrangement. Three different types of fuel elements are currently in use, viz. with high (93%), medium (45%) and low (20%) ^{235}U-enrichment fuel, respectively. The mean burn-up of the core is about 30% at begin-of-cycle, the target discharge burn-up of >60% for each fuel element being achieved by appropriate end-of-cycle shuffling of the fuel. As indicated in Fig. 3, five of the core positions are occupied by fork-type MTR control elements – four (with Ag-In-Cd blades) serving as shim and safety rods and the fifth (with stainless steel) providing fine control. The thermal neutron flux values are upto 1.2 x 10^{14} $cm^{-2}s^{-1}$ for the in-core irradiation positions, 0.8 x 10^{14} $cm^{-2}s^{-1}$ at the core edge and 1.0 x 10^{14} $cm^{-2}s^{-1}$ at the beam tubes behind the beryllium reflector.

The normal operational schedule for the reactor is three weeks at full power (three 8-hour shifts daily), followed by a week of low-power operation during which the reactor – apart from maintenance work and core reloading – is used for training and other experiments. With an average of ~5800 hours of full-power operation per year, SAPHIR figures among the most constantly used research reactors worldwide. The operational crew present in the reactor building consists of just two persons – an operator in the control room and a shift supervisor who is also responsible for carrying out most of the handling operations related to the irradiation experiments. A reactor engineer on call (common for all three shifts) effectively heads the operational team.

3 Radioisotope Production

The procedures developed and being applied for radioisotope production at SAPHIR rely heavily on manual handling of the irradiation targets by the reactor operational staff. The advantages thereby are flexibility in the choice of irradiation positions as well as easy access to the reactor core for experiments and operational changes.

Upto 19 doubly packed and sealed aluminium capsules can be loaded horizontally into a single, so-called isotope element (Fig. 4). These special elements which have the same outer dimensions as the MTR fuel elements can, in turn, be introduced into core-edge positions without shutting down the reactor. For irradiations at in-core positions (see Fig. 3), the reactor is shut down briefly before transferring an isotope element into or out of the core – in order to avoid any reactivity fluctuation effects (reactor period changes) during the operation.

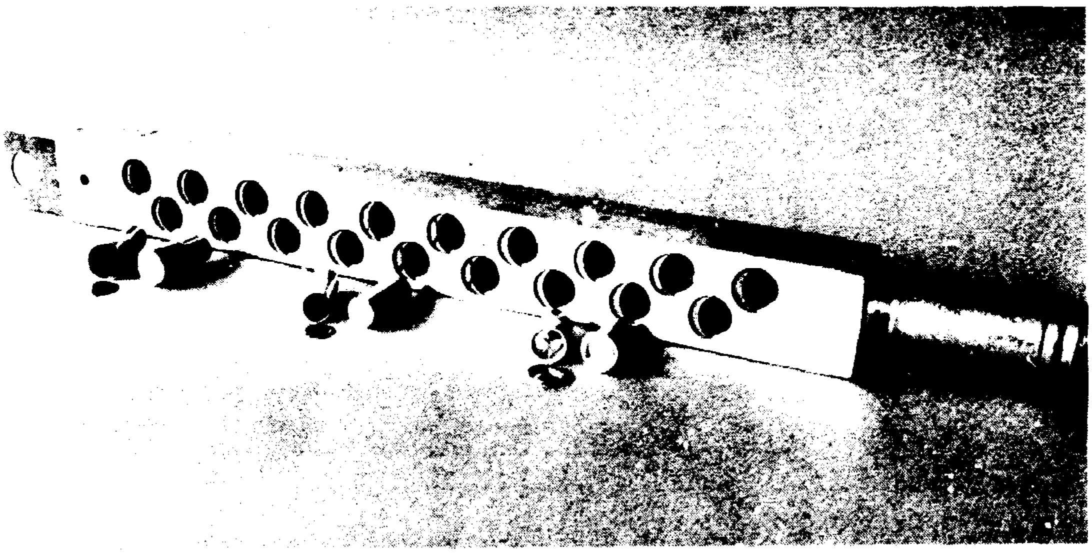

Fig. 4. A SAPHIR isotope element with different types of aluminium irradiation capsules.

After irradiation, the targets are transferred under water from the unloaded isotope elements into lead transport flasks which are then taken to a separate building for appropriate product preparation in hot cells (Fig. 5), packaging and dispatch. The various technical and administrative tasks for routine radioisotope production at the reactor are organized by the SAPHIR irradiation services co-ordinator.

In quantitative terms, ^{131}I has dominated the radioisotope production effort at the reactor in recent years, accounting for 50 to 70% of the total number of capsules irradiated (20 to 30%, in terms of isotope element transfers). A range of radiopharmaceuticals for diagnosis, therapy and labelling are marketed by PSI's Radiopharmacy Department using the ca. 3 kCi of ^{131}I produced in the reactor annually [2]. At the same time, increasing efforts are being invested in the development and production of the tumour-therapy nuclides ^{153}Sm, ^{111}Ag, ^{90}Y and ^{186}Re.

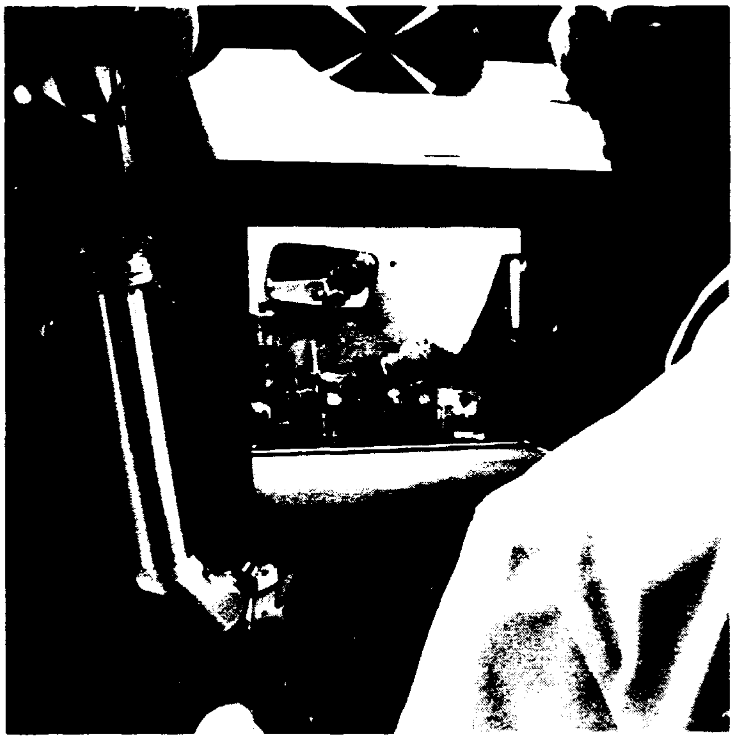

Fig. 5. Radioisotope products being prepared from SAPHIR-irradiated targets in a hot cell.

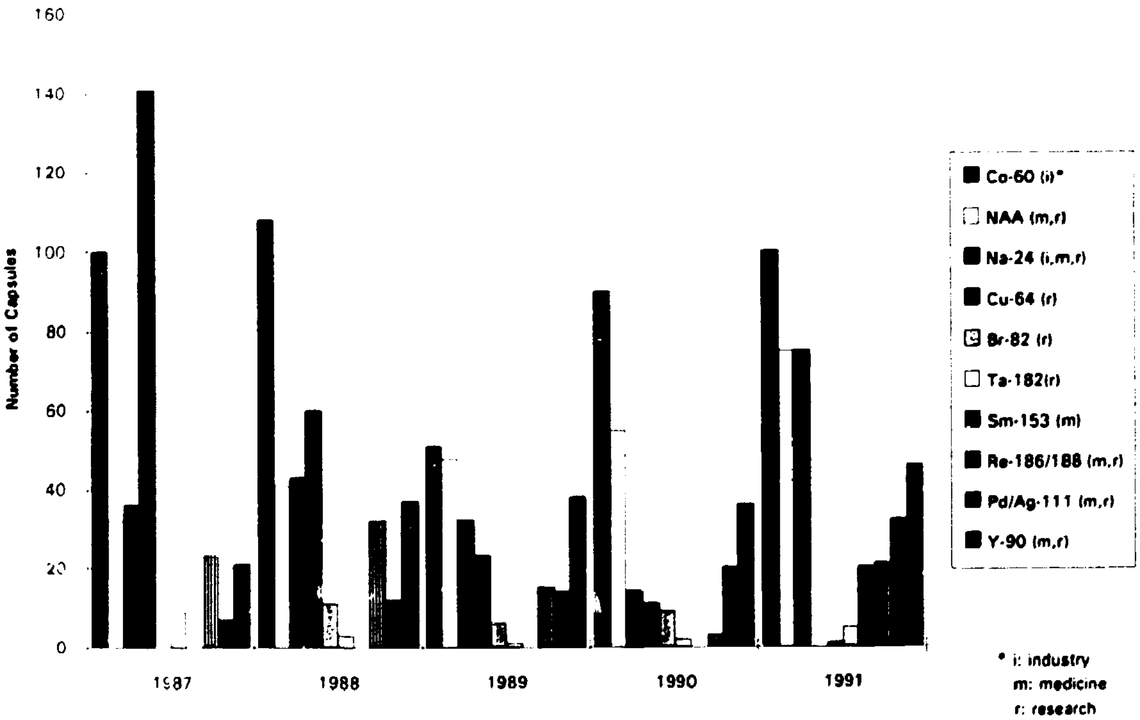

Fig. 6. 1987-91 statistics for capsules irradiated in SAPHIR for the production of radioisotopes other than ^{131}I.

Fig. 6 indicates the principal isotopes (other than ^{131}I) produced in SAPHIR during 1987-91 for various applications in research, medicine and industry [3]. (Neutron activation analysis

(NAA) samples irradiated in isotope elements are shown alongside for the sake of completeness.) It is seen that, in spite of significant annual variations in the relative amounts of the individual products, the total number of capsules irradiated has remained fairly constant for the isotopes shown in the figure.

4 Radiation Damage Studies

Radiation damage studies in the recent past at SAPHIR have employed special, so-called STILO (steel irradiation loops) rigs for the investigation of irradiation embrittlement and annealing of reactor pressure vessel steels [4]. Both PWR and BWR conditions have been simulated – in terms of neutron spectrum and irradiation temperature – and the investigation of specific effects (such as dose rate, sample composition, heat treatment, etc.) has been given due consideration in the experimental programme.

Fig. 7 gives a cut-away view of a fully instrumented STILO rig with a standard loading of test specimens. Irradiation positions for the rigs in SAPHIR are indicated in Fig. 3, along with those for the neutron spectrum "tailoring" elements (aluminium/INOX) in the case of the PWR experiments. For the close-core positions (fast flux values in the range of 1 to 4 x 10^{12} $cm^{-2}s^{-1}$), the irradiation temperature of ~290 °C is achieved via γ-heating of the specimens, a specific gas-gap geometry and a He/N_2 gas-mixture control. In the case of the off-core positions (fast flux of 1 to 4 x 10^{11} $cm^{-2}s^{-1}$), where γ-heating effects are negligibly small, a double-zone heater integrated into the specimen holder is installed.

The investigation of annealing effects in the STILO programme has included so-called IAR (irradiated-annealed-reirradiated) experiments, in which the rigs are removed from the reactor after receiving 50% of the target fluence. A specially designed multizone furnace is used for the desired annealing treatment of the entire specimen volume in a hot cell, after which the rig is reintroduced into the reactor for further irradiation.

In addition to mechanical (fracture mechanics) testing of the irradiated RPV steel specimens, solid state physics techniques (such as neutron scattering and positron annihilation methods) have also been applied in the STILO experiments. Not only have the microphysical methods provided useful insight into embrittlement and recovery mechanisms, they have also pointed out the need for complementary studies in pure systems, i.e. in related simple alloys.

A research programme on the fundamental questions of radiation-induced phase separation in alloys – as also concerning critical current enhancement in neutron-irradiated high temperature superconductors – has recently been initiated by PSI's Materials Science Department [5]. The versatile irradiation facility to be used at SAPHIR for the purpose has been named SPIRTS (spectrum, irradiation temperature, stress), its planned location with respect to the reactor core being indicated in Fig. 3. Small high-purity specimens ($\leq$ 3 cm^3) will be transferred under UHV conditions through three vertical lift tubes into separate irradiation blocks placed one above the other, each with a different spectrum tailoring material (tungsten, aluminium and water). Straining devices will establish desired in-situ specimen strain values, and the irradiation temperature will be variable down to liquid nitrogen. The first SPIRTS irradiations are scheduled for Autumn 1992.

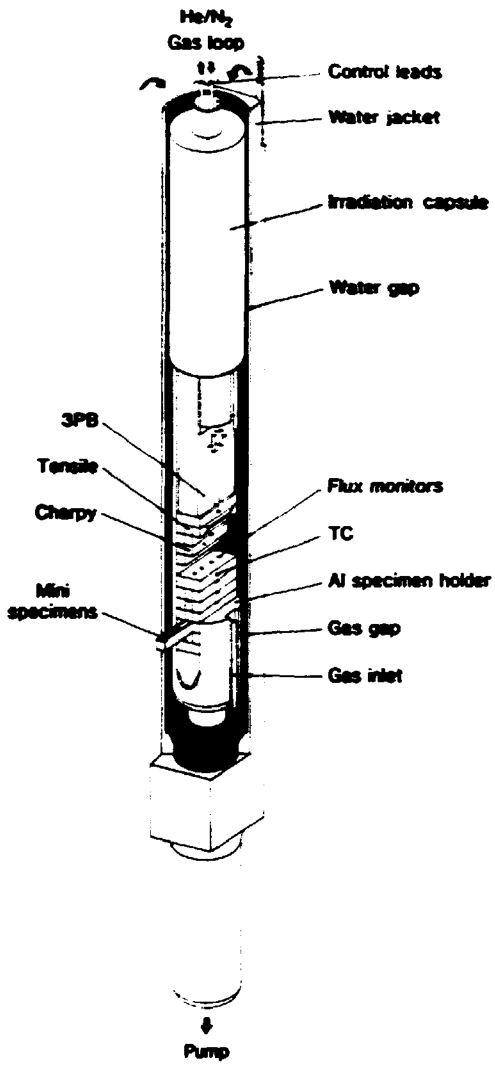

Fig. 7. Cut-away view of a STILO rig with a standard loading of test specimens (available test volume: 30 x 50 x 370 mm^3).

Modification of one of the STILO rigs to permit higher irradiation temperatures is part of the planning for a new PSI fusion-technology research programme concerning the irradiation behaviour of low-activation materials [6]. The proposed experiments in SAPHIR are planned for 1993-94.

5 Radiochemistry Applications

Neutron activation analysis (NAA) via irradiations in SAPHIR has been applied for trace element detection in a range of research projects – at the PSI Chemistry Department, as well as at various Swiss universities (see, for example, [7]). While the irradiation of NAA samples in isotope elements (Section 3) has been one method, a pneumatic "rabbit" system, BLASIUS, with two independent transfer stations has accounted for the bulk of NAA work at the reactor – with upto ~1000 activations carried out annually. The irradiation position in the reactor, located above the tangential beam tube, is indicated in Fig. 3, the neutron flux values being ~2 x 10^{13} thermal and 4 x 10^{11} fast ($cm^{-2}s^{-1}$). (For epithermal irradiations, the "rabbit" can be equipped with a boron-nitride insert.) One of the BLASIUS transfer stations is in the reactor building itself, but the more frequently used station is that in the Chemistry Building more than 100 m away, the transport time for an irradiated sample being ~10 s between reactor and laboratory.

For NAA samples of dimensions larger than can be accommodated in BLASIUS (or in an isotope element), a rotating-tray arrangement is available for use at core-edge positions. The loading/unloading operations are, once again, manual – with correspondingly longer reactor-to-laboratory transfer times. Groundwater studies have been the most common NAA application of this latter type.

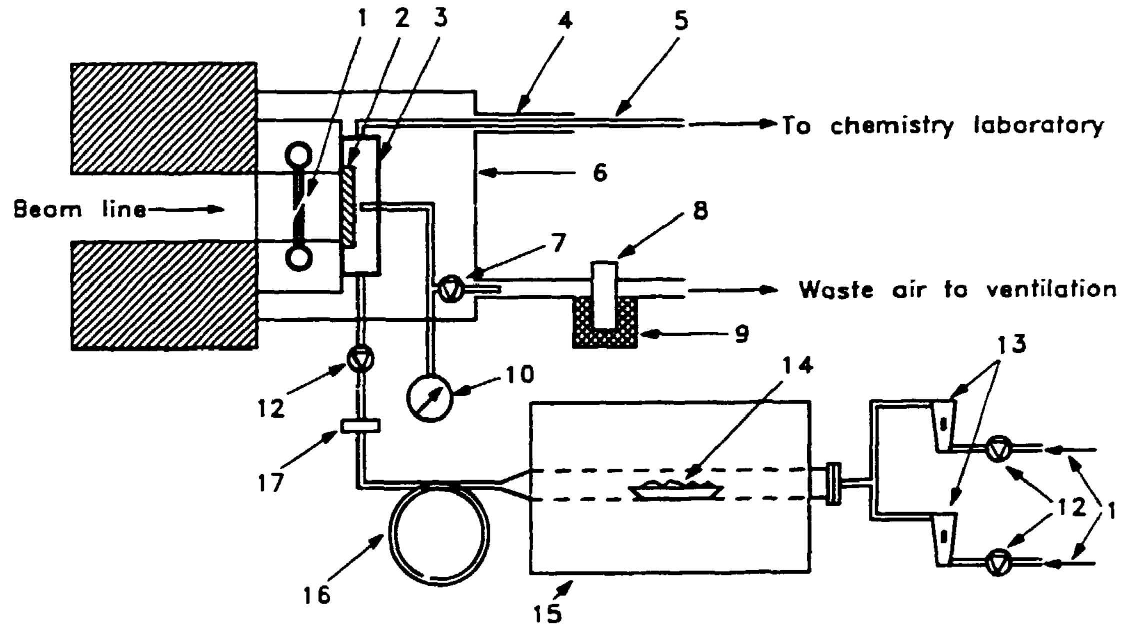

Fig. 8. Schematic view of the gas-jet facility at SAPHIR (from [8]; 1: chopper, 2: target, 3: target chamber, 4: outer tube, 5: capillary, 6: housing, 7: relief valve, 8: activity monitor, 9: charcoal trap, 10: pressure gauge, 11: gas supply, 12: solenoid valves, 13: flow controllers, 14: ceramic boat with KCl, 15: aerosol generator with tubular furnace, 16: polyethylene-tube loop, 17: glass filter).

Another facility available at SAPHIR for radiochemistry research is a gas-jet system, an arrangement often used for investigating the nuclear decay properties of short-lived nuclides. The experimental set-up at SAPHIR, however, has been specially designed for the investigation of adsorption/desorption processes for fission-product nuclides onto aerosol particles, as also for the development of fast chemical separation techniques for application in the field of superheavy element research [8]. Fig. 8 shows a schematic view of the facility, installed at the R0 radial beam port of the reactor (cf. Figs. 2, 3). Fission products, recoiling out of a 50 mm-diameter ^{235}U deposit (~1 mg/cm^2 thick) in the target chamber (thermal neutron flux ~5 x 10^6 cm^{-2}s^{-1}), are adsorbed onto KCl aerosol particles with He as the carrier gas – before being transported through ~130 m of polyethylene capillary tubing (2 mm i.d.) to the chemistry laboratory. The transfer time is about 12 s at a gas flow rate of 2 l/min. Some applications with very short-lived nuclides ($T_{1/2}$ ~1 s) are possible using a glove box installed ~10 m from the target chamber, i.e. in the reactor building itself. A feature of the SAPHIR gas-jet, which is a pre-requisite for kinetic studies, is the boral chopper (see Fig. 8), the two wings of which open and close within 25 ms.

6 Industrial Irradiations

6.1 Silicon Transmutation Doping

The demand for the neutron transmutation doping (NTD) of large-size silicon single crystals – for use in the industrial manufacture of various types of semiconductor devices – has grown markedly in recent years. The first such irradiations in the SAPHIR reactor were carried out as early as in 1975 [9], but the total annual quantities remained modest at less than 200 kg till about 1990. During the last 1-2 years, considerable effort has been invested to step up the silicon NTD activity at the reactor, and the current situation is that ~2 Te of silicon are irradiated annually. This more than ten-fold increase has been achieved within the constraint of not employing any additional personnel, the various manual handling operations involved (Fig. 9) being carried out by the reactor operational staff itself.

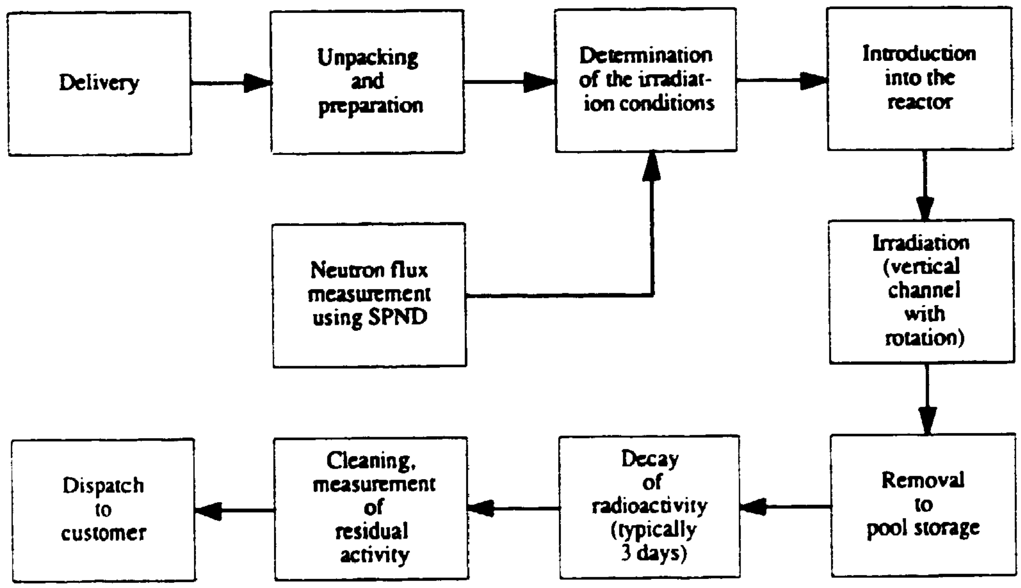

Fig. 9. Stages of the silicon transmutation doping procedure at SAPHIR.

Silicon ingots of 62.5 mm diameter and upto 300 mm length have been the type most commonly irradiated in the recent past, the use of core-edge positions (as indicated in Fig. 3) allowing loading/unloading operations to be carried out without shutting down the reactor. Measures implemented to achieve the desired accuracies on average target resistivity, as well as on axial and radial homogeneity, include: regular neutron flux measurements at the irradiation positions using dummy cylinders instrumented with SPND's, rotation of the ingot containers during irradiation and the insertion of short H_2O-displacing, aluminium cylinders above and below the silicon to flatten the neutron flux at the ends. Fig. 10 compares the axial neutron flux distribution measured in an "empty" (i.e. H_2O-filled) irradiation channel with that in a dummy cylinder, indicating thereby the flux-flattening effect of the ingot itself.

Techniques are currently being developed for the routine irradiation of larger (upto 100 mm-diameter) silicon ingots in SAPHIR. Careful optimization – in terms of high thermal neutron flux values and low radial/axial flux gradients – is necessary as the centre-line of the irradiation channel gets displaced further away from the core.

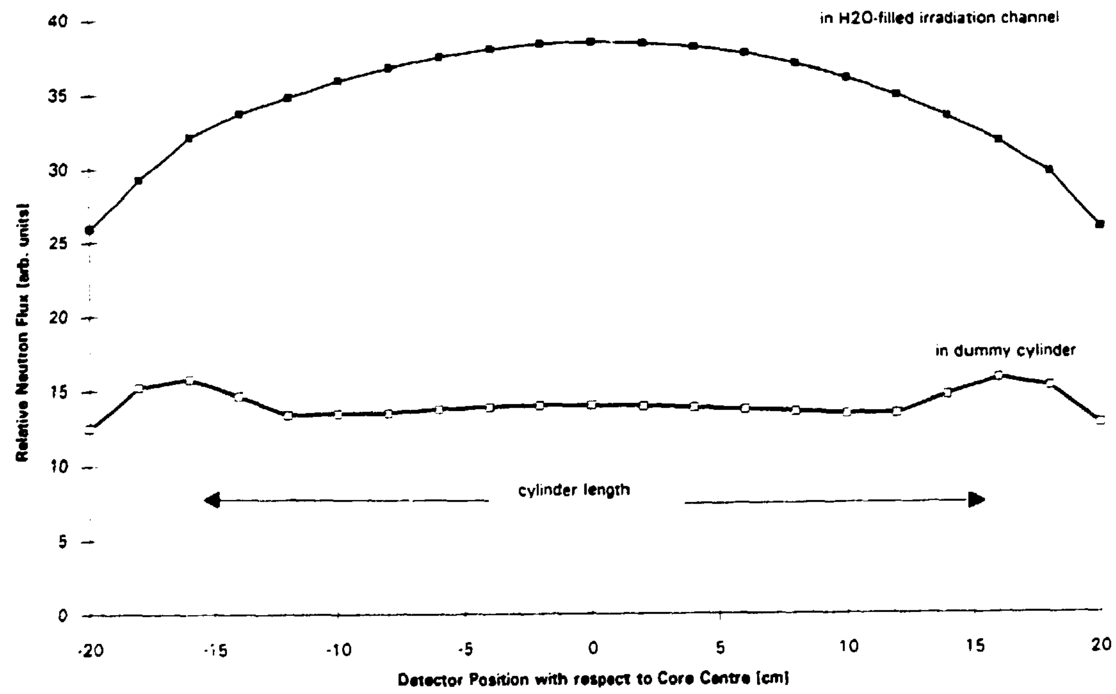

Fig. 10. Measured axial neutron flux distributions indicating the flux-flattening effect of the ingot itself during silicon transmutation doping irradiations in SAPHIR.

6.2 Epithermal Irradiations

For several industrial applications requiring epithermal and fast neutron irradiations, a particular problem is the minimization of residual radioactivity induced by thermal neutrons. A cylindrical rig, designed to occupy a single fuel-element position, is currently being tested for its effectiveness in such applications. Its main features are (a) the use of inserts with boron-carbide filled walls to reduce the incident thermal neutron flux and (b) dry conditions for the irradiations so that there is little thermalization of the transmitted fast neutrons.

6.3 Neutron Radiography

A neutron radiography facility, as indicated in Fig. 2, has recently been set up at the tangential beam tube port T1 of the SAPHIR reactor [10]. The facility has been designed within the constraints of the limited space available in the reactor building and is optimized with respect to the existing beam tube geometry.

A cylindrical graphite scatterer, ~300 mm long and 63 mm in diameter, is positioned in the tangential beam tube opposite the reactor core to serve as secondary neutron source. During full-power operation of the reactor, the temperature of the graphite cylinder is ~300 °C. A divergent collimator arrangement ensures a well-defined neutron beam geometry and has a 100 mm long bismuth single-crystal integrated into it. The latter, while causing ~80% suppression of the thermal neutron flux, reduces the γ-ray intensity by a factor of ~1000. This has made the application of direct imaging techniques with low γ-background effects possible and has, additionally, simplified the shielding requirements in the vicinity of the beam catcher considerably.

The primary beam-shutter function is provided by a collimator which extends to the edge of the reactor shielding and which can be filled with either water or helium. An evacuated multisection collimator, of total length 7.1 m, can be positioned in front of this – to yield a usable beam area of ~0.12 m^2 at its exit (12 m station, cf. Fig. 2).

Adjustment of the exposure time for the film, as also limitation of the exposure area (to minimize undesirable activation and scattering effects in the case of small objects) are achieved using a mechanical shutter device. Inactive objects of lateral dimensions upto ~1 m and thicknesses upto ~25 cm can be positioned together with a vacuum film cassette on a versatile object holder – an appropriately dimensioned, water-filled cylinder located behind this serving as beam catcher. The thermal neutron flux at the object position has been measured with gold foils to be ~3 x 10^5 $cm^{-2}s^{-1}$ with a Cd-ratio of ~20.

An alternative location for obtaining radiographic images is at the exit of the neutron beam from the reactor shielding (5 m station, cf. Fig. 2). A usable beam area of ~710 cm^2, with a thermal neutron flux value of ~2 x 10^6 $cm^{-2}s^{-1}$, is available here. With heavy shielding components (upto ~5 Te) provided around this location, it is possible to radiograph active objects such as segments of irradiated fuel rods and absorber plates. Transfer imaging and track-etch techniques can clearly be applied to advantage in such cases. The facility is being used in conjunction with transfer flasks and other equipment provided by PSI's hot laboratory, the current series of investigations being a co-operative effort to optimize the experimental techniques further.

7 Neutron Scattering

The Laboratory for Neutron Scattering (LNS) at PSI is run jointly in collaboration with the Swiss Federal Institute of Technology Zurich and has, over the years, developed an elaborate experimental infrastructure centred around the four spectrometers located at the SAPHIR radial beam tubes, R2-R5 (cf. Figs. 2, 3). Fig. 11 shows a view of part of the experimental area.

Fig. 11. A view of part of the experimental hall in SAPHIR showing spectrometers of the Laboratory for Neutron Scattering located at beam tubes R2-R4 (cf. Table 1).

With neutron scattering techniques representing a unique methodology for the investigation of structure and dynamics in condensed matter research, the experimental facilities at the reactor have been attracting users from a large number of both Swiss and foreign research groups covering a wide range of fundamental-research disciplines. High-temperature superconductors, materials research and magnetism are the three broad areas of investigation being pursued [11].

The radial beam tubes at SAPHIR, commencing from behind the beryllium reflector (Fig. 3), are helium-filled and provided with remotely controlled shutter plugs, as well as with liquid-nitrogen cooled silicon single-crystal filters for suppression of epithermal neutrons. The spectrometer shielding is designed such as to yield dose rates of <50 μSv/hr (n+γ) at all locations outside the line of the beam itself. It should be mentioned in passing that the tangential beam tube T6 – provided with a 3 x 5 cm^2 collimator – is often used for preliminary tests with new types of neutron scattering devices. (Education and training experiments are also conducted at T6 during low-power operation of the reactor (cf. Section 2)).

Table 1 gives some technical details for the four LNS spectrometers installed at SAPHIR – two 3-axis systems, a 2-axis diffractometer and a multicounter powder diffractometer [12]. Various auxiliary equipment is available for the conditioning of samples, e.g. a superconducting magnet for providing field strengths of upto 5 T, a He-3/4 dilution refrigerator for investigations at temperatures as low as 7 mK and furnaces for studies at upto 1100 °C. Even though, as mentioned earlier, the thermal neutron flux at the edge of the reactor core is slightly less than 1.0 x 10^{14} $cm^{-2}s^{-1}$ in SAPHIR (i.e. more than an order of magnitude less than at the high-flux ILL reactor at Grenoble, for example), the qualitiy of the LNS spectrometers and related instrumentation does provide optimal experimental conditions for a range of neutron scattering applications at thermal energies.

Table 1: Principal Technical Features of the LNS Neutron Spectrometers at SAPHIR (from [12]).

Type/notation (see Fig. 2)	3-Axis/D	2-Axis Diffr./C	Multicounter Power Diffr./B	3-Axis/A
Location (beam tube)	R2	R3	R4	R5
Incident wavelength range (Å)	0.9 to 4.4	0.8 to 4.1	fixed, 1.1/1.7	0.9 to 4.4
Max. flux at specimen ($cm^{-2}s^{-1}$)	10^6 (E=15meV)	10^6 (λ=1.05 Å)	10^6 (λ=2.3 Å)	10^6 (E=15meV)
Beam size at specimen (cm^2)	2.5 x 2.5	3 x 5	2 x 5	2.5 x 2.5
Collimation range	10′ to 60′	5′ to 60′	10′ to 12′	10′ to 60′
Monochromator angle range (2θ)	16° to 88°	14° to 90°	41° to 60°	16° to 88°
Scattering angle range (Φ)	-110° to 110°	–	–	-100° to 120°
Analyser angle range (2θ)	0° to 95°	0° to 120°/140°	3° to 140°	-110° to 110°
Other information	*a*), *b*)	*a*), *c*), *d*)	*a*), *e*)	*a*), *b*)

a) background <1 count/min.

b) variable horizontal curvature for monochromator and analyser systems

c) with tiltable detector (tilt angle range: -3 to 22°)

d) will also be used with polarized neutrons in the near future

e) resolution $\Delta d/d \geq 4\times10^{-3}$; can also be applied to the study of amorphous, liquid and quasicrystalline samples

8 Conclusions

It is seen that the 10 MW_{th} open-pool research reactor, SAPHIR, has – through the application of appropriate irradiation techniques – been meeting the requirements for various types of neutron irradiation experiments. At the present time, there is no other sufficiently intense neutron source in Switzerland which could replace the reactor in its multipurpose role.

As regards the outlook for longterm utilization of SAPHIR, an important factor is the planning and construction at PSI of a continuous spallation neutron source facility, SINQ, linked to the institute's large proton accelerator [13]. The new source – scheduled to be operational in 1995 – is projected to have major neutron scattering facilities (including a D_2 cold neutron source with a neutron guide hall) which will extend, both qualitatively and quantitatively, the fundamental-research possibilities at SAPHIR (cf. Section 7). The reactor, however, will clearly remain advantageous for a range of applications, e.g. those involving large-sample, high-fluence irradiations with easy access to regions of high neutron flux. It is particularly in such a context that the further development of irradiation techniques at SAPHIR will need to be continued.

Acknowledgements: The authors would like to express their sense of appreciation to the reactor operational team at SAPHIR, as also to the various researchers and experimentalists who have contributed to the development of the irradiation techniques described in this paper.

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