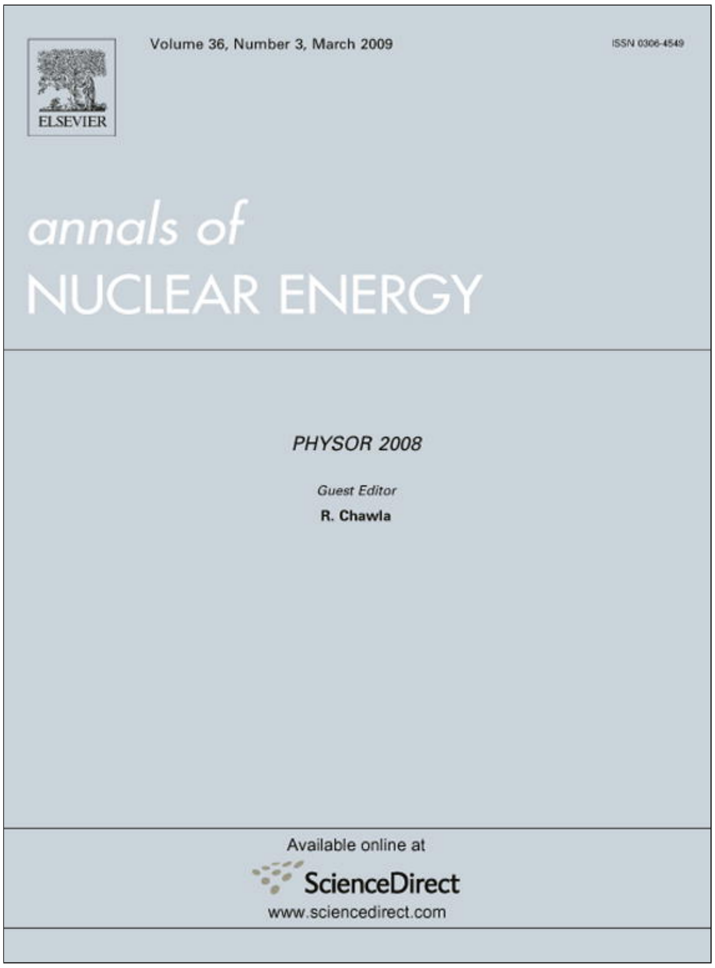
Volume 36, Number 3, March 2009
ISSN 0306-4549
ELSEVIER
annals of
NUCLEAR ENERGY
PHYSOR 2008
Guest Editor
R. Chawla
Available online at
ScienceDirect
www.sciencedirect.com

Annals of Nuclear Energy 36 (2009) 350–354

Contents lists available at ScienceDirect

Annals of Nuclear Energy

journal homepage: www.elsevier.com/locate/anucene

Neutronic characterization of the MEGAPIE target

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ARTICLE INFO

Article history:
Received 10 October 2008
Received in revised form 4 December 2008
Accepted 10 December 2008
Available online 6 February 2009

ABSTRACT

The MEGAPIE project aimed to design, build and operate a liquid metal spallation neutron target of about 1 MW beam power in the SINQ facility at the Paul Scherrer Institut (Villigen, Switzerland). This project is an important step in the roadmap towards the demonstration of the accelerator driven system (ADS) concept and high power liquid metal targets in general. Following the design phase, an experimental program was defined to provide a complete characterization of the facility by performing a "mapping" of the neutron flux at different points, from the center of the target to the beam lines. The neutronic performance of the target was studied using different experimental techniques with the goals of validating the Monte Carlo codes used in the design of the target; additionally, the performance was compared with the solid lead targets used before and after the MEGAPIE experiment.

1. Introduction

The MEGAwatt PIlot Experiment (MEGAPIE) project was started in 2000 to design, build and operate a liquid metal spallation neutron target of about 1 MW beam power (Bauer et al., 2001). The project is an important step in the roadmap towards the demonstration of the accelerator driven system (ADS) concept and high power liquid metal targets in general. The target was installed at the Paul Scherrer Institut (Villigen, Switzerland), and irradiated for four months in 2006. The target was inserted into the existing SINQ facility, replacing the former solid target. Following the design phase, an experimental program was defined to measure different neutronic and nuclear parameters. This is of interest both in the frame of ADS research, and for the SINQ users.

The neutronic performance of the target was studied with the goals of validating the Monte Carlo codes used in the design of the target; additionally, the performance was compared with the solid lead targets used before and after the MEGAPIE experiment. A detailed characterization of a compact molten metal target is clearly an asset in view of future target development, for applications in ADS or for neutron facilities.

1.1. Target description

The MEGAPIE target consists of a loop of lead–bismuth eutectic (LBE) inside a structure arranged vertically over a length of about 5 m.

The proton beam (575 MeV and 1.4 mA maximum current) penetrates the target from its bottom and the thermal energy deposited in the lower part of the target is removed by forced convection. The 82 l volume of LBE is driven by the main inline electromagnetic pump, then passes through a 12-pin heat exchanger and returns to the spallation region. The heat is evacuated from the heat exchanger through a diathermic oil loop to an external intermediate water cooling loop and then finally goes into the existing SINQ cooling system. The beam entrance window is cooled both by the main flow and also by a cold LBE jet extracted at the heat exchanger outlet, which is pumped by a second electromagnetic pump.

Since the goal of the SINQ facility is to provide thermal and cold neutrons to the users for various applications (mainly related to material science), the spallation target is inserted in a moderator tank of about 2 m diameter. A cold (25 K) liquid deuterium source provides cold neutrons to some of the beam lines. A system of inserts is placed in the moderator tank with the purpose to extract neutrons for the beam lines. The target-moderator system is

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doi:10.1016/j.anucene.2008.12.013

surrounded by a shielding block, consisting essentially of iron, with an external layer of borated concrete (Bauer et al., 2002).

1.2. Measurement program

The measurement program aimed at the characterization of the facility by performing a "mapping" of the neutron flux at different points, from the center of the target to the beam lines. For this purpose a measurement program was defined, using different experimental techniques depending on the neutron fluency varying from 10^8 n/cm^2/s to 10^{13} n/cm^2/s.

In particular, a set of measurements was performed during the startup phase, when the beam current was low. Part of these measurements could only be performed during this phase, as measurements with the nominal current would have led for instance to a saturation of the detectors. Other measurements were performed both in the startup phase and at nominal power to study the behavior of the facility in different irradiation conditions.

2. Target modeling

Monte Carlo calculations have been the essential tool during the neutronic design phase of MEGAPIE for various purposes, such as the design of the target and the assessment of safe operation. The bulk of the Monte Carlo calculations provided an overall characterization of the neutronic and nuclear behavior of the target prior to operation (Zanini et al., 2005). During the target irradiation, several neutronic and nuclear measurements were performed. In correspondence to each of these measurements, additional calculations have then been carried out to validate the employed codes. The codes used in the design phase were the comprehensive FLUKA 2006.3 (Fassò et al., 2005) and MCNPX 2.5.0 (Pelowitz, 2005) particle transport codes. The results presented in this paper are obtained with the MCNPX code with a statistical better than 1%.

Even though the various models used in the design phase were already quite detailed, a highly refined model of the SINQ facility and of the MEGAPIE target has been developed for a better comparison with the measurements. The main improvements consisted in modeling the SINQ facility around the target. An accurate geometrical description of the two latest employed solid targets and of MEGAPIE was performed. As the absolute neutron fluxes need to be calculated at the beam lines, at positions about 6 m away from the target center, the entire SINQ target block was modeled, and the beam lines collimators were included. A view of the geometry of the model with the MEGAPIE target is represented in Fig. 1.

The material definition is very important for flux calculations, as the presence of neutron capturing materials (such as the steel, or impurities in the LBE) may affect the target performances. Additionally, the presence of impurities may also affect the radionuclide inventory in the target. The actual composition of the steels (T91 and 316L) and of the $AlMg_3$ was used. A chemical analysis of a non irradiated sample of LBE (coming from the stock that was used in MEGAPIE) was performed, and a list of impurities extracted. Moreover, a precise model of the calculated two-dimensional beam profile was implemented in the MCNPX simulation.

MCNPX calculations have been performed for the three targets and at least 10^6 protons histories were run for each case. Neutron cross-sections up to 20 MeV were used and physics models were applied above this value.

For the simulations of the solid targets used in 2004–2005 (target 6) and in 2007 (target 7), the same model of the SINQ facility was used, where the solid rod targets replaced the MEGAPIE target.

The target 6 consists of a bundle of rods arranged in layers of 9 and 10 rods each, for a total of 37 rows and 351 rods. Each rod consists of a cylinder with 0.54 cm radius and 13.6 cm length.

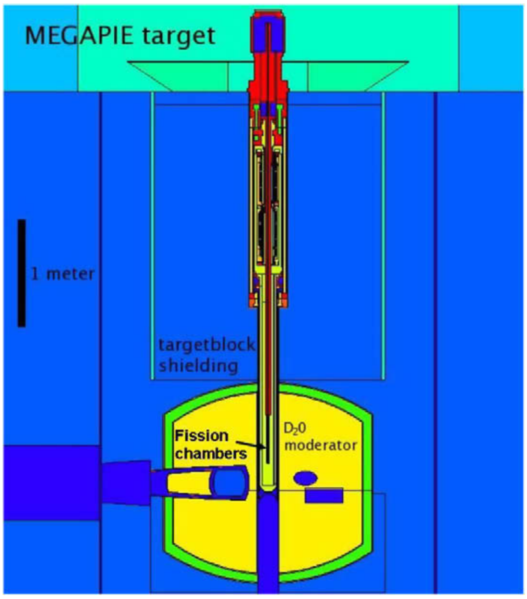

Fig. 1. MCNPX 2D visualization of the simulated system composed of the SINQ facility combined with the MEGAPIE target.

There are different types of rods:

the majority consists of Pb rods inside steel 316 L cladding, the volume of the cladding being filled with Pb to 90%;
the lowest row of nine rods consists of $AlMg_3$ cylinders which are open (therefore allowing the D_2O to circulate), for cooling purpose during operation;
for the rods at the sides of the bundle zircaloy replaces the steel;
several rods filled with specimens for the SINQ Target Irradiation Program (Dai et al., 2005) (mostly steel specimens) occupy some of the central positions in the target.

The target 7, which was inserted in the SINQ facility after the end of the MEGAPIE irradiation program, is very similar to target 6. The difference consists in the cladding (only zircaloy is used) and in a different arrangement of the STIP samples in the target.

3. Flux measurement inside the target

An innovative detector, based on micrometric fission chambers, has been designed and developed to measure the absolute neutron flux inside the MEGAPIE target with accuracy better than 5% and to follow the time and spatial evolutions of the flux over a large neutron intensity range.

The neutron detector, built in 2005, contains eight micrometric fission chambers adapted for the MEGAPIE specific environment, with dedicated cables, electronics and acquisition system. Fission chambers are embedded in pairs along the axis of the detector over a 50 cm length. Each pair (except one containing minor actinides) is made by a chamber containing ^{235}U fissile isotope and a chamber without deposit to compensate the fission signal from leakage currents or from currents induced by radiation fields. The bottom pair is shielded with natural metallic Gd filter (200 μm thickness) to absorb thermal neutrons and be more sensitive to epithermal neutrons. These different configurations are chosen to provide an overall characterization of the inside neutron flux, in terms of its intensity but also its energy distribution (Chabod et al., 2006a,b).

The current measured by fission chambers is proportional to the fission rate ($\sigma\varphi$) which depends on the neutron flux φ and the

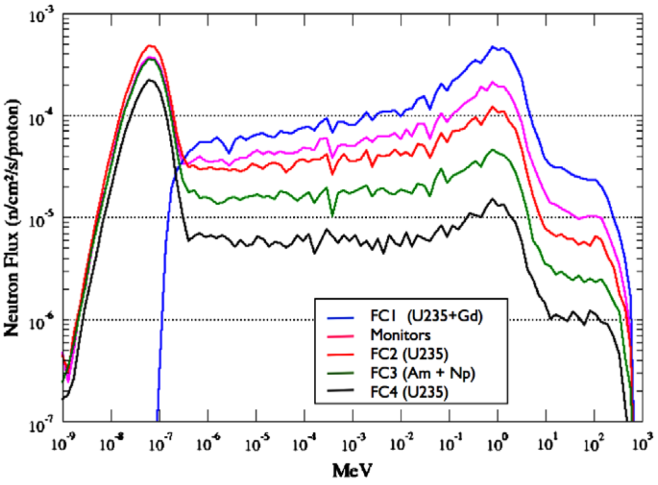

Fig. 2. Simulated neutron fluxes as "seen" by fission chambers (FC), along the beam axis. Minor actinide chambers are not discussed in this paper.

effective fission cross-section of the fissile isotope σ. The extraction of the neutron flux is then not straightforward and depends on a good characterization of the neutron energy distribution which is calculated using simulation codes (see Fig. 2).

To increase the accuracy on the energy spectrum determination, nine activation monitors were put inside the detector, between the first and the second chamber pairs, in a small stainless steel box (1.4 cm long and 6.5 mm in diameter). These nine monitors (Al–Co, Nb, Ni, Fe, Mn, Ti, Y, Rh, Gd) consist of ultrapure metallic disks or powder. They provide an integral measurement of the thermal, epithermal and fast neutron flux by capture and threshold reactions.

Since fission chambers are placed very close to the beam interaction point, they are very sensitive to the neutron flux distribution, both in position and energy. Thus we performed a deep study on the influence of the simulation parameters and physics models on the neutron flux properties. In particular, we studied the influence of the neutron detector geometry, the composition of the LBE, the beam profile and the different spallation models on the neutron flux (Panebianco et al., 2007). These studies showed for instance that the presence of some ppm of boron in the LBE affects the neutron flux mostly in the thermal region, as expected. In the lowest position, where the Gd shielding cuts away the thermal part of the spectrum, the effect of B is around 3% and does not exceed 20% when the boron concentration reaches a few per mil (which is far above the measured LBE composition). The largest influence (around 14%) on the simulated fission rate comes from the beam profile and it concerns mostly the lowest chambers, which are closer to the beam impact point. On the contrary, this effect does not exceed 4% for the upper chambers.

Taking into account all improvements and studies of the target system description and modeling, we could compare the measured fission rate to the simulated one (Table 1). The fission rate normalization is actually extracted from the measured fission current taking into account the chamber sensitivity which has been previously measured at the ILL reactor (Chabod et al., 2006a,b) with a precision

Table 1
Comparison of simulated and measured fission rates ($\sigma\varphi y$) (s^{-1} mA^{-1}) for different fission chambers. The first position is the lowest one (Gd shielded).

Position	1	2	5
Experim.	3.70×10^{-10} (3%)	2.85×10^{-9} (3%)	1.19×10^{-9} (3%)
C/E	1.80	2.98	2.78

Table 2
Comparison between measured and calculated reaction rates per atom (s^{-1} mA^{-1}).

Reaction	Experim.	C/E (Proc. 1)	C/E (Proc. 2)
$^{59}Co(n,\gamma)^{60}Co$	2.16×10^{-10}	1.02	1.92
$^{58}Ni(n,p)^{58}Co$	3.50×10^{-13}	0.95	2.09

of 3%. From this comparison we can see that there is a systematic over prediction of the measured values by a factor of 2–3.

In order to investigate this discrepancy we measured the activities from two flux monitors (Al–Co and Ni) by γ-spectroscopy with a high purity Ge detector. Data have been analyzed following two procedures. The first consists in calculating the reaction rate by using a mean flux value deduced from the fission chambers measurements, under the hypothesis that the mean cross-sections given by MCNPX calculations are correct. The second procedure is to get the total neutron flux value simulated with MCNPX and to perform the reaction rate evolution with the CINDER code (Wilson et al., 2001). Table 2 presents the comparison between the measured reaction rates and the calculated ones (using the two procedures) for the two reactions of interest: $^{59}Co(n,\gamma)^{60}Co$ (in the case of the Al–Co monitor) and $^{58}Ni(n,p)^{58}Co$ (in the case of the Ni monitor).

From the results presented one can see that the activation foil measurements are in much better agreement with the calculations performed starting from the flux measured by fission chambers than with the MCNPX–CINDER calculation. This proves the consistency of the two measurements. Monitors results confirm the discrepancy presented above between measurements and MCNPX calculations. We can go further in the analysis considering that the two gamma emitters are concerned by different energy ranges (essentially thermal for ^{60}Co, between 3 and 15 MeV for ^{58}Co). Further investigations on all the simulation details are underway.

4. Flux measurement outside the target

Besides the innovative measurements in the central rod, a large set of flux measurements have been performed in several points of the SINQ facility. Part of these measurements were already performed in previous years, prior to MEGAPIE operation, and repeated in 2007 with the new solid target. There is therefore the possibility to compare the performances of MEGAPIE with the conventional solid targets. Such information is of great interest for SINQ users, in view of their measurement programs and more generally for the target development program.

Activation foils for thermal and epithermal neutron determination were irradiated at three beam lines of the SINQ facility: ICON, NEUTRA and EIGER; additionally, measurements were performed at the NAA irradiation station located inside the moderator tank (see Fig. 3). The integral fluxes vary from about 10^{13} n/cm^2/s/mA at the NAA to about 10^8 n/cm^2/s/mA at the beam lines. All the results will be shown in the next section in comparison with simulated values.

4.1. NAA

The Neutron Activation Analysis (NAA) station is located inside the heavy water tank, at about 80 cm from the center of the SINQ target. At the NAA station, samples are placed inside polyethylene capsules, and sent to two irradiation positions by means of a pneumatic system. Gold and cobalt foils were irradiated to measure the thermal neutron flux. Measurements were performed in 2006 (MEGAPIE target) and 2007 (SINQ solid target). Reaction rates are obtained combining the average thermal cross-section deduced from simulation, with the activity extracted from γ-spectroscopy measurements performed using an HPGe detector. Additionally, a

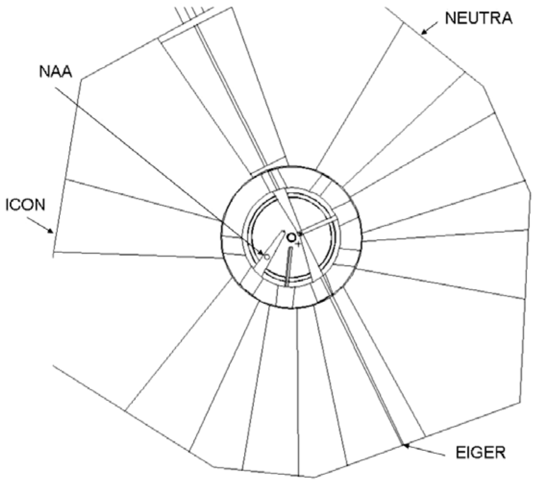

Fig. 3. Measurement positions for ICON, EIGER and NEUTRA beam lines, and for the NAA station.

set of threshold detectors (foils of Al, Ti, Mn, Fe, Ni and Cu) was irradiated to measure reaction rates induced by neutrons with energy greater than 1 MeV.

4.2. Beam lines

Several measurements were performed at ICON, NEUTRA and EIGER beam lines, right at the end of the SINQ target block (see Fig. 3). Measurements at the beam lines were performed with gold foils, with and without Cd cover (except for the NEUTRA measurement) in order to correct for the epithermal neutron component.

ICON is one of the neutron beam lines with direct view to the cold D_2 moderator. NEUTRA and EIGER are thermal beam lines.

The EIGER beam line has several special characteristics: the beam line looks on the water scatterer which is close to the target; the collimation is coated by supermirrors; a single sapphire crystal is placed in the beam line to reduce the epithermal component by around 90%. To observe the expected vertical neutron flux distribution and reduce the systematic uncertainties several gold foils were positioned along the vertical beam center.

4.3. Systematic corrections and uncertainties

There are a number of effects which might affect systematically the measurement of the neutron flux by foil activation. The most important are self shielding and neutron scattering. Other effects such as foil perturbation are not considered here.

Self shielding corrections are very different for thermal and resonance neutrons. For thermal neutrons, a systematic correction of 2% for NEUTRA and EIGER has been applied, in agreement with the attenuation law for thermal neutrons for foils of 20 μm thickness. A correction of 4% was applied for ICON (because of the lower energy of the cold neutrons) and NAA (which is inside the heavy water tank and therefore foils are crossed by a partially isotropic flux).

For epithermal neutrons the self shielding correction can be very large. Self shielding factors coming from epithermal neutron resonances have been measured and calculated in several works (see for instance Goncalves et al., 2002). An epithermal self-shielding factor of about 0.4–0.6 is expected for a gold foil of 20 μm thickness. A self shielding factor of 0.57 has been used for ICON, EIGER and NEUTRA (collimated beam), and 0.49 for the NAA (again because of a partially isotropic flux).

Systematic uncertainties on the measurements were estimated separately for each measurement point; uncertainties are indicated in the table of the results.

5. Results for outer flux measurements

5.1. Thermal neutron flux

Results concerning integrated thermal neutron measurements are shown in Table 3 for the target 6, MEGAPIE and target 7, respectively. MCNPX values represent the integrated fluxes up to 1 eV neutron energy, while experimental values come from the measurements, where the contribution from epithermal neutrons is subtracted, and the flux is obtained by dividing for the effective cross-section between 0 and 1 eV. These values are therefore directly comparable.

On average, MEGAPIE has a neutronic performance of a factor 1.74 ± 0.12 (calculated value 1.69) higher than target 6 and of a factor of 1.62 ± 0.08 (calculated value 1.54) with respect to target 7.

5.2. Epithermal neutron flux

Indicative values on the epithermal fluxes can be extracted from the activation measurements where the gold foils were enclosed in a Cd capsule. Gold has a strong resonance at 4.9 eV with a capture area of 1551 barn, corresponding to 99.7% of the resonance integral. It is however not possible to obtain a pure flux measurement at the resonance without using the double foil technique, and therefore these values are only indicative. The values in Table 3 represent the epithermal fluxes per logarithmic energy unit. To obtain the integral fluxes these values should be multiplied by $ln(E_{MAX}/E_{MIN})$ (under the assumption that the epithermal flux in lethargy units is constant), which gives a factor of 13.8 using E_{MIN} = 1 eV and E_{MAX} = 1 MeV.

The measured epithermal integral fluxes at NAA are about 4% of the total for target 7, and 6% for MEGAPIE. For ICON, the epithermal fraction at MEGAPIE seems higher, about 23%, as compared to the solid targets (around 14%), which would imply a higher background with the MEGAPIE target at the cold moderator beam lines. As expected, the calculated epithermal fraction at ICON is 10 times higher than measured, due to the effect of the sapphire crystal which has not been modeled.

5.3. Fast neutron flux

For the fast neutrons, good indications are obtained by reaction rate measurements with threshold detectors irradiated at NAA. Some relevant results are reported in Table 4. We can compare

Table 3
Summary of experimental and calculated integral thermal flux (cm^{-2} s^{-1} mA^{-1}) and epithermal flux results (flux per logarithmic energy unit).

	Thermal		Epithermal	
	Experim.	C/E	Experim.	C/E
Target 6				
ICON	4.19 × 108 (10)	1.17	5.70 × 106	1.18
EIGER	6.73 × 108 (7)	1.19	1.22 × 106	0.98
NEUTRA	2.47 × 107 (10)	0.97	–	–
MEGAPIE				
ICON	7.45 × 108 (10)	1.08	1.62 × 107	0.80
EIGER	1.14 × 109 (7)	1.23	2.61 × 107	9.5
NEUTRA	4.48 × 107 (10)	0.84	–	–
NAA	1.04 × 1013 (5)	1.25	4.43 × 1010	1.07
Target 7				
ICON	4.61 × 108 (10)	1.18	4.53 × 106	1.45
EIGER	7.45 × 108 (7)	1.16	–	–
NEUTRA	2.83 × 107 (10)	0.95	–	–
NAA	6.20 × 1012 (5)	1.29	1.97 × 1010	1.32

Table 4
Comparison between measured and calculated reaction rates per atom (s^{-1} mA^{-1}) for MEGAPIE and target 7. Values in brackets are uncertainties in%.

Nuclide	MEGAPIE		Target 7	
	Experim.	C/E	Experim.	C/E
Mn sample (89.8% Mn, 10.1% Ni)				
^{52}Mn	$1.99 \times 10^{-16\ (0.6)}$	0.93	1.85×10^{-16} (1.4)	0.93
^{54}Mn	6.60×10^{-15} (0.8)	1.30	5.29×10^{-15} (0.7)	1.19
^{51}Cr	4.91×10^{-16} (3.0)	2.20	4.51×10^{-16} (3.1)	2.00
Ni sample				
^{48}V	–	–	2.21×10^{-17} (0.6)	1.72
^{51}Cr	–	–	1.79×10^{-16} (2.8)	1.45
^{52}Mn	1.16×10^{-16} (1.0)	1.66	1.08×10^{-16} (0.6)	1.61
^{54}Mn	–	–	3.64×10^{-16} (0.8)	0.95
^{56}Co	1.04×10^{-15} (1.9)	2.38	9.62×10^{-16} (0.4)	2.30
^{57}Co	4.48×10^{-15} (3.3)	1.42	5.63×10^{-15} (3.0)	0.86
^{58}Co	6.61×10^{-15} (0.8)	1.42	5.55×10^{-15} (0.8)	1.26
^{56}Ni	2.07×10^{-17} (3.5)	7.05	2.26×10^{-17} (2.2)	6.02
Al sample (99.77% Al, 0.23% Fe)				
^{22}Na	1.77×10^{-16} (5.0)	1.18	–	–
^{24}Na	9.58×10^{-16} (1.3)	1.18	7.69×10^{-16} (0.5)	1.14
Ti sample				
^{44}Sc	2.57×10^{-16} (1.2)	1.30	$2.09\ 10^{-16}$ (0.4)	1.42
^{44m}Sc'	1.36×10^{-16} (2.5)	1.81	1.09×10^{-16} (2.3)	1.89
^{46}Sc	1.76×10^{-15} (2.3)	0.91	1.36×10^{-15} (0.5)	1.02
^{47}Sc	2.00×10^{-15} (1.8)	1.03	1.53×10^{-15} (0.5)	1.13
^{48}Sc	$5.85\ 10^{-16}$ (0.3)	1.53	4.32×10^{-16} (0.3)	1.69
Cu sample				
^{52}Mn	4.83×10^{-18} (0.8)	8.63	5.82×10^{-18} (1.9)	6.82
^{54}Mn	5.26×10^{-17} (2.4)	1.53	5.19×10^{-17} (0.2)	1.33
^{57}Co	2.26×10^{-16} (3.4)	1.84	2.17×10^{-16} (3.8)	1.70
^{58}Co	5.47×10^{-16} (0.9)	1.05	4.74×10^{-16} (0.9)	1.09
^{60}Co	5.54×10^{-16} (1.1)	0.73	4.81×10^{-16} (1.3)	0.69
^{59}Fe	3.72×10^{-17} (1.7)	0.32	3.19×10^{-17} (2.1)	0.33
Fe sample				
^{51}Cr	4.03×10^{-16} (9.0)	1.47	5.61×10^{-16} (2.6)	0.96
^{52}Mn	9.84×10^{-17} (1.0)	2.56	1.57×10^{-16} (0.5)	1.44
^{54}Mn	1.47×10^{-15} (2.7)	1.37	2.10×10^{-15} (0.7)	0.83

the reaction rates measured with the different targets to have an idea of the changes of the fast neutron flux at the NAA position. From the results in Table 4 we see that the reaction rates obtained in MEGAPIE and target 6 are on average very close.

These results are confirmed by the calculations. The calculation of reaction rates has been performed using the SNT code (Konobeyev et al., 2002) and the neutron spectrum calculated with the MCNPX code. It has been found that in most of the cases neutrons with energy from 20 to 150 MeV are the main contributors in the radionuclide production for all considered materials. The reactions responsible for the formations of the nuclides are (*n,xnp*) inelastic reactions. A given nuclide can be formed either by a direct reaction but also by the formation of parent nuclides and subsequent beta decay. In the latter case the cumulative cross-sections have also been considered. Direct cross-sections coincide with the cumulative ones in most of the cases, with the exception of a few isotopes where the parent contribution to the cumulative cross-section is about 10%.

In summary, calculations and measurements indicate a fast neutron flux in MEGAPIE only slightly higher than with target 7, contrary to the findings for the thermal and epithermal components. This interesting result comes probably from the different structures of the two spallation targets and from the influence of the heavy water inside the solid targets on the fast spectrum at the measured position.

6. Discussion and conclusions

A large data set was collected in the SINQ facility using the first high-power liquid metal target developed in the world, and complemented with corresponding measurements with the solid targets, and with detailed Monte Carlo calculations. The combination of these results provides very interesting information for the development of liquid metal targets and for the ADS.

1. Absolute flux measurements at the exit of the SINQ target block compare well (within 20% for thermal fluxes) with calculations, indicating that the neutron production and thermalization in the SINQ facility is correctly reproduced.
2. The MEGAPIE target is considerably more performing than the solid ones, with an average increase of the thermal fluxes to the users of a factor 1.74 (compared with the solid target 6) and 1.62 (compared with target 7). This result is also confirmed by calculations.
3. The flux measurements inside the MEGAPIE target, in the central rod close to the beam interaction zone, are a factor 2–3 lower than the calculations. It is important to note that the measurements with the innovative fission chambers are consistent with the results from the monitors placed in the same position. This result is very interesting, also because the measurement at the beam lines (point 1) indicate that the overall neutron production seems to be correctly modeled by MCNPX). These measurements were taken in a region where the neutron flux has a high gradient, it has strong thermal, epithermal and fast components, which all play a role in the average reaction cross-sections, and it is influenced by the presence of structural materials. All these factors compete in making this a very challenging measurement. Despite the vast effort also from the simulation side, the reasons for the disagreement are not yet fully understood and investigation continues.

Acknowledgements

The authors would like to acknowledge the MEGAPIE initiative and all financing institutions. This work is also supported by the European Community under the IP-EUROTRANS contract N FI6W-CT-2004-516520. We are grateful to PSI personnel for the great support given during installation, irradiation and post-test analysis.

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