

*Potential Containment Materials for  
Liquid-Lead and Lead-Bismuth Eutectic  
Spallation Neutron Source*

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# Potential Containment Materials for Liquid-Lead and Lead-Bismuth Eutectic Spallation Neutron Source

by

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## ABSTRACT

Lead (Pb) and lead-bismuth eutectic (44Pb-56Bi) have been the two primary candidate liquid-metal target materials for the production of spallation neutrons. Selection of a container material for the liquid-metal target will greatly affect the lifetime and safety of the target subsystem. For the lead target, niobium-1 (wt%) zirconium (Nb-1Zr) is a candidate containment material for liquid lead, but its poor oxidation resistance has been a major concern. The oxidation rate of Nb-1Zr was studied based on the calculations of thickness loss due to oxidation. According to these calculations, it appeared that uncoated Nb-1Zr may be used for a one-year operation at 900°C at  $P_{O_2} = 1 \times 10^{-6}$  torr, but the same material may not be used in argon with 5-ppm oxygen. Coating technologies to reduce the oxidation of Nb-1Zr are reviewed, as are other candidate refractory metals such as molybdenum, tantalum, and tungsten. For the Pb-Bi target, three candidate containment materials are suggested based on a literature survey of the materials compatibility and proton irradiation tests: Croloy 2-1/4, modified 9Cr-1Mo, and 12Cr-1Mo (HT-9) steel. These materials seem to be used only if the lead-bismuth is thoroughly deoxidized and treated with zirconium and magnesium.

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## 1. Introduction

In the current preconceptual design for the Los Alamos National Laboratory (LANL) Accelerator Based Conversion of Plutonium/Accelerator Transmutation of Waste (ABC/ATW), flowing liquid metal is the primary candidate target for use as a compact, high-power spallation neutron source. As 800-MeV protons generated from an accelerator impinge on the liquid metal target, neutrons are generated by a spallation process. These neutrons then are used to burn plutonium or to transmute high-level wastes. The advantages of a liquid-metal target for these applications are fast cooling of the target by flowing-liquid-metal coolant, great power density, and no irradiation damage to the target material. Lead (Pb) and lead-bismuth eutectic (44Pb-56Bi) have been the two leading candidate target materials for the production of spallation neutrons because of their high atomic numbers, low melting points, and low parasitic neutron capture. As shown in Table 1, lead has the advantages of less polonium (Po) production and less corrosiveness. However, due to its higher melting point (327°C) than Pb-Bi eutectic (125°C), Pb would require higher operating temperatures.

Table 1. Advantages and disadvantages of Pb and Pb-Bi eutectic targets.

	Advantages	Disadvantages
Pb	Less Po production Less corrosive	Higher operating temperature (m.p. = 327°C)
Pb-Bi eutectic	Lower operating temperature (m.p. = 125°C)	More Po production More corrosive Swelling after solidification

Because the beam window is an integral part of the target containment structure, the structural container for the liquid metal will be exposed to a significant flux of high-energy protons and neutrons as well as an extremely corrosive environment. Selection of the container material for the flow passage will greatly affect the lifetime and safety of the target subsystem. The material should have good compatibility with the liquid-metal target, sufficient mechanical strength at operating temperatures, a low-neutron-absorption cross section, and good performance under an intense proton environment. In this paper, we discuss the potential containment materials for the Pb and Pb-Bi eutectic target that were investigated based on the above criteria.

## 2. Lead Target

In the LANL ABC/ATW preconceptual liquid-lead target design, it is estimated that the maximum temperature of the system will be approximately 900°C at the point where the proton beam impinges on the window. In this temperature range, iron-base alloys are inadequate because of their high creep rates and poor oxidation resistance. Iron-base alloys are usually limited to up to 650–700°C service temperatures. Similarly, nickel-base and cobalt-base superalloys are only marginally acceptable because they are limited to up to 900–1000°C. Another consideration for the use of these superalloys is that nickel is incompatible with liquid lead, and cobalt has a very-high-absorption cross section. Refractory metals such as niobium (Nb), tantalum (Ta), molybdenum (Mo), and tungsten (W) are usually used at service temperatures well above 900°C.

It is anticipated that, for the LANL ABC/ATW system, temperature-gradient mass transfer will be the most damaging type of all materials degradation phenomena in liquid metal because of large temperature gradient in the system. The solubility of the container material in the liquid metal is a function of temperature. After a period of operation, solubility limits are reached, and the temperature-dependent solubility results in the transfer of the container material from the hottest location to the coldest location in the flowing loop. The effect in the circulating molten lead target is to dissolve container material from the

window and deposit it in the heat exchanger. Both the thinning of the window and potential fouling of the heat exchanger are undesirable.

The relative resistance to mass transfer of 24 metals and alloys in liquid lead with a temperature gradient of 300°C (800–500°C) was measured by Cathcart and Manly [1]. The results indicated that only Nb and Mo showed no mass transfer, while the other materials showed little to heavy mass transfer. Other parameters to consider in the selection of candidate materials are good mechanical properties in intense irradiation environments and low-neutron-absorption cross sections. Based on the mass-transfer behavior, proton irradiation behavior, mechanical strength at operating temperatures, and neutron-absorption cross section of candidate materials, Nb-1Zr has been selected as a containment material for the ABC/ATW liquid-lead target [2].

## 2.1 Oxidation of Nb-1Zr

Although Nb-1Zr has desirable properties for the liquid-Pb target, one major drawback is its low oxidation resistance at high temperatures. Thus, the ABC/ATW lead target is designed to be used in vacuum or at very low oxygen partial pressures.

Some preliminary oxidation experiments of Nb-1Zr were performed at LANL. Figure 1 shows the weight gain of Nb-1Zr in pure oxygen at a heating rate of 2°C/min. From nonisothermal thermogravimetric analysis, the oxidation rate was determined to be 62.2 mg/cm<sup>2</sup>·h in the temperature range of 700–800°C. The oxidation rate of Nb-1Zr in argon with 5-ppm oxygen at 900°C for 20 hr was 7.2 mg/cm<sup>2</sup> (0.36 mg/cm<sup>2</sup>·h). Oxide scales were formed on the surface of the sample. Figure 2 shows the weight changes of uncoated and plasma-sprayed MoSi<sub>2</sub>-coated Nb-1Zr at 900°C in pure oxygen. The experiment was terminated when the specimen broke loose from the platinum (Pt) hangdown wire. As expected, the oxidation kinetics under these conditions were extremely rapid. Based on the rapid oxidation rates and appearance of the sample after oxidation, the oxidation was thought to be autocatalytic. Although perfectly intact at the end of the experiment, the MoSi<sub>2</sub> coating provided no significant protection from oxidation. This was attributed to lack of time to develop the coating techniques.

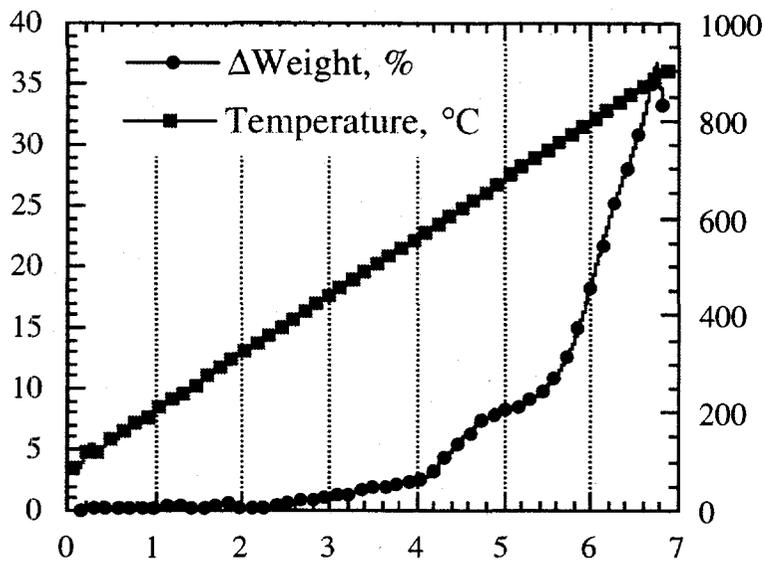


Figure 1. Weight gain of Nb-1Zr in pure oxygen at a heating rate of 2°C/min.

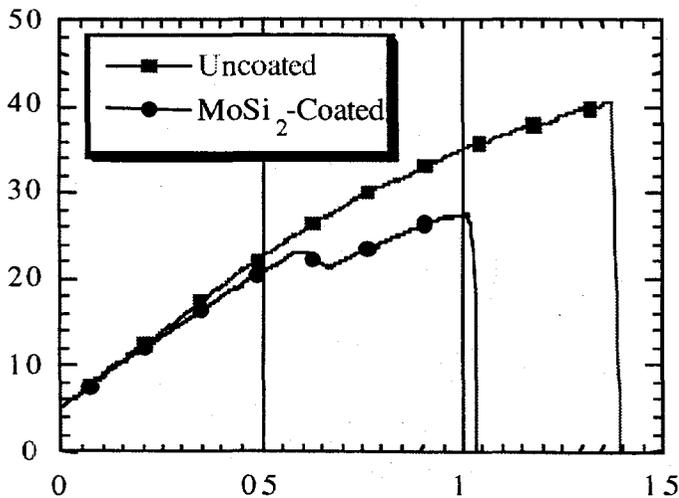


Figure 2. Weight changes of uncoated and plasma-sprayed MoSi<sub>2</sub>-coated Nb-1Zr at 900°C in pure oxygen.

The oxidation behavior of Nb-1Zr has been studied by many investigators. DiStefano and Hendricks [3] measured the oxidation rate of Nb-1Zr in vacuum at oxygen partial pressures of  $10^{-6}$  and  $10^{-7}$  torr at temperatures up to 1077°C for 100–150 hr. Under these conditions, no visible oxide films formed. The oxidation rate at  $P_{O_2} = 1 \times 10^{-6}$  torr and 900°C was expressed as  $r = 6 \times 10^{-3}$  mg/cm<sup>2</sup>·h. At  $P_{O_2} = 1 \times 10^{-7}$  torr and 900°C, the oxidation rate was expressed as  $r = 3.4 \times 10^{-4}$  mg/cm<sup>2</sup>·h. Delgrosso [4] measured the oxidation rate of Nb-1Zr in air at 871°C, and it was 36 mg/cm<sup>2</sup>·h.

Niobium pentoxide ( $\text{Nb}_2\text{O}_5$ ) is the end product of the oxidation. It is a white solid with densities that may vary between 4.3–5.2  $\text{g/cm}^3$  depending on the method of preparation and subsequent heat treatment [5]. The oxidation rate of Nb-1Zr can be converted to thickness loss due to oxidation. Assuming all the oxide is  $\text{Nb}_2\text{O}_5$ , the oxidation rate in argon with 5-ppm oxygen at 900°C measured by LANL (0.36  $\text{mg/cm}^2\text{-h}$ ) equals 2.5  $\mu\text{m/h}$ . The thickness loss during a one-year operation in argon with 5-ppm oxygen would be 2.19 cm. Also, the oxidation rate measured by DiStefano and Hendricks [3] at  $P_{\text{O}_2} = 1 \times 10^{-6}$  torr and 900°C equals 0.0417  $\mu\text{m/h}$ . Thus, the thickness loss during a one-year operation at  $P_{\text{O}_2} = 1 \times 10^{-6}$  torr and 900°C would be 0.0365 cm. Using the same reasoning, the thickness loss at  $P_{\text{O}_2} = 1 \times 10^{-7}$  torr and 900°C measured by DiStefano and Hendricks [3] equals 0.00207 cm.

If we assume that a loss of 20% of the wall thickness is acceptable during one year of operation, the required minimum wall thickness of Nb-1Zr that is exposed in argon with 5-ppm oxygen would be 10.95 cm. Under the same assumption, the required minimum wall thickness of Nb-1Zr exposed to  $P_{\text{O}_2} = 1 \times 10^{-6}$  torr at 900°C and to  $P_{\text{O}_2} = 1 \times 10^{-7}$  torr at 900°C would be 0.183 and 0.01 cm, respectively.

Calculations also were made based on an assumption that loss of 10% of wall thickness is acceptable. Table 2 shows the required minimum Nb-1Zr wall thickness at each condition for one-year operation at 900°C under the 10% and 20% thickness loss criteria.

Table 2. Minimum Nb-1Zr wall thickness required at each condition for 1-year operation at 900°C.

Environment	Thickness loss (cm)	Minimum wall thickness permitting 20% loss (cm)	Minimum wall thickness permitting 10% loss (cm)
Argon (5 ppm $\text{O}_2$ )	2.19	10.95	21.9
$P_{\text{O}_2} = 1 \times 10^{-6}$ torr	0.0365	0.183	0.3650
$P_{\text{O}_2} = 1 \times 10^{-7}$ torr	0.00207	0.01	0.02

The table suggests that the ABC/ATW target system may not employ uncoated Nb-1Zr in argon with a 5-ppm oxygen environment because it requires a minimum wall thickness of 10.95 cm using the 20% thickness loss criterion. Uncoated Nb-1Zr may be used for one-year operation at 900°C at  $P_{\text{O}_2} = 1 \times 10^{-6}$  torr with a minimum wall thickness of 0.183 cm and the 20% thickness loss criterion. At  $P_{\text{O}_2} = 1 \times 10^{-7}$  torr and 900°C, the thickness loss of Nb-1Zr after a year of operation is only 0.002 cm; thus, uncoated Nb-1Zr may be used for one year. These calculations are based on the assumption that all the oxidation phenomena results in the thickness loss from the surface of the material. However, the

internal-oxidation effects on both the mechanical properties of Nb-1Zr and corrosion resistance to liquid metals may be significant [6]. Further study is required.

During the operation of the ABC system, spallation products such as hydrogen and oxygen will build up through the interaction of high-energy protons with the target material. These products will behave as alloying or impure elements in the liquid-lead or lead-bismuth target material where they are formed, and thus will have beneficial, neutral, or detrimental effects on the containment material. Initial calculations showed that less than  $1 \times 10^{-15}$  atoms/proton of oxygen will be produced [7]. This amount seems to be negligible; however, more detailed study on oxygen production rate and on the interaction between the oxygen and other spallation products is required. Hydrogen embrittlement is another consideration. Stephens [8] conducted a hydrogen embrittlement experiment of Nb-1Zr by doping 10 ppm of hydrogen at 1040°C in a partial pressure of hydrogen of approximately 13 KN/m<sup>2</sup> for 10 min. Based on his experiment, he concluded that Nb-1Zr was not susceptible to hydrogen embrittlement at SP-100 operating conditions because of the small amount of hydrogen that would be encountered. For ABC/ATW systems, initial calculations showed that hydrogen is expected to be produced at a rate of 3.265 atoms/proton [7], which seems to be a significant amount. A further study on the hydrogen production rates and hydrogen embrittlement is required.

## **2.2 Coatings for Nb-1Zr**

A literature survey revealed that many coating technologies for niobium alloys have been developed. These coating techniques can be used to protect the Nb-1Zr component for longer service life.

Perkins and Meier [9] reviewed the status of recent technologies to protect niobium from oxidation. Silicide coating were considered to be the best available materials for the oxidation protection of niobium base alloys. Niobium silicides modified with silicides of chromium and iron (R512E) and chromium and titanium (R512A) have been used since the mid-1960s for this purpose. R512E-fusion-coated C103-Nb nozzle flaps have been used for over 15 years on the Pratt & Whitney F100 jet engine without significant failures. These components are exposed to air and products of combustion at temperatures up to 1427°C for hundreds of cycles. Similarly, orbital maneuvering engines for spacecraft, including the space shuttle and satellites, are made of niobium-based alloys coated with R512A or R512E. A major problem with this coating is the mismatch in thermal expansion coefficient, which places the brittle coating in tension on cooling from high temperature. The coating has a ductile to brittle transition temperature in the range of 982–1,093°C. The

mismatch in thermal expansion coefficient can form cracks in the surface during cooling. These cracks will normally heal by diffusion when reheated; however, after a number of cycles the cracks can become oxidized and fail to heal on reheating.

Mueller, Wang, and Rapp [10] developed multicomponent silicide coatings of (Mo, W)(Si, Ge)<sub>2</sub> to protect niobium-based alloys from high-temperature oxidation. A solid solution of molybdenum and tungsten disilicide doped with germanium constituted a primary coating layer that supported a slow-growing protective SiO<sub>2</sub> scale when exposed to high-temperature oxidation. Germanium was added to the cyclic oxidation resistance by increasing the thermal expansion coefficient of the silica film, and tungsten was added to strengthen MoSi<sub>2</sub> at high temperatures. The presence of germanium also avoided accelerated low-temperature oxidation by providing better sealant coverage at low temperatures. Coated niobium coupons at 925, 1,370, and 1,540°C in air successfully passed 200 cyclic oxidation tests of 1 hour each at 1,370°C and 60 cycles of 1 hour each at 1,540°C.

Packer [11] reviewed silicide coating technologies for refractory metals. In this technique, powders of silicon and coating alloying additions such as chromium, iron, titanium, and hafnium are mixed with an organic lacquer vehicle. Typically the powders are mixed in ratios of 60 wt% to 90 wt% silicon with one or more alloying agents. The resultant lacquer/metal powder slurry is handled much like a paint, with concerns over antissettling agents, viscosity control, and drying characteristics. The slurry is applied to a part by painting, spraying, or dipping. Some complex shapes are filled and then drained at a controlled rate. After application of the green, unfired slurry, parts are heat-treated in vacuum at 1,300–1,400°C. During this heat treatment, the powders react with one another as well as with the substrate, form eutectics, fuse, continue to react with the substrate to form high-melting-point metal silicides, and resolidify isothermally. After resolidification, heat treatment is continued for 30 minutes to one hour to permit interdiffusion between the coating and the substrate and the growth of intermediate silicides at the coating/substrate interface.

DiStefano and Hendricks [3] applied a molybdenum coating to Nb-1Zr and investigated the oxidation rate of molybdenum-coated Nb-1Zr. The thin layer (4 μm) of molybdenum coating was excellent in preventing oxidation at 500°C. Metallographic examination of the coated sample after oxidation showed that the molybdenum layer was intact and continuous. However, in a long-term test at 1,077°C and P<sub>O<sub>2</sub></sub> = 1 × 10<sup>-7</sup> torr, significant oxygen pickup by a coated sample was found (806 ppm).

### 2.3 Nb-1Zr Proton Irradiation Experiments at LANL

During the summer of 1994, three Nb-1Zr samples were irradiated by 800-MeV protons at the Los Alamos Meson Physics Facility (LAMPF) to test the change in material properties (i.e. strength, ductility, etc.) due to proton (and neutron) irradiation. A total of three samples were fabricated into an irradiation assembly, each 0.020 in. thick, as shown in Figure 3. Also included in the assembly was an aluminum (99.9408% purity) sample used to measure the distribution of the proton beam. All samples were marked to maintain proper orientation following the irradiation.

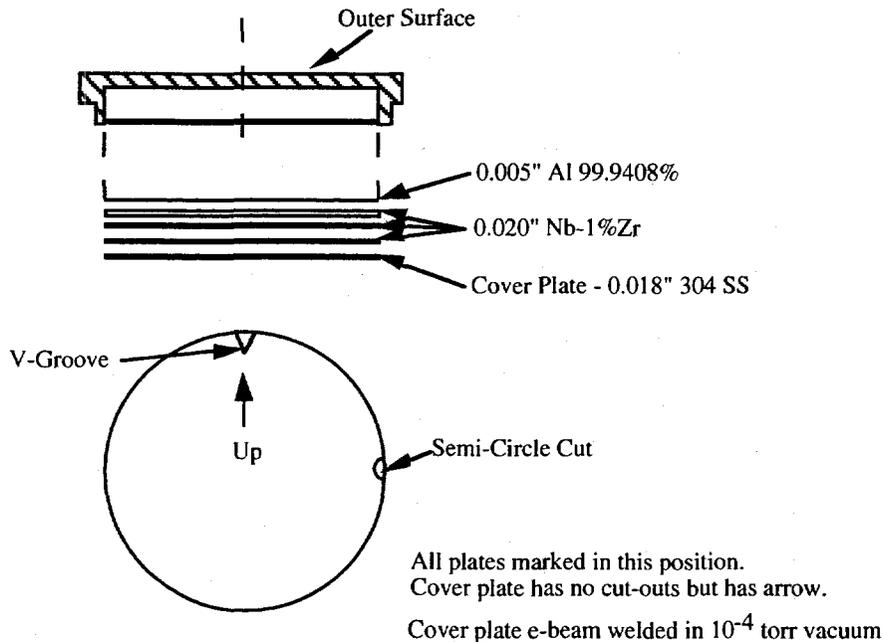


Figure 3. Irradiation assembly.

Initially, two coated samples were going to be included in the irradiation experiment to test the ability of coatings to maintain their integrity under proton irradiation. Samples were sent to the ALON Company for coating with alumina. Although experienced in using this type of coating on stainless steels, ALON was unable to successfully coat the Nb-1Zr. ALON attributed this failure to incorrect temperatures during the coating application and stated that a substantial experimental development effort would be required to be able to perform a successful coating. Hence, the alumina coating was abandoned for the irradiation experiment.

Earlier, small samples of Nb-1Zr were coated with MoSi<sub>2</sub>, and some simple oxidation experiments were performed. Although the MoSi<sub>2</sub> was ineffective in reducing the oxidation rates of the Nb-1Zr at high temperatures, an attempt was made to obtain an MoSi<sub>2</sub>-coated

sample for the proton irradiation in order to obtain some information on coating degradation. Unfortunately, attempts to coat the larger samples required also failed due to the mismatch in thermal expansion coefficient between the sample and the coating. Again it was determined that development of a successful coating technique was impractical under the time restraints. Thus, no coated samples were included. After proton irradiation, the sample assembly was moved to hot cell for post-irradiation examination.

## **2.4 Molybdenum, Tantalum, and Tungsten**

Brown and Cost [12] investigated the tensile properties of Ta and Mo after 800-MeV proton irradiation at LAMPF. Ta specimens retained significant ductility (approximately 17%) after proton fluence of  $1.5 \times 10^{20}$  p/cm<sup>2</sup>, while several Mo specimens broke at less than 0.2% strain after proton fluence of  $1.3 \times 10^{19}$  p/cm<sup>2</sup>. Thus, all the candidate refractory metals of Mo, Ta, and W have their own problems as containment materials for the ABC/ATW. Mo becomes too brittle after low-fluence proton irradiation. Ta has an unacceptably high thermal neutron absorption cross section (21.3 barns) and poor oxidation resistance. W also has a high thermal neutron absorption cross section (19.2 barns) and low ductility.

Another possible way to alleviate the materials problem for the lead target is the use of coolant to lower the window temperature. By reducing the maximum operating temperature at the proton window, more candidate materials would be available.

## **3. Lead-Bismuth Eutectic Target**

### **3.1 Materials Compatibility**

As an alternative to a liquid-lead target for the LANL ABC/ATW systems, a liquid-lead-bismuth eutectic target was investigated.

In the literature, several researchers used thermal convection loop tests to investigate materials compatibility with lead-bismuth eutectic. During the 1950s, Cygan [13, 14] performed Pb-Bi eutectic thermal convection and pumped loop tests for several steel alloys; the results of the tests are shown in Table 3. Main points of the results were the following: (1) when a steel surface is not completely deoxidized, impurities ( $\text{Bi}_2\text{O}_3$ ,  $\text{Pb}_3\text{O}_4$ ) rather than corrosion products appeared to be the major factors resulting in the stoppage of passage; (2) a possible method of metal purification might be the use of a magnetic trap; (3) low-carbon steel appeared to be a useful container for the eutectic up to 454°C; and (4) there was no advantage in using 410 or 446 stainless steel up to 454°C. Weeks [15] reviewed the

early work of metallurgical studies on Bi and Bi alloys for reactor fuels or coolants. Later in the 1960s, Romano, Klumet, and Gurinsky [16] investigated container materials for Pb, Pb-Bi eutectic, and Pb-Mg eutectic in thermal convection loops operating at temperatures up to 650°C and at average flow velocity of 1.5 cm/s. The results are tabulated in Table 4. A loop made of Croloy 1-1/4 (Fe-0.15C-1.25Cr-0.5Mo) with a temperature range of 200–400°C ran for 10,000 hours without signs of corrosion. A similar uninhibited loop with a temperature range of 300–450°C at Chalk River Nuclear Laboratories plugged in 1,700 hours [17]. At 650°C severe corrosion was observed in less than 100 hours.

Table 3. Results of lead-bismuth eutectic thermal convection loop test [13].

Container Material	Velocity (cm/s)	Hot Leg Temperature (°C)	$\Delta T$ (°C)	Time for Stoppage (hr)
1015 steel 1/4" ID 1/8" wall	3.3	454	277	450
410 stainless steel 1/2" ID 1/16" wall	8.8	454	500	1600
446 stainless steel 5/8" ID 1/16" wall	7.0	454	266–277	2000
1015 steel 1/4" ID 1/8" wall	4.5	454	288	220
410 stainless steel 1/2" ID 1/16" wall	3.6	454	277	950
446 stainless steel 5/8" ID 1/16" wall	6.1	454	222–277	1030

Table 4. Corrosion of steels in lead-bismuth eutectic [16].

Severe corrosion <1,000 hr	Croloy 1-1/4 (uninhibited) (500–625°C)
Severe corrosion 1,000–5,000 hours	Croloy 1-1/4 (uninhibited) (400–550°C)
No corrosion Up to 5,000 hr	Croloy 1-1/4 (Ti added) (350–650°C) Carbon steel (Ti or Zr added) (200–500°C, 450–600°C, and 500–650°C)
No corrosion Up to 10,000 hours	Croloy 1-1/4 (uninhibited) (200–400°C) Croloy 1-1/4 (Zr added) (500–650°C) Carbon steel (Zr added) (500–650°C)

Inhibitors are very effective in reducing the corrosion of steel by forming, for example, carbide and nitride films on the surface. Zirconium is perhaps the most effective inhibitor of

low-alloy steels [16], and it also has a lower thermal neutron cross section than titanium. The required concentration of titanium is slightly more difficult to maintain than that of zirconium. Zirconium nitride is the major component of the protective film at the steel/bismuth interface in a zirconium-inhibited system [18]. Magnesium or calcium normally are added to the liquid metal in concentrations below 1,000 ppm as an oxygen-getter. Magnesium concentrations above 1,000 ppm in Bi loops appear detrimental because of suppression of zirconium solubility and possible intermetallic compound formation. In Pb-Bi eutectic at 450°C, 350 ppm of Mg appears to be the upper desirable limit [19]. In Bi, about 250-ppm Mg and 250-ppm Zr were the optimum inhibitor concentrations [20]. It was necessary to add inhibitors during the loop operation [16, 21].

If inhibitors are used during the ABC/ATW operation, a variety of spallation products built up by the interaction of high-energy protons with target material will react with inhibitors in the Pb-Bi eutectic. These reactions may reduce the beneficial effect of inhibitors. More research on the interaction between the spallation products and the inhibitors is required.

Weeks, Rosenblatt, and Wilson [22, 23] tabulated the solubilities of several metals in Pb-Bi eutectic, as shown in Table 5. Both chromium and nickel are more soluble than iron. Thus, for stainless steels, selective leaching of these components takes place with increasing Cr and Ni concentration. In Pb-Bi eutectic [19] and Bi [16, 24], the corrosion increases with the chromium content in steel: Carbon steels are the most corrosion-resistant; low alloy steels (<2-1/4% Cr) are moderately corrosion-resistant; and high Cr steels (>2-1/4% Cr) are subject to gross attack [16, 24-26]. Nitrided Croloy 2-1/4 (Fe-0.15C-2.2Cr-1.0Mo) was more corrosion-resistant than the as-received material [16], and the corrosion rate decreased with increasing nitrogen content (0.01%–1%) of Croloy 2-1/4 [26]. The high solubility of nickel excludes the use of austenitic stainless steel or nickel-base alloys as a containment material for Pb-Bi eutectic.

Table 5. Solubilities of several metals in lead-bismuth eutectic [22, 23].

Metal	Temperature (°C)	Solubility (ppm)	Reference
Cu	500	7200	18
Fe	500	2.3	18
Cr	500	11.0	19
Ni	500	25000	19
Co	500	50.3	19
Ti	600	300	18
Zr	600	329	18

The corrosion is commonly velocity-dependent, the rate-controlling step being the migration of dissolved iron or chromium through the boundary layer of liquid adjacent to the steel surface [22]. However, Weeks and Klamut [24] noticed that the velocity in itself did not appear to accelerate corrosion in a liquid-bismuth thermal convection loop test. Very little difference in corrosion rate was observed between thermal convection loops in which the flow velocity was about 1.5 cm/s and pumped loops in which flow velocity was as high as 424 cm/s. However, the circulating system was sensitive to disturbances in the flow. Severe attack had been encountered in hot portions of loops in which there were changes in the direction of fluid flow.

### 3.2 Candidate Materials for Pb-Bi Eutectic Target

The highest temperatures in the LANL ABC/ATW liquid-lead-bismuth eutectic target, where protons impinge on the window, are estimated to be 530–650°C depending on the flow design. Even though carbon steels showed strong corrosion resistance in Pb-Bi eutectic and in Bi, their upper limit for use under continuous load usually is 370°C. At elevated temperatures of 370°–650°C, iron-based heat-resistant alloys are usually considered. The properties of carbon and low-alloy steels change considerably over this temperature range. Considering that there is not much stress exerted on the container material, all the low-alloy steels seem to have enough strength; however, a complete stress analysis in the target region of the ABC/ATW system will be necessary to verify this.

Brown, Wechsler, and Tschalaer [27] studied the tensile properties of pure iron, 2-1/4Cr-1Mo steel, and HT-9 after 800-MeV proton irradiation at LAMPF. The samples were sealed inside capsules containing lead-bismuth eutectic and were proton-irradiated to two fluences, 4.8 and  $54 \times 10^{23}$  p/m<sup>2</sup>. After high-fluence irradiation, both 2-1/4Cr-1Mo steel and HT-9 showed more than 10% ductility and yield strength of more than 600 MPa, while pure iron showed approximately 8% ductility and yield strength of 200 MPa. The two iron-base alloys also showed much smaller fractional changes in yield strength and ultimate tensile strength than the pure iron. The 2-1/4Cr-1Mo steel showed a little higher ductility and lower strength than HT-9. Both HT-9 and 2-1/4Cr-1Mo steels appear adequate for a proton beam window in contact with Pb-Bi eutectic.

Based on the literature survey, the materials discussed below are selected as candidate container materials for the Pb-Bi eutectic targets for the ABC/ATW system. The general tendency for these materials is that the lower the chromium content in the alloy, the higher corrosion resistance to the eutectic and the less strength, and vice versa.

### **3.2.1 2-1/4Cr-1Mo Steel**

The corrosion tests of Croloy 2-1/4 (Fe-2.2Cr-1Mo) in bismuth [16, 22, 24, 26] and of Croloy 1-1/4 (Fe-1.25Cr-0.5Mo) in lead-bismuth eutectic [16, 21] have been reported by many researchers. The 2-1/4Cr-1Mo steel also has been used extensively throughout the power industry as the final superheater for oil-fired power stations operating at a main steam temperature of 540 °C and midwall temperature of 580°C [28]. It has too low of a creep strength to be accepted generally as a final-stage superheater material for use with a steam temperature of 565°C. It is ductile and readily fabricated and can be welded using electrodes of the same composition. It is available commercially in a variety of product forms. The elevated behavior of this steel has been studied more thoroughly than that of any other Cr-Mo steels [29]. Additional creep strength and oxidation resistance can be provided by increasing the Cr content. Alloys containing 5%, 9%, and 12% Cr have been used for nuclear industry superheaters, usually where the oxidation or creep strength of 2-1/4Cr-1Mo is not adequate.

### **3.2.2 Modified 9Cr-1Mo Steel**

9Cr-1Mo steel has been used for superheaters in both coal- and oil-fired stations. The vanadium-niobium-modified 9Cr-1Mo (Fe-0.1C-9Cr-1Mo-0.5Mn-0.4Si-0.2V-0.08Nb- $<0.2\text{Ni}$ ) was developed by Oak Ridge National Laboratory and has greater strength than 2-1/4Cr-1Mo at elevated temperatures. The modified 9Cr-1Mo steel, previously covered in Code Case 1943 of the ASME Boiler Code, now is covered in the regular code. The alloy is designated ASTM grade 91 and is specified in ASTM standards for boiler tubes (ASTM A 213), forgings (ASTM A336), seamless pipe (ASTM A335), and forged and bored pipe (ASTM A369). The corrosion behavior of this alloy in Pb-Bi eutectic is not known.

### **3.2.3 12Cr-1Mo Steel**

This steel has the highest creep strength and oxidation resistance of the Cr-Mo steels. It does, however, present some problems in fabrication, and it requires special precautions. The application of this steel in the U.S. has been hindered by its relatively difficult welding characteristics [29]. It is not a listed material in the ASME Boiler and Pressure Vessel Code.

HT-9 (Fe-12Cr-1Mo-0.2C-0.4Si-0.5Ni-0.5Mn-0.3V-0.5W-0.02P-0.01S) is a brand name of Sandvik of Sweden. HT-9 is widely used for boiler tubing in Europe, but it is not an ASME Code material and therefore is not available commercially in the U.S. at present.

#### 4. Other Low-Melting-Point Alloys

It is desirable, from a materials standpoint, to reduce the operating temperatures of the target region of the ABC/ATW, and this can be achieved by employing less-polonium-producing and less-corrosive low-melting-point alloy targets. Lead-magnesium eutectic (98Pb-2Mg) has a melting point of 248.7°C. The addition of 2 wt% Mg to Pb increased the severity of corrosion of Croloy 2-1/4, and uninhibited Pb-Mg eutectic cannot be contained in Croloy 2-1/4 [16]. Zr inhibitor was added to the eutectic but would not go into solution, which indicated low solubility. The corrosion experiment results of this alloy are shown in Table 6. Lead-tin eutectic (38.1Pb-61.9Sn) has a melting point of 183°C. The neutronic analysis for the ABC/ATW system showed that this alloy does not produce any serious Po or toxic daughter material, so it is considered as a coolant material to reduce the temperature of the proton window for the liquid-lead target. No data have been found on corrosion behavior of this alloy in Pb-Bi eutectic.

Table 6. Corrosion of Croloy 2-1/4 in lead-magnesium eutectic [25].

Hours to Severe Corrosion	Temperature Range (°C)
<1000	488-550
1000-5000	510-550

Alloys with melting points below the boiling point of water are known as fusible alloys. These are used for binding plugs in automatic sprinkler systems, low-temperature boiler plugs, solder for pewter and other soft metals, for tube bending, and for casting patterns for many ornamental articles and toys. Their compositions and melting points are shown in Table 7. Unfortunately, all of these alloys contain Po-producing bismuth, and it appears that there are not many advantages to using them.

Table 7. Alloys with melting points below 100°C.

Alloy	Composition				Melting point (°C)
	Pb	Bi	Sn	Cd	
Original Newton's alloy	31.25	50	18.75	—	92
Newton's metal	—	50	25	25	95
Wood's metal	25	50	12.5	12.5	70
D'Arcet's alloy	25	50	25	—	93
Lichtenberg's alloy	30	50	20	—	92
Malotte's metal	20	46	34	—	95

## 5. Conclusions

For the lead target, only refractory alloys of Nb, Mo, Ta, and W are worth consideration because of its high operating temperature. Each of these materials has its own disadvantages: Nb and Ta are easily oxidized; Mo becomes too brittle after proton irradiation; Ta and W have high-thermal-neutron-absorption cross sections; and W is very difficult to fabricate. Based on the assumption that all the oxidation phenomena results in thickness loss from the surface, thickness loss of Nb-1Zr in various environments were calculated. The results showed that uncoated Nb-1Zr may be used for one year at 900°C at  $P_{O_2} = 1 \times 10^{-6}$  torr, but the same material may not be used in argon with 5-ppm oxygen. At  $P_{O_2} = 1 \times 10^{-7}$  torr at 900°C, the thickness loss due to oxidation for Nb-1Zr does not seem to be a problem. Further study of the internal oxidation and hydrogen embrittlement of Nb-1Zr is required. For the lead-bismuth eutectic target, three candidate container materials were selected based on a literature survey. These are Croloy 2-1/4, modified 9Cr-1Mo, and HT-9. These materials may be used only if the lead-bismuth is thoroughly de-oxidized and treated with Zr or Mg inhibitor. The interaction between the spallation products and the inhibitors needs to be studied. The corrosion resistance of these Cr-Mo steels to the eutectic will be improved by nitriding.

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