THE VACUUM INTERFACE COMPATIBILITY EXPERIMENT (VICE) SUPPORTING THE MYRRHA WINDOWLESS TARGET DESIGN

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Abstract

The MYRRHA accelerator-driven system (ADS) is being developed in Mol, Belgium incorporating a windowless spallation target, as this is the only option within the constraints of the envisaged neutron source performance. In this paper the research programme and experimental set-up required for the R&D effort towards the conditioning of Pb-Bi eutectic for use in a *windowless* liquid-metal spallation target are presented. The experiences acquired commissioning the set-up, the current status of the experiment and first results are discussed.

Introduction

Lead-bismuth eutectic (LBE) has interesting properties as a target material for high-power spallation sources. It is sufficiently heavy to provide an adequate yield in the spallation reaction, rather transparent for fast neutrons and has a quite low melting point of LBE (123 C) so the forced convection method to remove heat deposited in the target by the proton beam by circulating the liquid LBE can be used at modest temperatures. However, "off the shelf" LBE is not necessarily suited for use in a spallation loop because possible insoluble contaminations might accumulate at particular positions in the loop (e.g. at ridges), thus giving rise to an increased pressure drop, diminished flow and possibly complete blocking. Thus, a cleaning procedure is required. In addition, LBE is quite corrosive as regards stainless steel due to the high solubility of Fe and Ni. The process can be kept under control by establishing a dynamic equilibrium between the oxygen in the LBE and that on the surface of the vessel. This implies a control of the oxygen level in the LBE to the level of about 10^{-6} wt.% [1], whereas untreated LBE is almost saturated with oxygen; here also, a conditioning procedure is needed.

For MYRRHA [2] a windowless spallation target design was chosen because of the space permitted in the high-performance subcritical core to accommodate the spallation loop. The 120 mm central hole limits the proton beam to about a 70 mm diameter since a minimal lateral target volume is needed for an efficient spallation reaction. A 350-MeV, 5-mA proton beam was chosen for reasons of geometry and the performance requirements of the subcritical core. The resulting current density runs up to about 150-200 $\mu A/cm^2$ due to beam profile shaping. This value exceeds other attempts for a window design for spallation sources that are already at the cutting edge of present-day technology by more than a factor three, and thus a window between target and beam line is not feasible in MYRRHA. This fact raises a third item in the preparation of LBE for use as spallation target material. The common vacuum between target and beam line allows the beam to interact with the gas above the target coming from initial outgassing, liquid-metal evaporation and later emanations of volatile spallation products from the target surface. The proton beam will trigger an ionisation cascade by generating secondary electrons from the gas and thus cause the formation of secondary plasma that may transfer a part of the megawatt beam power to the beam line walls. In turn, the plasma causes sputtering on the inner beam line walls and an increase in desorption and evaporation since both electrons and ions in the plasma will have enough energy to drive out atoms from the walls. Eventually a plasma formation runaway could take place, leading to beam tube damage and eventually to beam clogging.

The plasma problems can be solved if a sufficiently high vacuum above the target ($<10^{-3}$ mbar) can be achieved. The vacuum pumping speed (ref: H_2) in the direct area of the target surface, however, is limited to about 100 l/s due to space constraints in the subcritical core. With these values, the highest acceptable emanation rate is around 10^{-1} mbar l/s. This represents 1 g of H_2 per day being emanated from close to 5 000 kg of LBE in the spallation loop. Since the amount of plasma formation critically depends on the competition between the pumping capacity and the outgassing/evaporation rate, both should be investigated in a realistic situation where quantitative data are obtained to establish and/or confirm design criteria for the spallation loop. It is indeed not *a priori* clear to which level volatile elements are removed in the conditioning process of the LBE before operation of the target. In any case, the problem is too complex to be dealt with reliably on the basis of theoretical model calculations alone.

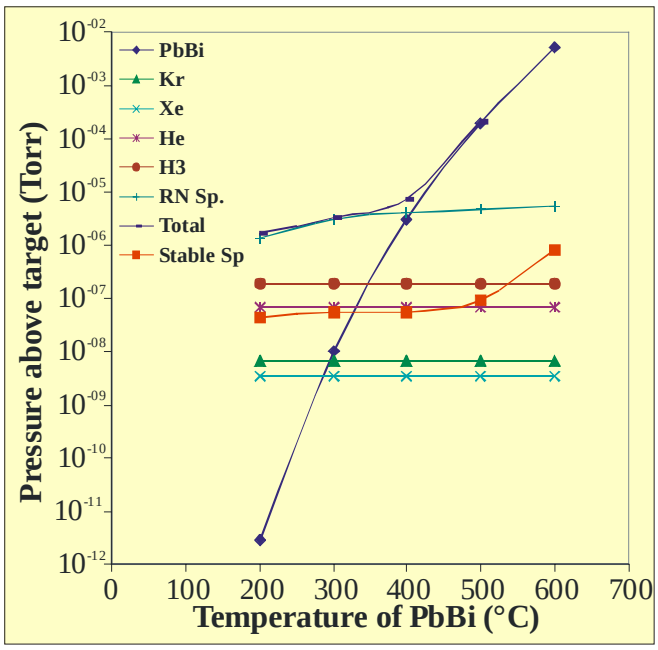
Other issues to be investigated that are closely related to the outgassing problem are the dynamical behaviour of volatile spallation elements like Hg and Po: to what is the emanation rate from the free surface and how do molecules propagate towards the vacuum system. Obviously, this has direct implications on radioactive waste management and operational control of the spallation source.

Experimental programme

As MYRRHA is the first ADS project to clearly adopt a windowless design, it is necessary to obtain a quantitative experimental compatibility assessment of the free LBE target surface with the beam tube vacuum. To investigate the items raised above an experimental set-up was built combining two vessels, in each of which a part of the experimental programme will be carried out. In the barrel-shaped preconditioning vessel (PCV) the procedures for the initial cleaning, oxygen level control and coarse outgassing of the LBE will be studied. The second vessel, built to perform the vacuum interface compatibility experiment (VICE), is largely a one-to-one ultra-high vacuum (UHV) model that mimics the lower part of the beam line connecting to a liquid-metal target in the centre of the subcritical core. The large scale avoids the need to extrapolate the results from small-thimble experiments, which would not necessarily scale in all respects. In the device we will study metal evaporation, emanation of volatile spallation products and the initial and long-term outgassing of the target material. Possible compound formation and the gas transport properties of the beam line will also be studied. The main detector is a high-resolution quadrupole mass spectrometer placed near the top of the vessel. Data will be taken as a function of pressure and temperature. The measured flow rates will be calibrated by comparison with known externally introduced mass flow rates using the mass spectrometer as the differential detector. It may be noted that related work is being undertaken in the framework of TRASCO-ADS [3]. In the next sections, both experimental vessels, the equipment and operational procedures that will be used in the gas experiments and the present status are described [4].

As a first step in the programme the emanation of all types of spallation products and liquid-metal evaporation was estimated [5] (Figure 1) using a deterministic model of the vacuum pumping scheme of the lower end of the spallation loop and beam line. Instant release of all products and zero sticking coefficients were assumed, representing, at least from a vacuum point of view, a worst-case scenario. It was found that this contribution to the atmosphere above the target can easily be tolerated if the particle emanation of the free surface is less than the equivalent emission occurring when the full surface is at 500-600 C. This indicates the importance of the limited acceptability of hot-spots.

Figure 1. Estimated emanation of spallation products and liquid-metal evaporation in the MYRRHA spallation source as a function of temperature



The preconditioning vessel

A scheme of the precondition vessel (PCV) is shown in Figure 2. It is a 68.5 l barrel-shaped 10 bar pressure UHV vessel for temperatures up to 500 C. The ultra-high vacuum (UHV) preparation includes inner surface cleaning and electro-polishing to avoid any contamination of the LBE material, thus ensuring clean experimental conditions. The vessel can simply be cleaned and re-polished to return it to its initial state should any damage occur. This also makes the PCV suited to simulate and study possible oxygen ingress and the subsequent reconditioning of the vessel and the LBE. Its main use, however, is in the initial conditioning procedure of the LBE. For this, 100.00(1) kg (9.6 l) of LBE was molten in the PCV (Figure 3) at a pressure of 10^{-7} mbar. Temperatures are measured by K-type thermocouples and controlled by a Labview® based data-acquisition and control program (DAC) that also monitors the status of other sensors (pressure, oxygen control). During commissioning a bake-out sequence of 5 h at 200 C, 11 h at 330 C and 6 h at 400 C was used. It was shown that the vessel can be heated homogeneously to 2% and temperatures can be kept stable to within 1 C.

After the LBE melt, the liquid surface is inspected visually via two 67-mm quartz windows on top of the vessel that can withstand an interior pressure of up to 10 bar. A double set of helicoflex-type metal seals are used to allow window temperatures up to 450 C. An internal shutter system is foreseen since the windows become opaque after a few weeks at 400 C due to LBE vapour deposits. When surface contamination of the liquid metal is too severe – which may occur since most insoluble impurities will be lighter than the LBE and thus float – a first coarse cleaning can take place by opening the vessel under an inert gas flow and skimming the surface. Subsequently, excess PbO must

Figure 2. A schematic overview of the preconditioning vessel

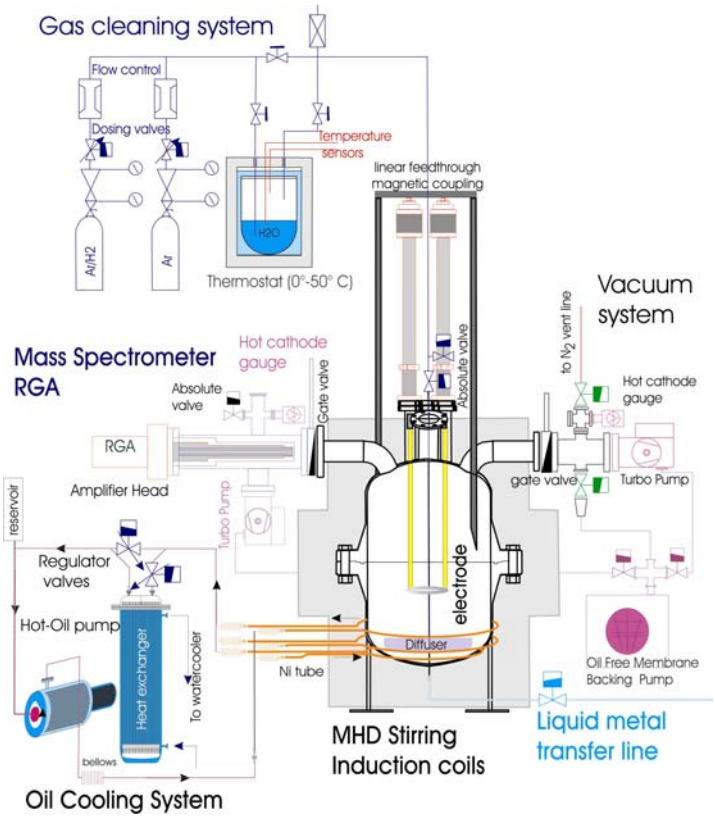
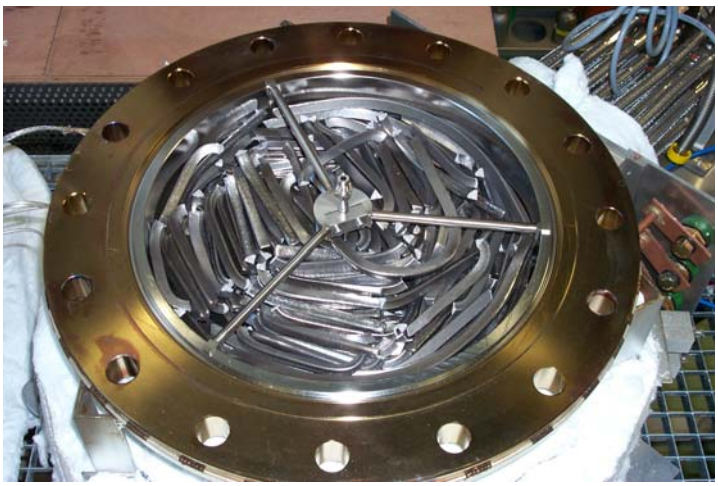
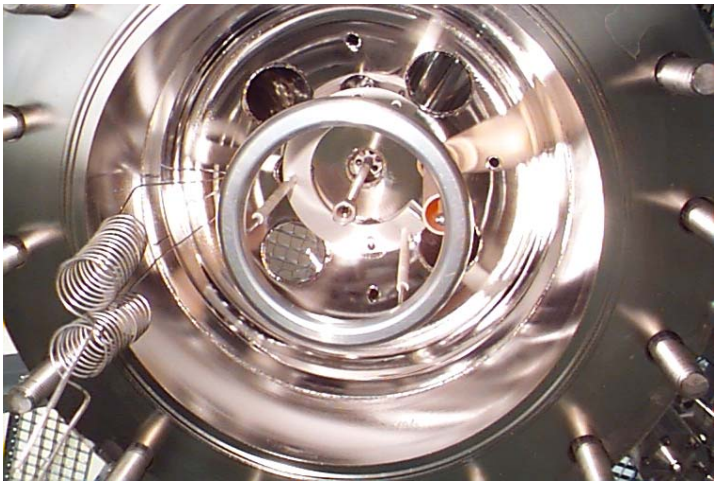


Figure 3. The bottom shell of the PCV being loaded with LBE bars. In the top right corner the connections to the Ni coils are seen.



be removed by lowering the oxygen content in the liquid LBE. For this purpose the PCV is equipped with a gas cleaning system where water vapour is added to a 5% H_2/Ar mixture by bubbling it through water that can be set at any stable temperature between 0 and 50 C. By tuning the gas flow and the water temperature the H_2/H_2O ratio that governs the oxygen potential is set to the desired level. The gas mixture is then spread out in the liquid metal via a stainless steel diffuser. At the exit, the water content of the gas is verified using a chilled-mirror dew point meter. A solid zirconium-oxide electrolytic cell with a Pt/air reference system monitors the oxygen content in the vessel while probes with Ni/NiO and Bi/Bi_2O_3 references are dipped into the LBE melt. After the gas cleaning, most insoluble material will have been removed. Yet, for the small amount of debris that may still be present on the face of the melt, electrical glow discharge cleaning is foreseen (Figure 4). A Cr-coated electrode is mounted on a magnetically coupled linear motion feed-through. After the chamber is evacuated, an appropriate reducing gas, e.g. deuterium, is injected to create a background pressure of 10^{-4}-10^{-2} mbar. A bias voltage on the electrode in the gas creates plasma that bombards the surface of the liquid metal and the walls of the vessel, thereby reducing and removing outer layers. This material is then eliminated from the vessel by the pumping system.

Figure 4. The inside of the upper lid of the PCV showing from left to centre: internal thermocouples, glow discharge electrode, inspection window and inlet tube for gas treatment



The liquid metal contained in the PCV is stagnant, although the experiments discussed here are to represent a moving free surface in a liquid-metal loop. Thus, it is essential to be able to renew the surface of the melt. Also, for most cleaning processes that only work efficiently on good contact of the gas with the LBE, surface renewal is important. To achieve this, magneto-hydrodynamic (MHD)–driven stirring is installed using three sets of Ni-tube coils, put around the vessel. Each set is connected to one of the three phases of a special 50-Hz mains transformer that delivers up to 600 A RMS current in each phase. The low frequency assures a penetration depth of the AC magnetic field of a few cm. Finite-element calculations have shown that, e.g. at 2 cm from the inside wall of the vessel, a magnetic field of up to 10 mT can be reached. With the viscosity and density of the molten LBE given, this suffices to create a recurrent flow in the crucible going up (down) along the walls and down (up) through the centre of a few cm/s.

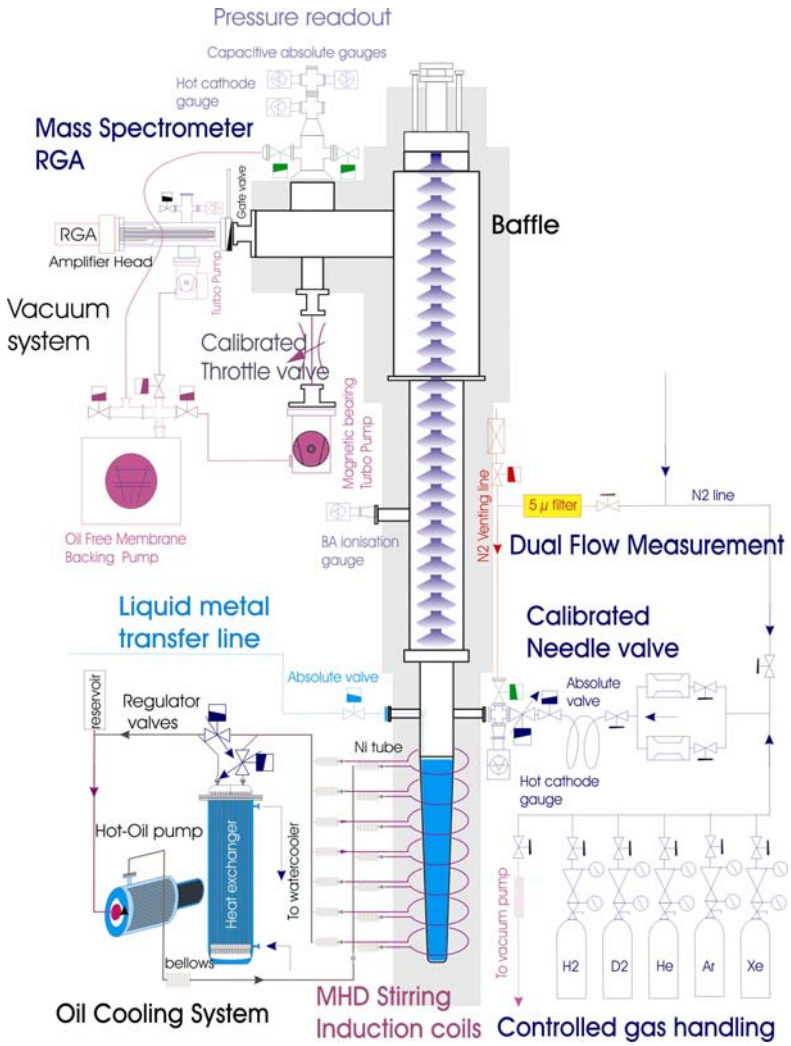
The UHV in the PCV is generated by a small 60 l/s turbopump with greased bearings backed by a diaphragm pump. The system yields a minimal hydrocarbon contamination in the vacuum vessel without using a turbopump with full magnetic bearings. It has an end-vacuum capability for leak testing down to mass spectrometer conditions below 10^{-7} mbar and can cope with the expected emanations. Pressure readout from a full range Bayert-Alpert (BA) ionisation/pirani gauge ($5\ 10^{-10}$-1 000 mbar) is stored by the DAC system. The vacuum pump and gauge are separated from the main vessel by a 63-mm UHV gate valve. Opposite to this, a 100-mm UHV gate valve allows the installation of a rest gas analyser (RGA) based on a triple-quadrupole mass spectrometer. A more detailed description of the RGA will be given in the next section.

Before and after each cleaning step in the conditioning of the LBE, the overall outgassing and evaporation flow rate will be assessed by measuring the pressure reached in the vessel against a known pumping speed and by observing the time evolution of the pressure rise in the closed-off vessel. Also, the qualitative appearance of the eutectic mirror will act as an important guide in the cleaning process. In the development of the cleaning procedure, these data will be taken after repeated exposures to air and following application of the various cleaning methods. Once the process is complete, the liquid LBE is shifted to the VICE vessel. A 6-mm stainless steel transfer tube connects the bottom of the PCV with the inlet port in VICE. On both ends, a diaphragm valve allowing a temperature of up to 260 C terminates the line. To be able to transport the LBE mainly by gravity, the PCV is positioned on a platform such that the bottom of the vessel is about 2 cm above the level of the inlet port in VICE. However, the possibility exists to pressurise the PCV during the move with an inert gas that is eventually used at the end of the transfer procedure to purge the transfer line.

The VICE vessel

The VICE vessel is a 5-m high elongated UHV chamber resembling the lower part of the proton beam line near the liquid-metal target. A schematic drawing of the device is shown in Figure 5. At the bottom of the column the LBE, coming as pre-treated from the PCV, is stored in a 900-mm high crucible. The entry point of the transfer line is at the lowest port above the surface level of the melt. The line itself continues below the surface level to ensure that any material added later on (e.g. tracer Hg or Te) is deposited into the melt. In its top part, the VICE vessel holds a ca 3-m long baffle system that gives VICE a conduction rate limitation for the assessment of flow rates and to possibly limit (retard) the migration of condensable metal vapours to the top. It consists of 36 truncated cones with a base diameter of 240 mm and an 80-mm free bore. Thermocouples are installed on the baffle that can be cooled or heated separate from the vessel by circulation liquid of the correct temperature through a built-in circuit. A quartz oscillator microbalance is foreseen to be mounted to assess the evaporation and outgassing rate of condensable materials in the free bore. Also, dismountable surface elements on the baffle will permit post-mortem study of deposits on test areas. At the very top, a single window is

Figure 5. An overview of the technical layout of the VICE experimental vessel



placed, similar to the ones on the PCV. Besides visual inspection of the melt surface, it allows experiments concerning laser-based level detection (LIDAR) that could possibly be carried out in VICE in preparation for the real level monitor to be employed on MYRRHA. As before, the inner surface of VICE is electro-polished and cleaned to UHV requirements. However, because of the specific geometry of the vessel, reconfiguration of the surface finish after extended damage has occurred is not easy. The full VICE vessel can be baked to 500 C, for which purpose a 7-kW heating and temperature control system, similar in design and operation to the one on the PCV, is installed. The MHD stirring device is also similar as before, though here 12 coils are installed because of the elongated shape of the crucible. Each of these is connected alternately to one of the three phases of the 600-A mains transformer. A glow discharge cleaning electrode can also be installed in the VICE vessel, albeit in a fixed position. It will solely be used for minor corrections since the bulk of the conditioning should already have taken place in the PCV.

At the top of the vessel a 400-l/s magnetic bearings turbopump with an integral Holweck stage, able to discharge against 10 mbar and backed by a 3-m^3/h membrane pump, is placed. To ensure that the total pumping speed is dominated by the dimensions of the vessel and not by the pump, it is oversized relative to the geometrical pumping speed of the vessel of less than 100 l/s (H_2 value). The

vacuum system in VICE is completely oil-free, thus preventing any hydrocarbons from entering the vacuum. Still, the pump is able to provide the vessel with initial clean UHV conditions reaching a base pressure below 10^{-8} mbar. At the entrance of the turbopump the pumping speed can be throttled over nearly four orders of magnitude to almost zero by a calibrated variable conduction valve. This device is implemented for flow rate measurements and to obtain a means of changing the pressure over a reasonable range independently of the flow rate in the system in a stable manner. The valve can also be fully closed so that it acts as a shut-off valve (albeit with internal Viton seal). The entire VICE-PCV vacuum system is metal-sealed to the outside atmosphere to achieve absolute He leak-tightness ($<10^{-9}$ mbar.l/s for components, $<10^{-8}$ mbar.l/s for the system) and a maximum bake-out temperature.

The vacuum gauges that provide pressure readout are not merely process control devices but form an integral part of the data-collecting equipment. During the outgassing and evaporation experiments the pressure over the full vessel must be known as accurately as possible. To this end, two hot-cathode/ pirani full range gauges ($2\ 10^{-6}$-1 000 mbar) are placed, one just above the liquid-metal surface and one at the top end of the column. In the centre of the vessel a Bayert-Alpert (BA) ionisation/pirani full range gauge ($5\ 10^{-10}$-1 000 mbar) is mounted and a pair of absolute capacity pressure gauges providing a total range of 10^{-4}-110 mbar is put at the top of the set-up. All vacuum gauge readings are registered in the DAC computer. A known drawback of a hot-cathode gauge is its long-term drift that limits its accuracy to about 30%. In our set-up, they can be calibrated against the BA gauge and the capacitive gauges in the low- and high-pressure ranges respectively. The latter two types have an accuracy of about 5% and 0.15% over their respective useful ranges. The (re-)calibration is practically done by gradually changing the pressure via the calibrated throttle valve or by slowly venting the vessel through the needle valve after the pumps have been closed off. It is foreseen to calibrate the gauges for a particular type of gas using one of the species available in the controlled gas handling system.

The rest gas analyser (RGA) is the most important diagnostic tool in the liquid-metal outgassing and evaporation experiments discussed here. The apparatus is based on a Hiden triple-quadrupole mass spectrometer. In combination with a dedicated 0-50 amu RF generator, the RGA reaches a mass resolution $\mathrm{D}m/m > 2\ 000$ in the 0-50 amu range. Although mass calibration of the spectrometer is relatively straightforward, an absolute signal calibration is not possible since it depends on the efficiency of the ion source and the detector, which may drift for various reasons. Thus, differential calibration against a known gas flow is necessary. The spectrometer itself is housed in its own small vacuum chamber that has a turbopump and a full range hot-cathode vacuum gauge. At the entrance of the RGA chamber an additional variable aperture is introduced. This makes it possible to match the required vacuum for the RGA to the experimental vacuum that is to be changed as matter of the experiments, by differential pumping. The chamber also acts as a protection for the fragile quadrupole structure and makes is relatively easy to move the RGA to either one of the PCV or VICE vacuum vessels.

A gas-inlet port is placed just above the level of the liquid metal to allow gas flow calibration. The flow is controlled by an all-metal regulating needle valve with a minimal flow of 10^{-10} mbar l/s for pure gasses. A shut-off diaphragm valve is placed in series. The mass flow is measured by a set of two parallel mounted differential heat transfer type flow meters giving a total measurement range of almost four orders of magnitude down to 10^{-2} mbar l/s with a relative accuracy of 1%. For lower flow rates, the needle valve calibration will be used. In the gas-handling system H_2, D_2, He, Ar and Xe are presently foreseen as calibration gases. N_2 is also available, although it is taken from a supply of liquid nitrogen at hand in the laboratory. The calibration gases are not limited to the species mentioned here since in principle, any known gas can be used. Once a known gas flow is allowed into the vessel, its signal can be measured by the mass spectrometer. However, the transport properties of the gas through the VICE column (and through the beam tube in reality) will depend on the pressure and temperature and thus the calibration must be done all temperature and pressure regimes used in the real experiment. They should also be repeated in close time-wise proximity before and after the actual measurements.

When the calibration is established, mass flow rates of emanated gases and vapours measured in the RGA in given circumstances can be linked to an evaporation or outgassing rate. The composition of the gas flow will be resolved using the mass characteristics of the gas as measured by the quadrupole spectrometer. In this way the emanated species can be identified. In these conditions the eventual experiment can be performed measuring the mass flow rates of material outgassing or evaporating from the liquid melt. These values are expected to lie between 10^{-6} and 10^{-1} mbar l/s. The data will be taken as a function of the pressure in the vessel, the liquid melt temperature, the stirring rate and the baffle temperature. The experimental data will serve as input into the modelling of the interaction of the proton beam with the emanated gasses and vapours in order to eventually assess the plasma formation problem described in the introduction.

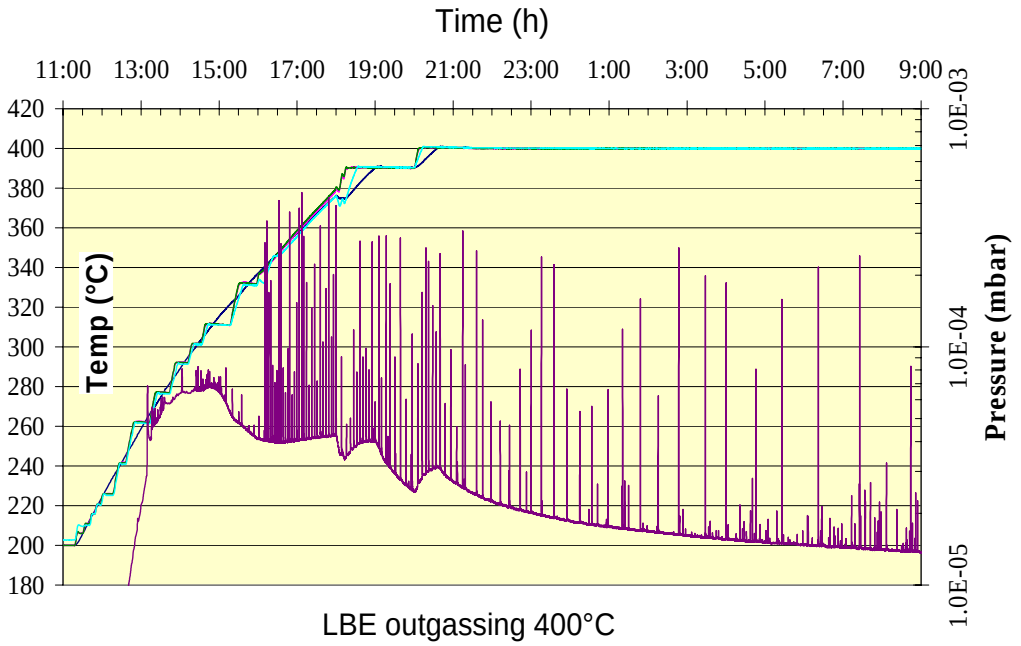
In a second stage, stable isotopes resembling the spallation products like, e.g. Hg and Te, may be added to the LBE via the liquid-metal transfer line. By using the MHD stirring device the materials should be adequately mixed such that in effect the outgassing of spallation products is simulated. Subsequently, the mass flow of the spallation product will be measured as a function of time at a given temperature and pressure. In this way the measurement will yield information on the dynamical gas transport properties of the beam line and the time-integrated signal will give data on the emanation probability of spallation products produced in the target. Of course, because of the time dependence, the tracer material will have to be inserted in a pulsed mode.

At present, the commissioning and testing stage of the VICE vacuum vessel and instrumentation is well underway while the commissioning of the PCV is finished. The vessel has been loaded with 100 kg of LBE and the initial coarse outgassing tests have taken place. The characteristic spikes in the vacuum pressure readout due to the release of gaseous inclusions in the LBE ingots during melt were observed. After 24 h of pumping with the LBE at 150 C a base pressure of $1\ 10^{-6}$ mbar was reached. However, it was found that even after several days, stirring of the melt induced additional gas emanation to a sufficient level to increase the pressure to 10^{-3} mbar. This shows the importance of stirring the melt during the outgassing process. When heating the LBE to 400 C, the bulk of the outgassing appeared to start only above 320 C (Figure 6), with the development of bubbles yielding pressure spikes. With the LBE at 400 C, a pressure of $4\ 10^{-6}$ mbar was eventually achieved after seven days of pumping. These values are two orders of magnitude below the minimal requirements mentioned in the introduction. However, the presence of a PbO layer on the liquid LBE possibly inhibits gas emanations from the surface and thus these preliminary results do not show the intrinsic vacuum compatibility of the windowless target design. To check this issue, the vessel was opened with the melt at 150 C and exposed to air for 24 h without stirring. Prior to re-closing the vessel, the surface of the LBE was mechanically skimmed. After subsequent reheating and pumping, a pressure below 10^{-6} mbar was reached within 24 h after reheating the melt to 400 C. This indicates that initial outgassing only needs to be done once as long as the melt is kept stagnant when a cover gas is used.

The next step is the (time consuming) development of the cleaning and oxygen control process. In the first instance a gas diffuser with a sintered stainless steel surface was employed to distribute the H_2/Ar gas in the melt in very fine bubbles. However, the sintered material turned out not to be able to withstand immersion in LBE for extended periods of time. This is probably due to wetting of the LBE on the large surface of the sintered part. At present, tests are undertaken with a much coarser diffuser using thick-walled stainless steel tubing with 2-mm holes.

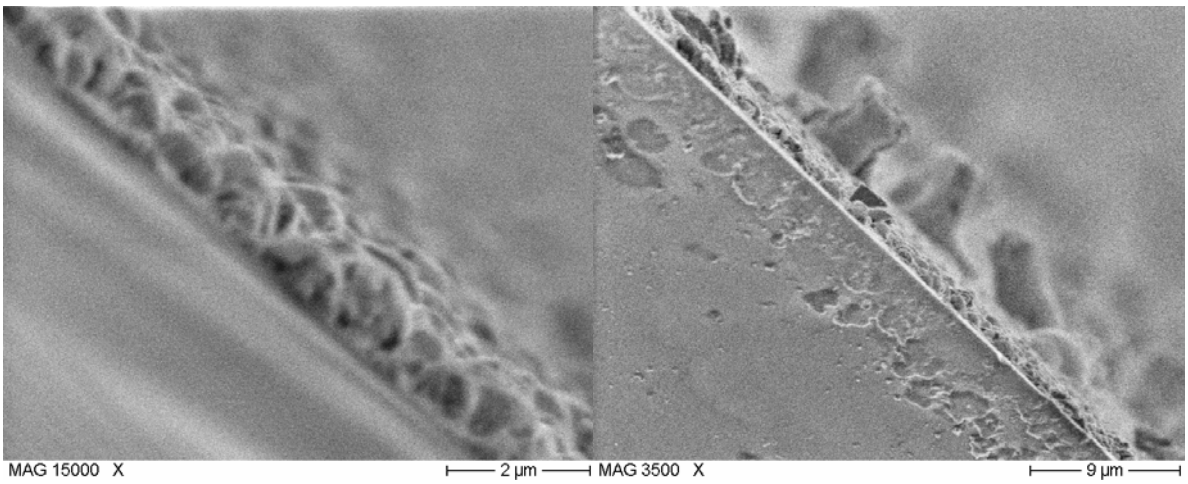
During the outgassing and conditioning experiments, a layer of “black dust” was formed on the quartz viewport windows. The study of this layer is relevant since the nature of the deposit will not only have implications on the instrumentation in the vacuum of the spallation loop but also on that in the gas plenum above the main LBE pool [2]. Chemical analyses have shown that the deposit consists

Figure 6. Initial outgassing of LBE at 400 C



of 69% Bi and 31% Pb with other detectable elements (Zn and Sn) only present at the ppm level. Comparing this to the composition of the LBE in the vessel (54.3 wt.% Bi and 45.7 wt.% Pb by analyses) we see that there is a small preference for Bi deposition and thus there is no indication that the "black dust" should be mainly PbO. On electron microscope images of the layer it can be seen that the layer is built up from small 500-nm droplets. It is not clear whether these are formed by surface tension when Pb and Bi vapour condenses on the quartz windows or that the droplets are formed as an aerosol during the gas treatment phase of the conditioning process. This question will only be resolved when the analyses of a "black dust" layer deposited in clean UHV conditions is complete.

Figure 7. Electron microscope images of LBE deposited on quartz windows. In the background SiO_2 fragments of the substrate are seen. The bulk of the substrate is seen on the bottom left.



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