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Gallium-Cooled Target for Compact Accelerator-Based Neutron Sources

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Abstract

This paper discusses the motivation for gallium cooling of targets of compact accelerator-based neutron sources (CANS); summarizes features of the low-power alternative, i.e., water cooling, and the limitations of boiling water heat transfer; lists the properties of liquid gallium; and cites its low hazards potential. I set out working equations for heat transport and fluid flow in liquid gallium and present a concept for a gallium-cooled system, including a scoping calculation of temperatures and pressure drops, and present conclusions and a recommendation.

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1. Introduction

Recent interest in compact, low-power, low-energy-particle accelerator-based neutron sources (CANS) comes about in response to the need for slow neutron sources that are relatively inexpensive to build and operate and suitable for university and industrial locations. These provide for scientific applications less demanding of neutron flux than the high-power sources and for tuneup experiments to be performed at the large sources. They can serve as test-beds for developing new techniques and components for use in high-power installations and are safe and convenient venues for training and education where learning, not time, is of the essence. Moreover, recycled components and infrastructure from abandoned facilities

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are often adaptable to CANS concepts. The enthusiastic attendance at this meeting and the recent formation of the UCANS collaboration attest to this interest.

The central features of these sources, namely low particle energy and modest beam power, nevertheless lead to high power densities in the target and yet demand low-cost, simple cooling systems. The coolant of choice is water, but heat fluxes at the cooled target surface in these sources can approach or exceed the maximum values feasible with water, the critical heat flux (CHF) nucleate boiling limitation (about 300 W/cm², although this can be exceeded using extreme measures).

Liquid metal coolants offer prospects for higher heat fluxes than water, but the most common ones suffer drawbacks of flammability (alkali metals and their eutectics), high melting temperatures (Pb, Bi, Pb-Bi eutectic, Sn, Pb-Sn—i.e., solder—and Bi-Sn alloys), radioactivation (Cd, In, Hg, Bi), and toxicity (Cd, Hg). Almost uniquely, gallium offers the benefits and lacks the drawbacks of other liquid metal coolants for these applications. Its low vapor pressure and high boiling temperature imply that liquid gallium remains in single phase even at elevated temperatures. Its activation products have short half-lives. Moreover, gallium is liquid at temperatures above 30°C: melts in your hand, solid on the floor. Fifty-fifty gallium-tin alloy, i.e., GaSn, an alternative to pure gallium, melts at 25°C.

2. Water cooling

Heat transfer by flowing water (forced convection) is the “engineers’” choice for most applications. Figure 1a [1] shows the heat flux as a function of the temperature difference between the heated wall and the saturation (boiling at prevailing pressure) temperature of the bulk fluid.

![Fig 1a Low-temperature convective heat transfer to water](image-url)
Figure 1b. General heat transfer to water. a-b, single phase liquid water; b-c, nucleate boiling; c-d, mixed; d-e, film boiling; e-f, film boiling and radiation. [2]
At low wall temperatures, the water is in liquid phase. At high wall temperatures, steam bubbles form at the surface (two-phase flow) and the heat flux increases rapidly with temperature. The heat flux increases as the wall temperature increases and as the flow velocity increases, ultimately approaching an asymptote independent of flow velocity. Figure 1b [2] shows that this trend does not continue indefinitely. The region a-b-c is the practical regime illustrated in Fig. 1a. The heat flux at c is the critical heat flux, beyond which the heat flow is unstable, vapor blankets the heated surface, and temperatures jump to very large values at c’, governed by film boiling and radiation heat transfer, called burnout. The critical heat flux is the maximum feasible for given flow conditions. In extreme circumstances, $q''_{\text{max}}$ can be as large as 1.0 kW/cm², but in practice is 300-500 W/cm². The figures use English engineering units: $10^6 \text{ Btu/hr-ft}^2 = 315 \text{ W/cm}^2$, $1.8^\circ\text{F} = 1^\circ\text{C}$.

3. Properties of gallium

Physical properties
- Atomic number $Z = 31$.
- Mass number $A = 69$.
- Stable isotopes Ga69 (60.1%), Ga71 (39.9%).
- Melting temperature $T_m = 29.8^\circ\text{C}$.
- Boiling temperature $T_b = 2403^\circ\text{C}$.
- Vapor pressure [3] less than 10-19 Pa at temperatures below 500°C.
- Heat of fusion $Q_f = 5.59 \text{ kJ/mol}$.
- Density [4] @ 30°C, = 6.095 g/cm³.
- Thermal conductivity $k = 0.406 \text{ W/cm} \cdot ^\circ\text{C}$.
- Specific heat $C_p = 0.37 \text{ J/gm} \cdot ^\circ\text{C}$.
- Viscosity = 0.019 poise = 0.019 g/cm-s.
- Price @ 99.9999% purity $\$500/\text{kg}$.

Most data above from [5].

Nuclear properties [6]
- Thermal neutron capture crosssection (2200 m/sec) = 2.9 b/atom
- Neutron activation crosssections and products
  - $\sigma_{n,\gamma}$ (2200 m/s, Ga69) = 1.7 b/atom; resonance capture integral = 14 b/atom; T1/2 (Ga70) = 21 min.
  - $\sigma_{n,\gamma}$ (2200 m/s, Ga71) = 4.8 b/atom; resonance capture integral = 31 b/atom; T1/2 (Ga72) = 14 hr.

Hazards of gallium
- The Materials Safety Data Sheet for gallium and gallium-containing alloys indicates caution in handling (use gloves) and avoidance of inhalation of powder and contact with eyes (may cause irritation).
- Liquid gallium metal is corrosive against many metals, notably aluminum, but compatible with stainless steels, titanium, beryllium, water, and rubber-like elastomers.
- Bulk gallium metal is not flammable. A general appraisal of personal hazards is that gallium is, in most ways, benign.

A useful treatise on liquid-metal/materials compatibility is the Kelman compilation [7].

Heat transport and fluid flow
- Heat transport from a heated surface to flowing liquid involves numerous defined dimensionless quantities and empirical data correlations: [2,8].
The (dimensionless) Prandtl number (a property of the fluid only) is

\[ Pr = \frac{C_p \eta}{k} \]  

For liquid gallium at about 30°C, the Prandtl number is

\[ Pr = (0.37)(0.019)/0.406 = 0.0173. \]  

The (dimensionless) Nusselt number, \( Nu \), so that

\[ Nu = \frac{hD_e}{k} \]

where the equivalent diameter of the channel, \( D_e \), is

\[ D_e = \frac{4(\text{flow cross sectional area})}{(\text{wetted perimeter of the flow channel})}, \]

\( k \) = thermal conductivity,

and \( h \) is the convective heat transfer coefficient.

The heat flux \( q'' \) is

\[ q'' = h\Delta T \]

where \( \Delta T \) is the difference of temperature between the heated surface and the bulk fluid.

Seban’s correlation for flow between heated plates gives the Nusselt number for situations like CANS targets with liquid metal coolants

\[ Nu = 5.8 + 0.02 Pe^{0.8} \]

where \( Pe \) is the Peclet number,

\[ Pe = RePr \]

and \( Re \) is the (dimensionless) Reynolds number.
\[ Re = \frac{vD \rho}{\eta} \]  

(7)

in which
- \( v \) is the bulk-average flow velocity,
- \( \rho \) is the density, and
- \( \eta \) is the viscosity of the liquid.

Accordingly, quiescent liquid metals offer significant heat transfer coefficients even in the absence of flow (\( Re = 0 \)) because of the high thermal conductivity of liquid metals compared to nonmetallic liquids.

[The Dittus-Boelter Correlation gives the Nusselt number for single-phase heat transfer from a heated surface to flowing liquid (quoted just for reference—it applies only for nonmetallic coolants like water),

\[ Nu = 0.023 Re^{0.8}Pr^n, \]  

(8)

and for heat flow from the heated surface into the fluid, \( n = 0.4 \); for heat flow out of the fluid into a cooled surface, \( n = 0.3 \).]

The Darcy equation gives the pressure drop \( \Delta p \) between the ends of a channel of constant cross section

\[ \Delta p = f \left( \frac{L}{D_e} \right) \rho \frac{v^2}{2} \]  

(9)

where \( f \) is the Darcy-Weisbach friction factor, which depends on the roughness of the channel walls and the Reynolds number, usually taken from the Moody Friction Factor Chart. Smooth channels are the usual case, and for large Reynolds numbers (\( Re > 2 \times 10^4 \)),

\[ f = 0.184/Re^{0.2}. \]  

(10)

4. A calculation

Here I provide a preliminary evaluation of a gallium-cooled CANS system based on an analytical formalism. Detailed design will require modern numerical thermal-hydraulic analysis.

The temperature of the heated surface is the critical quantity for CANS targets. Beryllium is the target material and, in the absence of other considerations, can withstand rather high temperatures. The average heat flux, assuming 20 kW over an 8-cm diameter, is

\[ q'' = \frac{20000}{[(\rho/4)(82)]} = 398. \text{ W/cm}^2, \]  

(11)

but the peak heat flux may be about twice that because the proton beam may be non-uniform, so I calculate for

\[ q_{\text{peak}}'' = 800 \text{ W/cm}^2. \]  

(12)
(LENS employs nonlinear optics in the proton beam line to flatten the power density distribution.) Before launching detailed design analysis, it will be necessary to obtain a realistic beam power distribution from which to determine the peak-to-average power ratio. The thickness of the beryllium target is less than the range of protons in the material so that most of the protons do not stop in the target, where they would eventually build up and cause blistering, destroying the target plate. Moreover, the heat deposited in the target plate is less than the beam power, so needs to be calculated in the process of cooling system analysis. Protons that stop in the gallium coolant are expected not to cause problems.

I assume that the flow channel is of rectangular cross section 8 cm wide and 2 mm thick, so that the equivalent diameter is

\[ D_e = \frac{4 \times (8) \times (0.2)}{2 \times (8.2)} = 0.390 \text{ cm} \]  

(13)

and the flow cross sectional area is

\[ A = 8 \times 0.2 = 1.6 \text{ cm}^2, \]  

(14)

as shown in Fig. 2

![Fig 2. Schematic diagram of a gallium-cooled target](image-url)
Let the flow velocity be $v = 1 \text{ m/sec}$. Then the Reynolds number is

$$\text{Re} = (100)(.390)(6.095)/(0.019) = 12511. \quad (15)$$

The Peclet number is

$$\text{Pe} = (0.0173)(12511) = 216.4, \quad (16)$$

the Nusselt number is

$$\text{Nu} = 5.8 + .02(216.40.8) = 7.28. \quad (17)$$

and the heat transfer coefficient is

$$h = (0.406)(7.28)/(0.390) = 7.58 \text{ W/cm}^2\text{-°C.} \quad (18)$$

Then the wall-fluid temperature difference is

$$= 800/7.58 = 105.6 \text{ °C}, \quad (19)$$

which is an acceptable figure for the gallium-cooled system.

Figure 3 schematically illustrates a gallium-cooled target system. The volume flow rate is

$$V' = vA = (100)(8)(.2) = 160 \text{ cm}^3/\text{s}. \quad (20)$$

The frictional pressure drop through the target region, which I assume to be 10 cm along the flow path, is

$$\Delta p = (0.028)(10/0.390)(6.095)(1002)/2 = 2.188 \times 104 \text{ g-cm/s2/cm2} =
= 2.188 \times 103 \text{ kg-m/s2/m2} =
= 2.188 \times 103 \text{ N/m2} \approx 0.022 \text{ atm}, \quad (21)$$

which is practically negligible.

Figure 3. Schematic illustration of a gallium-cooled target system.
The temperature rise in the target channel is
\[
\delta T = \frac{20000}{(6.095)(0.37)(160)} = 55^\circ C, \quad (22)
\]
which is an acceptable figure.

I assume the external circuit to consist of D = 2-cm diameter tubing (for a circular tube, De = D) L = 5 m long. The flow velocity in the tube is
\[
v_t = \frac{V'}{\left( \frac{D^2}{4} \right)} \quad (23)
\]

The frictional pressure drop for the external loop depends on the Reynolds number for the tube,

\[
Re = \frac{(50.9)(2.0)(6.095)/0.019}{3.266 \times 10^4}, \quad (24)
\]

for which the friction factor is \( f = 0.023 \), and the frictional pressure drop is

\[
\Delta p_{friction} = 0.023 \times 500/1.0 \times 9.08 \times 10^4 \times 105 \text{ g-cm/s2/cm2} = 9.08 \times 10^4 \text{ N/m2} = 0.83 \text{ atm.} \quad (25)
\]

The acceleration pressure differences at the entrance and exit from the target region more or less cancel around the loop, but the pressure in the target region is smaller than the pressure in the entry tube according to

\[
\Delta p_{accel} = 3.102 \times 10^4 \text{ g-cm/s2/cm2} = 3.102 \times 10^3 \text{ N/m2} = 0.282 \text{ atm.} \quad (26)
\]

I have not accounted for the pressure drop in the heat exchanger except to the extent that it is included in the pressure drop in the assumed 5-meter-long external piping.

The power required to circulate the fluid is

\[
P_{pump} = V' \Delta p_{friction} = (160 \text{ cm3/s}) \times (9.08 \times 10^4 \text{ N/m2}) = 14.53 \text{ N-m/s} = 14.53 \text{ W}, \quad (27)
\]

which does not account for the efficiency of the pump. A centrifugal pump or an electromagnetic pump might be appropriate to circulate the coolant.

5. Compatibility of liquid gallium with components

Liquid gallium is apparently compatible with 304 stainless steel piping components, as described in the invention disclosure of Smither, et al. [9] These authors used an electromagnetic induction pump to circulate the liquid gallium. And liquid gallium is apparently compatible with beryllium metal, as described in the paper of Blackburn and Yanch [10,11], who tested the capabilities of a gallium-cooled beryllium target system for use in accelerator-based neutron capture therapy (ABNCT). Those authors
used a centrifugal pump to circulate the liquid. Blackburn’s thesis is a thorough study of a gallium-cooled beryllium-target system, with experimental and successful prototype demonstration.

6. Conclusions and a recommendation

The scoping study reveals that a gallium-cooled system maintains target temperatures (~100°C above ambient), with modest coolant velocities (~1 m/s, 0.16 l/s), modest coolant temperature rise (~55°C), affordable coolant mass (~10 kg: ~$5000), moderate pumping power (~1 atm pressure drop), and moderate target coolant pressure (~0.3 atm below system driving pressure).

One might design a high-power (>~10 kW beam power) CANS for use with gallium coolant, but begin operation with water coolant. Existing systems would need back fitting of some components. New systems would require engineering for the less familiar gallium coolant, but this seems to present no big problems. Titanium rather than steel (aluminum is incompatible with gallium) used in the body of the target and nearby (Fig. 2), might be desirable to reduce the radioactivity of the target and facilitate access, and stainless steel used in the rest of the cooling circuit (Fig. 3.) The ABNCT source, with beryllium target and gallium cooled, built and operated at MIT, is a proof-of-principal prototype and source of experience. A windowless X-ray source based on gallium, still existing at ANL, is also a valuable prototype.

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References