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MERCURY AS A TARGET MATERIAL FOR PULSED (FAST) SPALLATION NEUTRON SOURCES SYSTEMS

Günter S. Bauer

Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland

ABSTRACT

For spallation neutron systems which are not adversely affected by the high thermal absorption cross section, mercury seems to be a good target a material because it is liquid at ambient temperatures, shows good compatibility with low nickel content steels, has a high neutron yield, is easy to purify and has no truly long lived isotopes. Its performance in an engineered target for pulsed spallation sources is expected to be superior to that of its solid competitiors Ta and W and equivalent to that of Pb or Pb-Bi but at much less overall technical effort.

1. Introduction

Using a flowing, molten, heavy metal as target material has been considered at least as an option for all medium-to-high-power spallation neutron sources studied in the past [1], [2], [3], [4] The obvious advantages are

- a high heat removal capacity by convective flow of the heated material as a whole
- a high average density of the target material in the beam interaction zone, giving a high source brightness
- no moderation of the primary neutron spectrum inside the target, reducing the likelihood of resonant absorption and resulting in a high reflector flux and good neutronic moderator coupling
- no water present in the proton beam interaction zone, reducing radiolysis and radioactivity in the cooling water circuit substantially
- no structural radiation damage in the target material, making regular replacement necessary only for the part of the target shell exposed to high particle flux

Keywords: Mercury, Liquid Metal, Vapour Pressure, Solubility

- the content of hazardous volatile species in the target material that could be released in an accident scenario is low because they will escape from the target continuously and be safely removed on line.

Desirable properties of a material to be used as liquid target for a spallation neutron source are:

- a high atomic number to give good neutron yield
- a high density to give high source brightness
- a low melting point to make choices for structural materials and operation easier
- a low corrosive activity (solubility of elements of the container material) to avoid problems with mass transport from hot to cooler parts of the system
- chemical inertness to minimize formation of possibly abrasive compounds that would be transported in the system
- a low neutron resonance integral to minimize absorption of neutrons before they get thermalized
- a high scattering cross section for fast and epithermal neutrons to improve reflector action of the target
- a low thermal neutron absorption cross section, at least if the target is to feed a neutron source which builds up a high thermal flux through a long life time of the neutrons in the reflector.

Table 1 gives an overview of the candidate material and their properties.

Table 1: Liquid Metal Target Candidate Materials and their Relevant Properties in Comparison

Property	Pb	Bi	LME	LBE	Hg
Composition	elemental	elemental	97.5%Pb +2.5%Mg	45% Pb +55%Bi	elemental
Atomic Number Z	82	83			80
Atomic Mass A (g/mole)	207.2	209			200.6
Density (g/cm^3) solid (20°C)	11.35	9.75			
liquid	10.7	10.07	10.6	10.5	13.55
Coefficient of thermal expansion (K^{-1})	$2.91 \cdot 10^{-5}$	$1.75 \cdot 10^{-5}$			$6.1 \cdot 10^{-5}$
Contraction on solidification (%)	3.32	-3.35	≈0	≈3.6	
Melting point (°C)	327.5	271.3	250	125	-38.87
Boiling point at 1 atm. (°C)	1740	1560			356.58
Specific heat capacity (J/g/K)	0.14	0.15	0.15	0.15	0.12
Thermal neutron absorption (barn)	0.17	$3.4 \cdot 10^{-2}$	0.17	0.11	389
Corrosivity	moderate	highest	moderate	high	low
Chemical activity	moderate	highest	moderate	high	low

Since all neutron sources that considered liquid metal targets so far were conceived with a large and non-absorbing reflector to produce a high time average thermal neutron flux, mercury was excluded as a potential candidate because of its high thermal neutron absorption cross section. If, however, thermal neutron absorption is not a primary concern - or is even a desirable property to ensure proper functioning of the system, as in short pulse spallation sources, pulse shape optimized intermediate pulse length sources or fast neutron systems involving fissionable material, mercury certainly deserves special attention.

2. Neutronic properties of mercury

In order to get a full picture of the neutronic performance of a spallation target, it is necessary to perform Monte Carlo calculations, but certain general aspects can also be derived from rules-of-thumb and other known facts. As noted before, mercury, in its natural isotopic composition, has a high thermal neutron absorption cross section, which is mainly due to the isotope of mass number 199, which accounts for 367 of the total of 389 barn (Table 2). Another 13 barn are due to the isotope of mass number 200. While one might expect the isotopes with high neutron absorption cross section to "burn" in a high thermal neutron flux, this will not play a significant role in a liquid metal spallation target, because only a small fraction of the roughly 1 m^3 of mercury in the circuit will be exposed to the neutrons at any moment.

Table 2: Thermal neutron properties of Hg and its stable isotopes

Thermal neutron absorption cross section			389 barn
Resonance integral			75 barn
Scattering cross section		bound	26.3 barn
		epith.	15 barn
Neutron mean free path:			
		scattering	0.93 cm
		thermal neutron absorption	0.06 cm
Stable Isotopes			
Mass	Abundance (a/o)	Therm. neutr. absorption @ 2200 m/s	
196	0.15	3080 barn	
198	10.1	1.9 barn	
199	17	2162 barn	
200	23.3	60 barn	
201	13.2	8 barn	
202	29.6	4.9 barn	
204	6.7	0.4 barn	
all	100	389 barn	

On the other hand, the high thermal neutron absorption is not necessarily an undesirable property in a pulsed spallation neutron source: In order to keep thermal and cold neutron pulses short, the moderators are normally surrounded by a material which is transparent for epithermal and fast neutrons and highly absorbing for slow neutrons, in order to prevent slow neutrons from returning to the moderator after rattling around in the reflector for some time. Such a material is boron. The problem with boron at high neutron flux is that it emits an α-

particle of short range and therefore heats up substantially, requiring intensive cooling. In Fig. 1 the thermal neutron absorption cross sections of boron and mercury are compared to one another. Up to about 200 meV the cross section of Hg is less than a factor of 2 lower. The mean free path of thermal neutrons in Hg is about 0.06 cm, compared to 0.04 in B_4C and 0.025 cm in BN. Other boron-containing materials such as Boral have a much longer mean free path. It is also important to note that the ratio of the mean free paths for absorption and for scattering is 0.065, which shows that mercury is indeed a good "decoupler" for thermal neutrons. The absorption cross section vanishes at about 10 eV. In order to achieve a short pulse throughout the range of interest for an ambient temperature moderator, decoupling becomes important below about 0.4 eV. It is obvious that mercury should serve well in the whole regime, whereas boron tends to reduce also the intensity of higher energy neutrons, which could still be accepted as return flux from the "reflector".

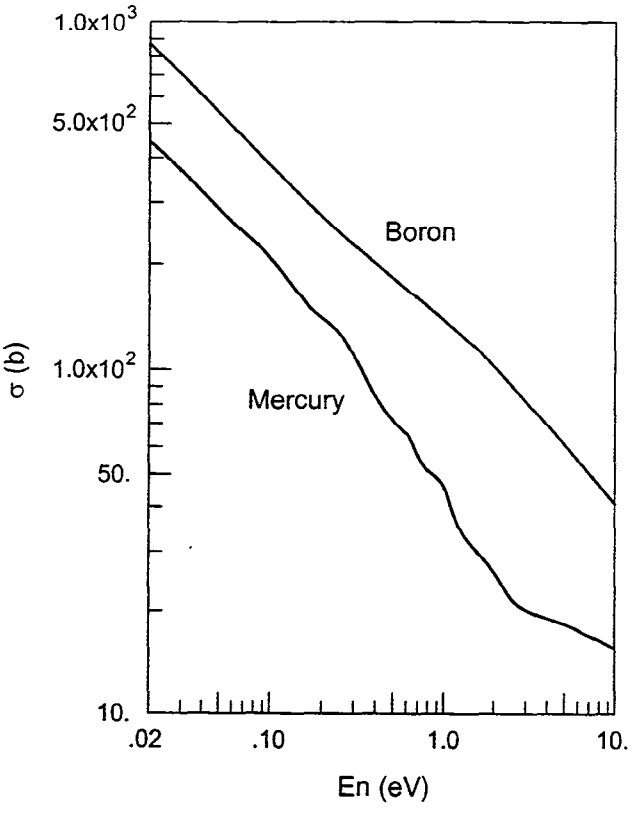

Figure 1: Thermal neutron total cross section of mercury and boron

In Table 3 a few data for the high energy regime are given. The range of 1.35 GeV protons, the neutron yield and the mean free path between inelastic collisions were calculated by the relations given by Carpenter [10]. It can be seen that almost no slowing-down of fast neutrons takes place in a Hg-target because the mean free path between elastic collisions is about 2.5 cm in the keV-MeV range and 1800 collisions would be required for complete thermalization. We may therefore expect the leakage spectrum from a mercury target to be harder than that from a H_2O- or even D_2O-cooled Ta or W-target, where the volume fraction of coolant would have to be of the order of 30 ÷ 50 % at ESS-power densities of up to 3 kW/cm^3.

Table 3: Some high energy neutron data for mercury

Range of 1.35 GeV protons	70 cm
Approx. mean free path between inelastic collisions	15 cm
Approx. neutron yield at 1.35 GeV from spallation (20 cm diam. target)	27 n/p
Slowing down properties for neutrons:	
Log. enegy decrement per collision	0.01
Nr of collisions 2 MeV -> 25 meV	1832

This may have some effect on n-2n or n-3n processes in some reflector materials. Or, conversely, reflector materials containing elements with high n-2n, n-3n cross section and low threshold energies should be particularly desirable.

Table 4: Radioactive isotopes of mercury and their half lives

Radioactive isotopes	mass	T 1/2	Comments
from n-capture (ß-active)			
	203	46.59 d	
	205	5.2 min	
	(206)	8.15 min	Unlikely to be
	(207)	2.9 min	produced
from spallation			
	197	64.1 h	Where two decay modes
	195	40 h	exist, the longer half life
	194	**367 a**	is given
	193	17.65 h	
	192	5 h	
	191	3.18 h	
	190	42.8 m	
	189	8.7 min	
	188	8.8 min	
	187	2.4 min	
	186	1.4 min	
Hg<186 have half lives less than some alpha-activity		1 min	

Listed in Table 4 are the radioactive isotopes of Hg, together with their half lives. Radioactive isotopes can be generated by neutron capture or in the spallation process. With one exception, all radioactive isotopes decay with half lives less than 50 hours, most of them significantly less. While this may not be very significant for handling and maintenance work during the service time of the target material, it may well be significant when it comes to final disposal. Since Hg is relatively easy to purify by chemo-physical means, all the non-Hg radioactive spallation products can be separated off and disposed of separately. The remaining mercury will be of very low specific activity and have negligible surface dose after a year or so, because only Hg-194 will be left as β^+-emitter. It is of particular significance to the general risk perception, that there is no polonium production, in particular no ^{210}Po (α-emitter with

half life of 138.4 days) to be expected in a mercury target. Some α-active isotopes of Hg, Au, Pt etc. may be generated during target operation, but they all have half lives well below 1 minute.

3. Physical and chemical properties of mercury

Since, obviously, from a neutronic point of view, mercury is a very promising candidate for fast neutron spallation systems, the next question is about its physical and chemical properties.

Listed in Table 5 are the relevant physical properties for the design and operation of a mercury spallation target. The most important feature, of course, is the fact that Hg is liquid at room temperature, thus avoiding the need for auxiliary heating to keep the metal molten even if the source is not in operation. This is important for two reasons: a) it takes a long time to safely melt a large mass of the order of 10 tons in a complicated target circuit with pumps, heat exchangers, diagnostics equipment etc. and b) for all other candidates listed in Table 1, there is either a large volume change at the solid-liquid phase transition, or a long term expansion after solidification (case of Pb-Bi). In both cases the risk of undesirable mechanical load on circuit components exists (enclosure in the case of expansion, internal structures in the case of contraction) and would require complicated heating systems and process protocols for solidification and melting in order to avoid damage.

A potential problem could arise from the high vapour pressure of mercury, shown in Fig. 2. Certain materials considerations may require operating temperatures of a few hundred degrees [11], which will require a system pressure of several bars, in particular if mechanical pumps are to be used and cavitation effects must be avoided. On the other hand care must be taken to avoid local overheating to more than about 500 °C, if the system pressure is to be kept at 10 bars or below[1].

Hg is known to have a very high surface tension and to form tiny droplets when spilled. This might require special care when the liquid metal circuit must be emptied for repair or maintenance purposes. On the other hand the surface tension seems to depend strongly on the degree of purity of the mercury. Also, opportunities exist to provide cooling for parts of the circuit near the most likely positions which have to be opened (e.g. the target shell) to less than -40°C and thus solidify any Hg-residues for the period when the circuit is open. The high thermal expansion coefficient of Hg will require a sufficiently large ullage tank to accommodate the volume change upon heating to operating temperature. It also has the effect of generating a large (hindered) expansion during each power pulse of the beam in a pulsed system. Fortunately, Hg also has a relatively high compressibility, which helps to keep the

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[1] Note: The high vapour pressure in principle opens up the interesting opportunity to design an isothermal target cirucit with heat removal based on evaporation of the target material. Mercury has been widely used in this mode in diffusion pumps in the past. This would, in view of the rather high latent heat of vaporisation (almost 300 J/g) not only mean that only a small amount of the Hg is in circulation (10 kg or 0.8 litres per second would have to be evaporated for a 5 MW beam depositing 3 MW in the target), it would also minimize thermal and static mechanical stress on the target and make removal of gaseous products straight forward. In view of the uncertainties related to the pulsed nature of the power deposition in ESS, this is not considered, for the time being. It would, however, be an interesting option for a large cw fast neutron system, where otherwise very large mass flows of liquid metals are necessary to remove the heat. In such systems direct use of the Hg-vapour for energy conversion could be an option. Hg-gas turbines have already been built and operated sucessfully.

pressure level that builds up in the liquid in the same regime as for other target materials such as lead. The problem of pressure waves and their effect on the container material is common to all liquid metal targets in pulsed operation [5] and has been discussed elsewhere [6].

Table 5: Physical properties of mercury

Atomic number		80	
Atomic weight		200.61	
Density	°C	g/cm^3	
	20	13.55	
	100	13.35	
	200	13.12	
	300	12.88	
	350	12.76	
Coefficient of thermal expansion	(0-100°C)	61 E^{-6}	K^{-1}
Melting point		-38.87	°C
Boiling point (1 atm)		357	°C
Critical temperature		1477	°C
Vapour pressure (298-630 K),mm Hg log p = -3308/T+10,373-0,8 log T			
	mm Hg	°C	
	1	126.2	
	10	184	
	40	228.8	
	100	261.7	
	400	323	
Latent heat of fusion		11.72	J/g
Latent heat of vaporisation		291.82	J/g
Specific heat	°C	J/g/°C	
	0	0.1396	
	100	0.1373	
	200	0.1359	
	300	0.1354	
	450	0.1363	
Thermal conductivity	°C	J/cm/sec/°C	
	0	0.0821	
	60	0.0967	
	120	0.1093	
	160	0.1168	
	220	0.1269	
Surface tension	°C	N/cm	
	20	0.00465	
	100	0.00456	
	200	0.00436	
	300	0.00405	
	350	0.00395	

Table 5 cont.

Viscosity	°C	centipoise	
	20	1.554	
	100	1.24	
	200	1.032	
	300	0.95	
	350	0.914	
Compressibility		$3{,}6 \cdot 10^{-11}$	m^2/N
Electrical resistivity	°C	microhm*cm	
	50	98.4	
	100	103.2	
	200	114.2	
	300	127.5	
	350	135.5	
Coefficient of resistivity (0-100°C)		$0{,}9 \cdot 10^{-3}$	K^{-1}

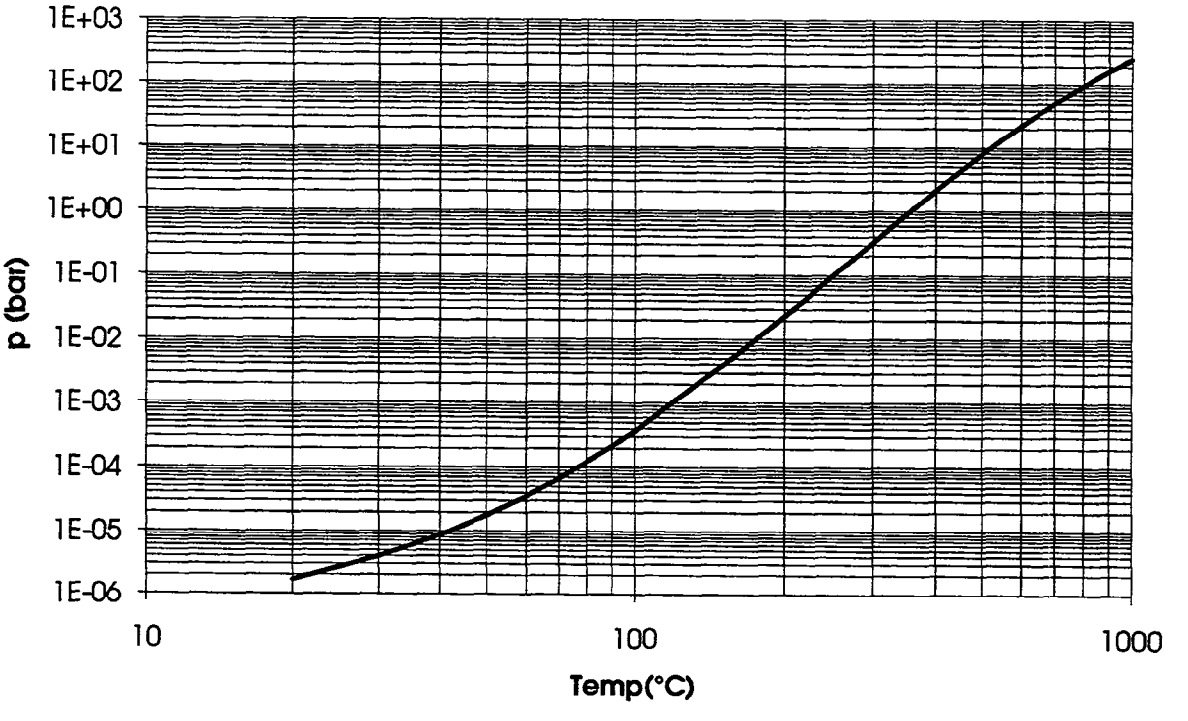

Figure 2: Vapour pressure of mercury

The high vapour pressure of Hg has yet another consequence: While Hg, as a spallation product, would evaporate from any other candidate liquid metal target circuit (based on Pb and/or Bi), all non-gaseous elements are expected to remain in the mercury. This can be seen from Fig. 3, where the calculated partial molar enthalpies of evaporation are given and compared to those at infinite dilution in a Hg-bath [7]. Obviously, there is no element with a lower enthalpy of evaporation than Hg itself.

Another question of interest is the chemical behaviour of the spallation products generated in the mercury. Some intermetallic or metal-non metal compounds can be quite abrasive and produce damage when swept across the target window at high velocity. Also, it is important to know whether such compounds or the pure elements would remain in solution or segregate to certain regions of the circuit (e.g. the heat exchanger). As a first step towards an assessment of these problems, the enthalpies of formation of intermetallic compounds with Hg were examined [8]. Some results are shown in Fig. 4. Negative values of ΔH_f show a

tendency to form amalgams, whereas elements for which ΔH_f is positive will segregate out if their limit of solubility is exceeded. Prominent examples of the latter group are the heavy 3d-metals Mo and Ta and the elements of the iron group (i.e. the main constituents of steels).

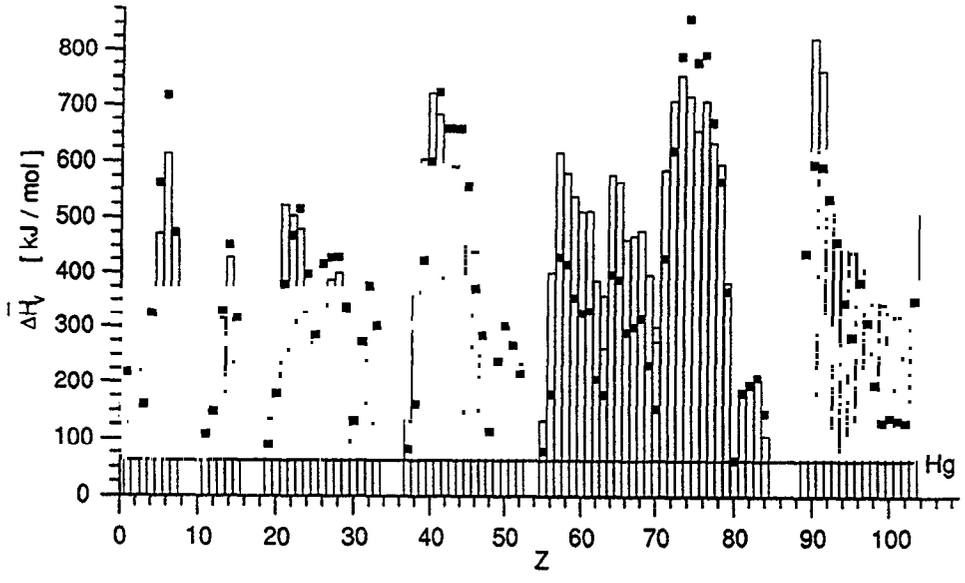

Figure 3: Calculated molar enthalpies of vaporization of the pure elements (black dots) and in infinite dilution in mercury (open bars).

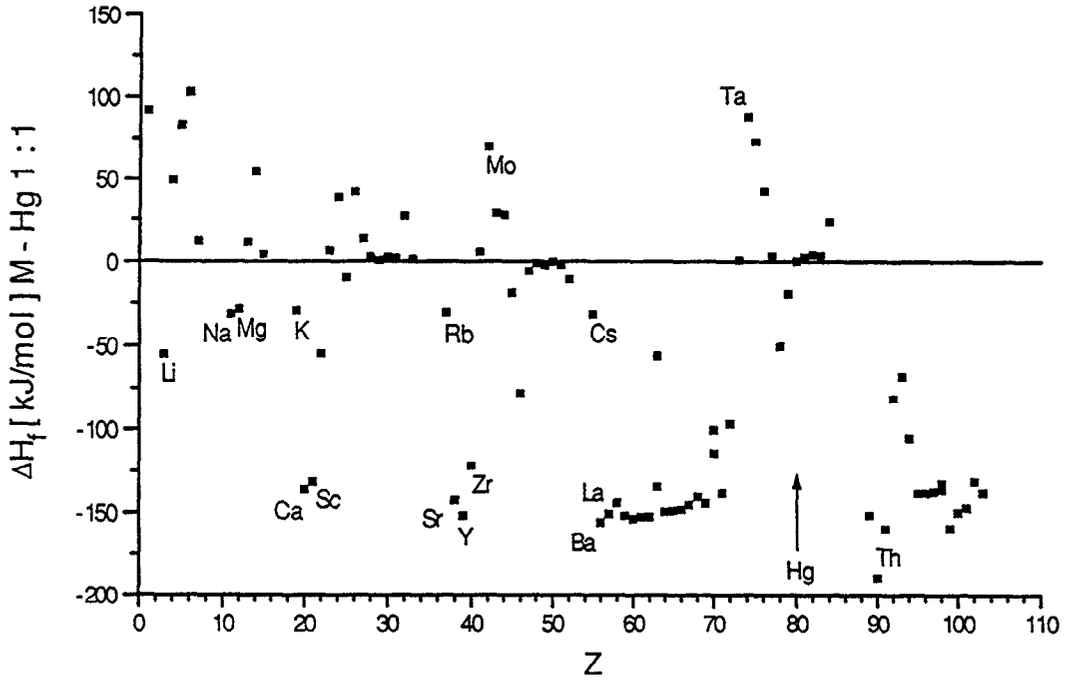

Figure 4: Enthalpy of formation of intermetallic compounds M-Hg

The solubility of non-amalgamating elements in Hg, in particular those which are components of the container material, is important because liquid metal corrosion very often results from dissolution of parts of the container material at the hot parts of the system and segregation at the cooler parts (this is one of the reasons, why too large temperature differences in the

system should be avoided). For the most important elements, Fe, Cr, V, Ni and Ti, experimental data have been compiled [8] and are shown in Fig. 5. As a general feature, solubilities in Hg are about 2 orders of magnitude lower than in Bi (and about 1 order of magnitude lower than in Pb). As in other liquid metals, Ni has the highest solubility of the ones shown. While perhaps not a problem for reasons of corrosion in Hg, Ni should be avoided in the container material also because of its He-production under slow neutron irradiation. Low nickel content or nickel-free steels are therefore the preferred container material for liquid mercury spallation targets for the time being, but other favourable options may exist and further research is needed also in this respect.

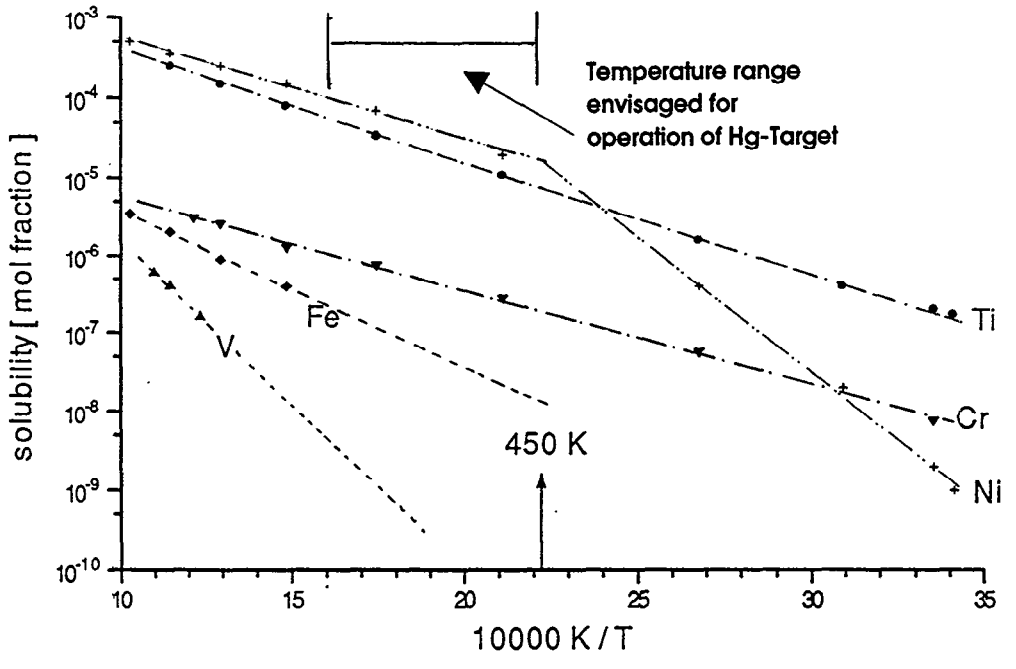

Figure 5: Solubility of metals Ti, V, Cr. Fe, Ni in Hg (exp. data); lines are guides to the eye only.

It has also been pointed out [9] that attack of low carbon steels by mercury can be inhibited by small additions of Ti and Zr (at a level of about 1 to 10 ppm) due to the formation of insoluble titanium or zirconium carbide-nitride films on the surface of the steel. Furthermore, addition of about 50 ppm of Mg has been recommended to (a) improve the wetting properties of the mercury by removing the oxide layer on the steels and thus improve heat transfer and (b) prevent the inhibitors from being removed from the Hg by oxidation.

Finally, it should be noted that mercury does not react with water, an important feature if a leak springs between the mercury circuit and a possible secondary water cooling loop. On the contrary, water is used in the purification process to obtain high purity mercury. Since, as mentioned before, purification of mercury may be desirable not only during operation of a mercury target, but, in particular, also at the end of its service life, we briefly quote the procedure used [9]: To purify mercury it is first treated in a stoneware crock with diluted nitric acid (1 part and in 4 parts water) accompanied by vigorously bubbling air up through the mercury. This is followed by thorough washing with water to remove the residual acid. The noble metals are then removed by destillation (evaporation) of the mercury under reduced pressure at 250 °C. Impurities not left behind in the destillation process will float on the mercury after a period of standing and are removed by filtering through Pyrex glass wool. The purity of the mercury obtained in this way is between 99.99998 and 99.99999 %.

4. Conclusions

Although the present assessment is far from complete, a large number of details need still to be looked at and several technical problems must still be solved, mercury seems to be a promising candidate target material, avoiding many of the problems associated with water cooled solid targets and relaxing the technical efforts and risk potential particular to other heavy liquid metal options, especially those containing bismuth. Presently no basic problems can be seen that would impede the implementation and safe operation of a target system based on liquid mercury. By developing such a system, the route might be opened up to spallation neutron sources well beyond the 5 MW level of the current European Spallation Source (ESS) project, for which a mercury target has now been adopted as the first priority concept. Also accelerator-based fast neutron systems for nuclear power might profit from the use of mercury as spallation target material.

5. Acknowledgment

Mercury was suggested for consideration as a target material for pulsed spallation sources during a workshop held at PSI in 1992 in the group chaired by B. Sigg. Subsequently, many discussions with my colleagues at PSI and elsewhere helped to clearify the picture and lead to the final publication of this article. The work was performed with support from the Swiss Bundesamt für Bildung und Wissenschaft (BBW) under contract Nr. 94.0134 in the frame of a contribution to the ESS-project study funded in part through the EU-Human Capital and Mobility program, Contract Nr. CHRX-CT94-0503.

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