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Measurement of Volatile Radionuclides Production and Release Yields followed by a Post-Irradiation Analysis of a Pb/Bi Filled Ta Target at ISOLDE

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A crucial requirement in the development of liquid-metal spallation neutron target is knowledge of the composition and amount of volatile radionuclides that are released from the target during operation. It is also important to know the total amount produced, which could be released if there was an accident. One type is the lead-bismuth eutectic (LBE) target where different radionuclides can be produced following interaction with a high-energy proton beam, notably noble gases (Ar, Kr, Xe isotopes) and other relative volatile isotopes such as Hg and At. The results of an irradiation experiment performed at ISOLDE on a LBE target are compared with predictions from the MCNPX code using the latest developments on the Liège Intranuclear Cascade model (INCL4.6) and the CEM03 model. The calculations are able to reproduce the mass distribution of the radioisotopes produced, including the At production, where there is a significant contribution from secondary reactions. Subsequently, a post-irradiation examination of the irradiated target was performed. Investigations of both the tantalum target structure, in particular the beam window, and the lead-bismuth eutectic were performed using several experimental techniques. No sign of severe irradiation damage, previously observed in other ISOLDE targets, was found.

I. INTRODUCTION

Several high-power liquid metal spallation neutron sources are currently operating or under development. The Spallation Neutron Source (SNS) in Oak Ridge, USA, and the neutron source at the Japan Proton Accelerator Research Complex (J-PARC) in Tokai, Japan, are dedicated to condensed matter studies. In both cases the spallation material is mercury. The Multi-Purpose Hybrid Research Reactor for High-Tech Applications (MYRRHA) will use lead-bismuth eutectic (LBE) for neutron production in a subcritical core. Lead-bismuth eutectic (LBE) is one of the candidate materials for the spallation target of an accelerator-driven system (ADS). The MEGAwatt PIlot Experiment (MEGAPIE [1]) was the first high-power liquid metal target, which was irradiated at nearly 1 MW power in 2006 at the Paul Scherrer Institut.

In licensing and safety studies of such complex systems, production and release of volatile products (often radioactive) are in need to be addressed. In this paper we report the final experimental results from a LBE target irradiated at CERN-ISOLDE. An experiment such as that performed at ISOLDE can provide valuable information on both aspects. For the production of radionuclides, Monte Carlo codes to calculate the production yields can be benchmarked. The use of an extended target enables the contribution of secondary reactions to the total production yield of a given isotope to be studied. The diffusion process for all the elements that are not immediately released must be taken into account for a proper comparison with Monte Carlo predictions. Information on the diffusion time in such a target, and in full scale experiments such as MEGAPIE, at different operating temperatures, can tell us how fast volatiles diffuse out of liquid metal targets at different temperatures.

Additional valuable information can be obtained from the investigation of the target after irradiation, to study radiation damage effects on the target structure and to perform additional analysis on the spallation material.

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The target structure and the LBE were investigated after irradiation. This included the analysis of possible radiation-induced damage (pitting effects, cracks) or corrosion effects in the target structure and spectroscopic studies of LBE samples after irradiation. We report briefly on the main findings, which have been described in other papers[2, 3].

II. IRRADIATION

A. Measurements

The experiment is described in Ref. [4]. The target consisted of a Ta cylinder, 20 cm long, with radius of 1 cm, filled with 547 g of LBE. Isotopes released from the target were subsequently ionized, accelerated to 60 keV, mass separated by a magnetic dipole and sent to the experimental area, where yields were measured. The target was operated at various temperatures up to 600 °C. The proton beam energy was of 1 or 1.4 GeV, with pulses of various intensities, typically 10^{13} protons/pulse.

All the isotopes released from the target at temperatures up to 600 °C were investigated, and release yields, i.e. the product of thick target yield and released fraction were measured for most of them, based on a comprehensive study of the efficiency of the apparatus. Measurements were performed off-line, by means of gamma spectroscopy using an HPGe detector of Al foils into which the ionized beam had been implanted, and on-line using another HPGe detector and a plastic scintillator measuring beams deposited on a tape. However, since it was difficult to determine the absolute efficiency of the plastic scintillator, only data from the HPGe detectors were used for absolute release yield measurements.

B. Experimental Results and Comparison with Calculations

Several elements were observed in the experiment; quantitative measurements of Ne, Ar, Kr, Xe, Hg and At isotopes were performed. Some release of Cd and I isotopes was also observed. Results for Ar, Kr, Xe, Hg and At isotopes at proton beam energies of 1 GeV and 1.4 GeV are shown in Figs. 1 - 5. The uncertainties in the data points are statistical uncertainties based on the analysis of the HPGe spectra.

The release yields exhibit rather large variations due to different factors, in particular contributions from the decay of the progenitors, the inherent complexity of the release process and uncertainties in the diffusion times of the isotopes. Release times of noble gases (the time of the release of half the produced quantity) are between 0.5 (He) and 3 (Xe) minutes at the maximum temperature, and of the order of 10 minutes for mercury. This information was obtained from measurements done with a few proton pulses on target, followed by a measurement of the current of the released (ionized) isotopes with Faraday cups up to about 30 minutes. The diffusion times for other isotopes, such as astatine, were deduced a posteriori by comparison with Monte Carlo simulations.

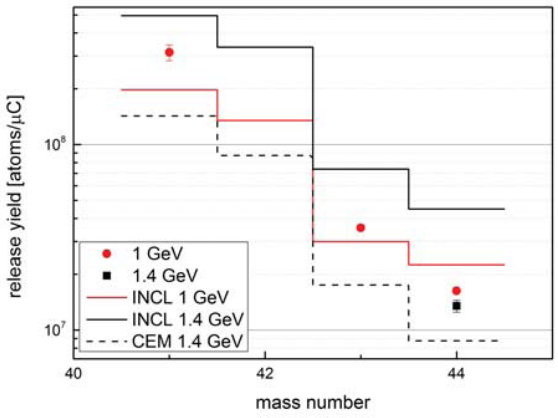

FIG. 1. Measured release yields of Ar isotopes (in atoms for a proton charge of 1 μC on target) for 1 and 1.4 GeV protons (red and black dots, respectively). Calculations using MCNPX using the INCL4.6[5]-Abla07[6] model combination, for 1 and 1.4 GeV proton energy, and the CEM03 model, for 1 GeV proton energy, are also shown. In the calculations instantaneous release was assumed.

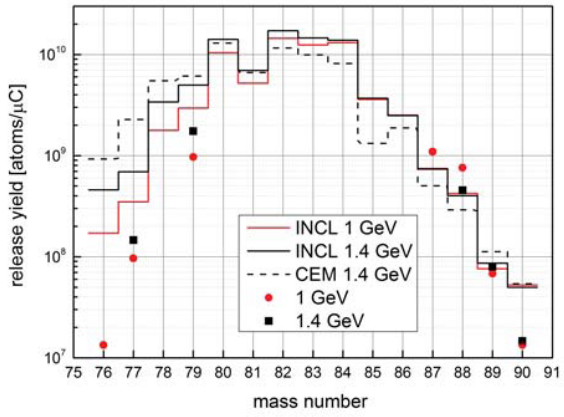

FIG. 2. Like Fig. 1 for Kr isotopes.

Systematic uncertainties in the ion source efficiency and in the transmission efficiency were estimated to be about 20%. Many data points were obtained by averaging results from different measurements. By averaging over several measurements, confidence in the accuracy of the experimental results was gained. However, since the contribution of decay of progenitors is significant for several volatile isotopes, some variation in the total yields

can be expected if the amount of the progenitors had not reached saturation, as was the case for long-lived progenitors. In some cases, spectra were measured at different times using the same technique (off-line collection measurement or on-line measurement at the tape station). In other cases measurement of the same isotope was performed both off-line and on-line. In most cases the results from different measurements were within 20-30%, i.e., within the other systematic uncertainties. However, in some cases the results differed by a factor of 2 or more, possibly because of different contributions from parent decay. In those cases the uncertainty in the results is higher, as shown for some data points in the plots.

Earlier comparison with Monte Carlo calculations showed good agreement for most of the isotopes studied [4]. One exception were At isotopes, where none of the model combinations or codes used could reproduce the experimental results. Recent developments in the INCL4 Intranuclear Cascade Model [5] have shown that astatine production can be successfully modeled if the pion exchange reactions and secondary reactions induced by ^{3,4}He are taken into account. Additionally, the diffusion time inside the bulk material must be considered for a precise comparison between data and calculations. Information on the diffusion time was only available for mercury (a diffusion time of about 10 minutes was measured at 650 °C). For astatine, a diffusion time of about 10 hours must be assumed for a proper comparison with the experimental data. Results of MCNPX[7] calculations for astatine have recently been published [8]. Using the same INCL4.6 model, implemented into a private version of MCNPX2.7.b, coupled to the latest version of Abla07[6], we calculated the production of the other isotopes measured at ISOLDE. Results for Ar, Kr, Xe, Hg and At isotopes are shown in Figs. 1 to 5 together with the data. The contribution of the progenitors to the radioisotope production was taken into account. We assumed saturation activity for the progenitors, and the contribution of all the relevant progenitors was added. As discussed above, this was not always the case, and an overestimation of the calculations can be expected in some cases. Calculations were also performed with the CEM03 model[9].

Overall, for Ar, Xe and Hg, agreement between data and calculations with all the models can be considered satisfactory. For Kr there are too few data points (placed in the tails of the mass distribution) to draw conclusions. For At, clearly the INCL4.6 calculations give better results, in light of what was discussed above.

In addition to the isotopes shown, release yields of ^{23,24}Ne were measured giving for both isotopes values in the range of 1.5 $\times 10^7$ atoms/μC. For these two isotopes, the CEM03 calculations are in good agreement with the data, while the INCL4.6 results are about 10 times higher. Unfortunately data for these isotopes were scarce, therefore it is difficult to make a firm conclusion about the validity of the codes in this case; during the "Benchmark of spallation models" [10] calculation results

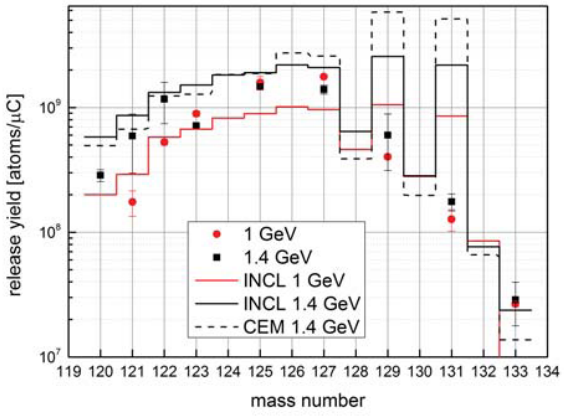

FIG. 3. Like Fig. 1 for Xe isotopes. For the ^{125}Xe, ^{127}Xe and ^{133}Xe data points the sum of ground and metastable states is shown. For ^{129}Xe and ^{131}Xe only the metastable states was measured, which explains the discrepancy with respect to the simulated value that represents the sum of metastable and ground states.

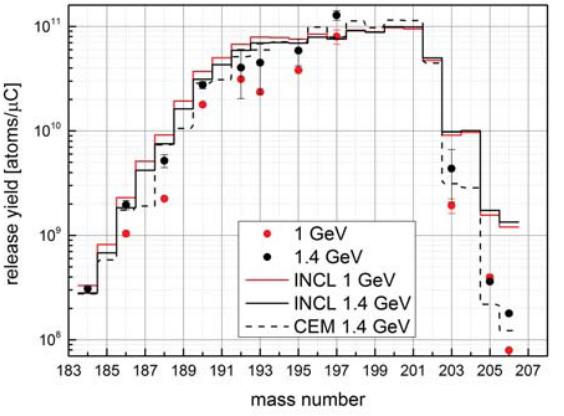

FIG. 4. Like Fig. 1 for Hg isotopes. For the ^{193}Hg, ^{195}Hg and ^{197}Hg data points the sum of ground and metastable states is shown. In the calculations diffusion time of 10 minutes was assumed.

of the CEM03 and INCL4.5-Abla07 models (INCL4.5 being close to from 4.6 in this case) were compared to experimental production cross sections of ^{21}Ne and ^{22}Ne [11] giving a good agreement. Nevertheless the present finding should be further explored, if future data will be available.

Other isotopes measured were ^{107,111m}Cd and several iodine isotopes. Comparison with calculations were not performed with the latest code models. Results for iodine were already presented in Ref.[4], showing predicted values two order of magnitude higher than measured data, thus indicating a very slow release for these isotopes.

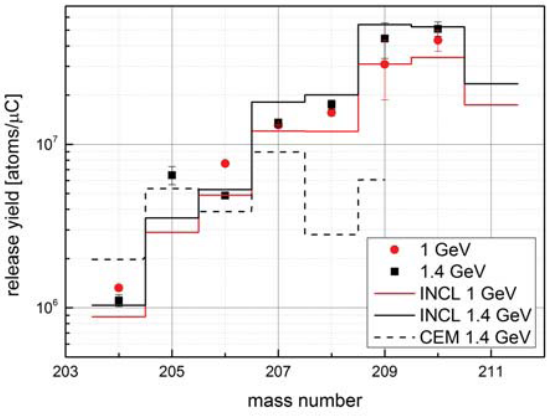

FIG. 5. Like Fig. 1 for At isotopes. In the calculations diffusion time of 10 minutes was assumed.

III. POST-IRRADIATION

A few years after irradiation at ISOLDE, the target was transported to the Hot Lab at the Paul Scherrer Institut for a post-irradiation analysis of the Ta container and the LBE material. Results from the post-irradiation analysis were reported in Ref. [2]. The LBE was analyzed by means of alpha, gamma, and mass spectroscopy[3]. The polonium content was measured and found to be in good agreement with predictions from calculations.

The Ta target was analyzed by visual inspection, and by scanning electron microscope (SEM) and energy-dispersive X-ray spectroscopy (EDX) analysis of samples prepared with a focused ion beam (FIB). The results indicated only minor radiation damage, much lower than in targets previously irradiated at ISOLDE. Cavitation-induced pitting and thermal-stress-induced cracks have been confirmed in previous work to be the origin of liquid metal target containment failures at ISOLDE in the 1990s. Mitigation of these effects was addressed by lengthening the proton beam pulse from sub-μs to 16 μs, defocusing the beam and hence reducing the beam energy density, and modifying the geometry of the target container. Therefore, none of the severe damage effects seen in the previous targets were observed. Full results are discussed in Ref. [2].

IV. FEEDBACK TO ADS AND CONCLUSIONS

The experimental program at the ISOLDE LBE target started as part of the MEGAPIE project. Similar measurements of gas emission were performed during the irradiation of MEGAPIE in 2006. Results from calculations and measurements in MEGAPIE are reported in Refs. [12–14]. The irradiation temperature in MEGAPIE was much lower than that of the ISOLDE target, with a maximum temperature of about 400 °C in the hot spot, but below 300 °C in the free surface between the target and the cover gas system.

The combination of volatile element measurements from the ISOLDE and MEGAPIE experiments has provided important information on the gas release process in an ADS target. Noble gas release is a slow process in a large scale liquid metal spallation target; the release yields of noble gases in the MEGAPIE system are at the 1% level after 1−2 days of operation, and the release in the cover gas reaches saturation a few weeks after the beginning of operation. The mass distributions of the released nuclides are reproduced well by calculations. The diffusion of mercury is slower than the noble gas release, and the diffusion of astatine (which decays into polonium) is much slower.

In summary, we have shown how a full investigtion of a small-size LBE target from irradiation to post-irradiation analysis can provide very important data, which are relevant to the development of high-power neutron producing targets. Experimental determination of production and release of radioisotopes is critical for the safety of liquid metal spallation targets. The post-irradiation analysis provided useful information about the radiation damage of structural components.

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