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# FAST COOLING TECHNOLOGY FOR SAMPLE STICK IN TOP LOADING CRYOSTATS

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## ABSTRACT

Cryostats are essentially important sample environments for low-temperature neutron scattering experiments. They are primarily used to investigate materials of interest, i.e., samples of crystalline solids or powders, at temperatures below 100K. Dry cryostats, where the samples are cooled by helium exchange gas, are favored due to their simple operation, low maintenance, and, most importantly, moderate usage of progressively more expensive helium. However, samples in dry cryostats usually take almost two hours to cool from room temperature to $\sim$80 K, causing time-consuming changeover delays. Since neutron scattering experiments are usually conducted under strict time restraints at large-scale user facilities, maximizing instrument time utilization is of great significance. A novel sample stick with an embedded liquid nitrogen transport line is presented and shown to achieve improved cooling performance by involving the boiling of liquid nitrogen. The preliminary experiments demonstrate a cooling time of $<$2 min, compared to $\sim$2 hr for a typical dry cryostat.

## NOMENCLATURE

h Heat transfer coefficient
q“ Heat flux
$T_{sur}$ Surface temperature
$T_{sat}$ Fluid saturation temperature
$T_{ONB}$ Onset of boiling temperautre
$T_{Lei}$ Leidenfrost point
$\Delta T$ Superheat

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## 1 INTRODUCTION

Neutron scattering experiments involve passing the artificially created neutron beam through a sample and detecting where (and possibly when) the scattered neutrons encounter a detector. Subsequently, the researchers can extract a wide range of material information from collected data, i.e., atom arrangement and material properties at atom scale. Most neutron scattering experiments require special physical or chemical conditions/environments to induce the tested sample into a phase or state of particular interest [1, 2]. Cryogenic temperature, as low as mili-K scale, is one of the common environments. Such low temperature is usually achieved using cryostats. Among various designs, a top loading cryostat style refers to one with a sample loaded/unloaded from the top (1).

The top loading cryostats can be categorized into wet and dry (cryogen-free) cryostats, based on the cooling mechanism. In wet cryostats, a liquid helium bath is used to cool samples down to low-K scale, with another wrap-around liquid nitrogen outer bath serving as a 77 K shield (Figure 1a) [3]. The dry cryostats can be further divided into two types: one uses a cryocooler as the cold source to cool the sample down to the desired temperature and is called a closed cycle refrigerator (CCR, Figure 1b) [4]; the other type still uses a closed helium loop to cool sample down, where liquid helium vaporizes in a heat exchanger to absorb heat from the sample while the helium vapor dumps heat to a cryocooler and condenses back to liquid (Figure 1c) [5]. Although wet cryostats have the advantages of high energy efficiency and low vibration, there is a strong trend to move forward to dry cryostats due to their low maintenance cost, simple oper-

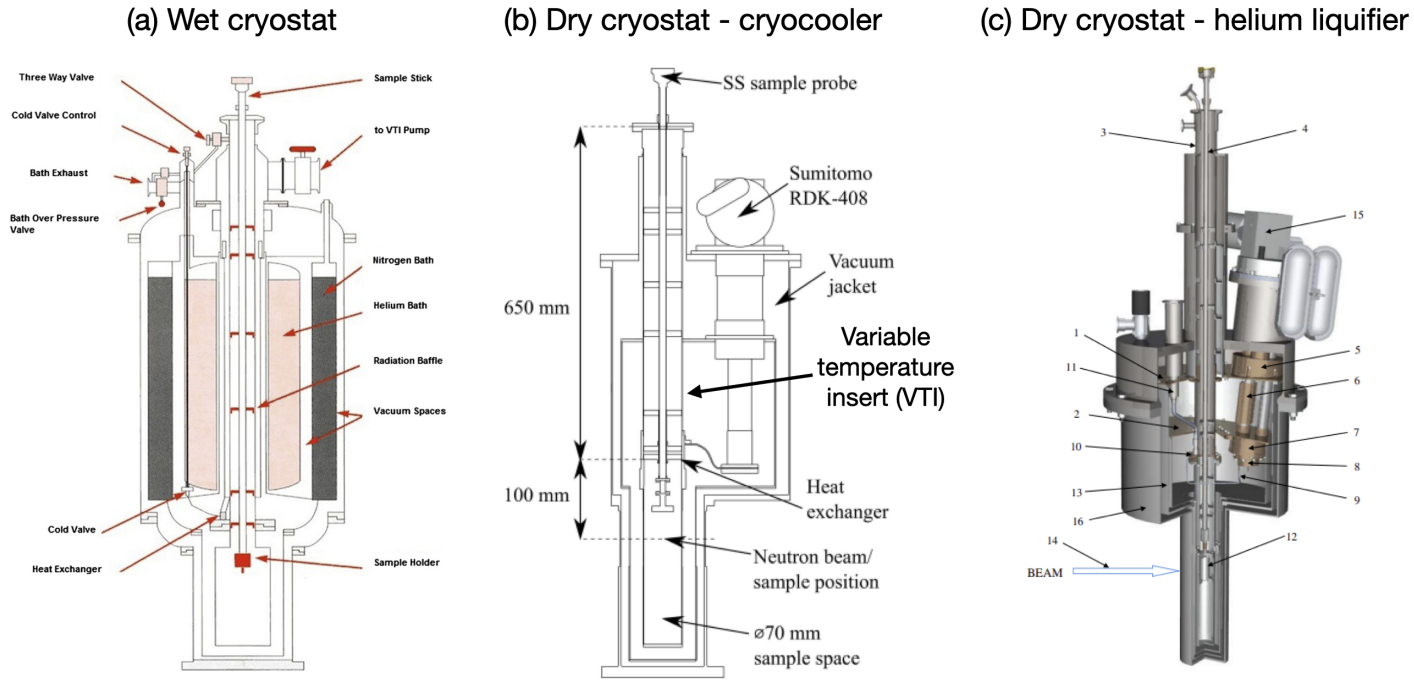


**FIGURE 1**. Typical cryostats: (a) wet cryostat [3], (b) dry cryostat with cryocooler [4], (c) dry cryostat with a closed loop for venting and liquifying helium [5].

ation, and, most importantly, not consuming more and more of the expensive liquid helium.

The CCR cryostat, a device of interest in current work (Figure 1b), uses an intricate sample holding and cooling system. The actual sample is either a solid "crystal" or powder inside a dedicated can. The size varies between experiments, but usually is between small pin and nearly filling the allowable sample volume. A crystal sample or sample can is mounted on a holder at the bottom of the sample stick, the object of current study. Then, the sample stick is inserted into the sample space of the cryostat, which is filled with low-pressure helium exchange gas. The sample space is surrounded by a Variable Temperature Insert (VTI), which is connected to the cold stages of a cryocooler through thermal links. Thus, the cryocooler cools the VTI through thermal conduction, and then the VTI cools the helium exchange gas through inefficient natural convection. Finally, the sample itself is cooled by the exchange gas through natural convection with the low-pressure cold exchange gas. This inefficient heat transfer path impacts the cool-down time from room temperature to $<4$, typically taking $>3$ hours to complete.

Figure 2 shows a typical cooling curve of a dry "Orange" style CCR cryostat [6]. It takes almost 2 hours to cool down from room temperature to $\sim$80 K, and less than 10 minutes from $\sim$80 K to 10 K. The discrepancy in cooling rates is associated is substantial decrease in materials' specific heat, hence the thermal mass, as the temperature reaches cryogenic range [7]. Ultimately another $>1$ hour is required to cool from 10 K to 2.7 K due to the

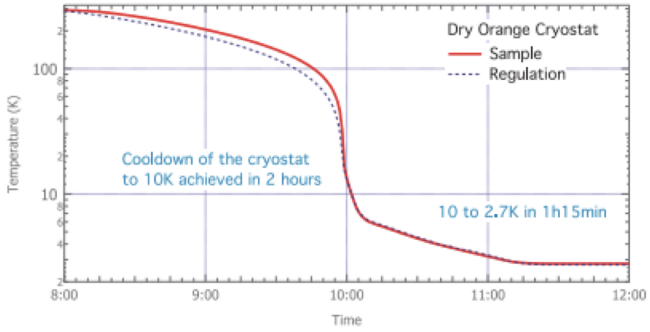


**FIGURE 2**. Cooling curve of a dry Orange cryostat [6].

severely reduced cooling power of the cryocooler in that temperature range [6]. The current work aims to reduce the cooling time from room temperature to $\sim$80 K down to minutes. Such improvement dramatically reduces the overall sample change time. Considering the fact that the neutron scattering experiments are usually performed under strict time restraints, as well as the high neutron beam cost (tens of thousands of dollars per hour), reducing sample change time is of great significance to utilize the beam time efficiently.

## 2 DESIGN

### 2.1 Sample Stick with Embedded Liquid Nitrogen Transport Line

A new sample stick design (Figure 3) has been developed to aid the initial sample cooldown from room temperature to $\sim$80K

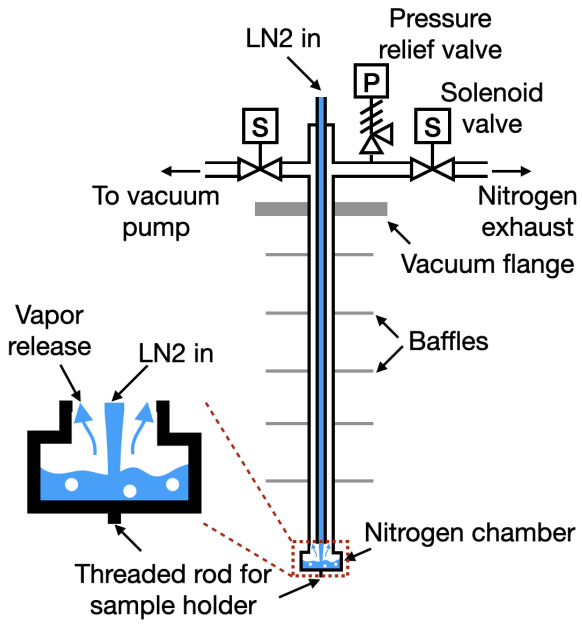


**FIGURE 3**. Concept: sample stick with embedded liquid nitrogen transport line.

in minutes by involving boiling of liquid nitrogen, an efficient heat transfer mechanism utilizing nitrogen's latent heat [8, 9]. A small tube is embedded in the sample stick to transport liquid nitrogen to a receiver chamber at the bottom, near the sample position. This chamber holds liquid nitrogen (not shown) and rapidly cools the sample holder via liquid nitrogen boiling. The generated nitrogen vapor is released to the ambient through the annular space between the inner transport tube and the outer sample stick. A solenoid valve controls the exhaust release, and a pressure relief valve is added to the line to avoid overpressure. An additional connection at the top enables a vacuum pump connection. The line is pre-evacuated before introducing liquid nitrogen to remove moisture and prevent nitrogen ice formation inside the sample stick. After the sample reaches ~80 K, the space in the sample stick will be drawn vacuum again to remove the remaining nitrogen, as well as provide thermal insulation between the sample and the outside environment. Five reflective baffles are fitted along the length of the sample stick to reduce radiation heat transfer between the sample and the outside environment.

## 2.2 Boiling Curves

The sample is cooled by boiling off nitrogen in the chamber. Hence, the complex boiling nature of a highly superheated surface influences the cooldown time. Figure 4 shows a typical boiling curve, where the heat flux, q", transferred from solid surface to working fluid is plotted against surface superheat, which is defined as the difference between surface temperature and fluid saturation temperature, $T_{sur} - T_{sat}$. With surface superheat increasing from zero, single-phase convection is the heat transfer mechanism, as bubble nucleation is not triggered due to the low superheat. At a certain superheat (point A, onset of boiling, $T_{ONB}$), vapor bubbles form and depart from the surface as boiling occurs. The heat transfer improves dramatically due to the incorporation of latent heat. However, as the superheat keeps increasing to point B, the vapor generation rate overcomes the vapor removal rate. Thus, the vapor starts to accumulate on the surface and form local vapor layers. This vapor layer acts as thermal insulation due to its low thermal conductivity, jeopardizing local heat transfer. Thus, point B is the maximum heat transfer that nucleate boiling can reach. With increasing surface superheat, the heat transferred from surface to working fluid decreases as a vapor layer covers more and more locations until the entire surface is covered by an intact vapor layer (point D, defined as Leidenfrost point, $T_{Lei}$). In this range, the heat transfer mechanism is transition boiling. After reaching the Leidenfrost point, heat is transferred through film boiling, where heat flux is increased again with increased surface superheat [10].

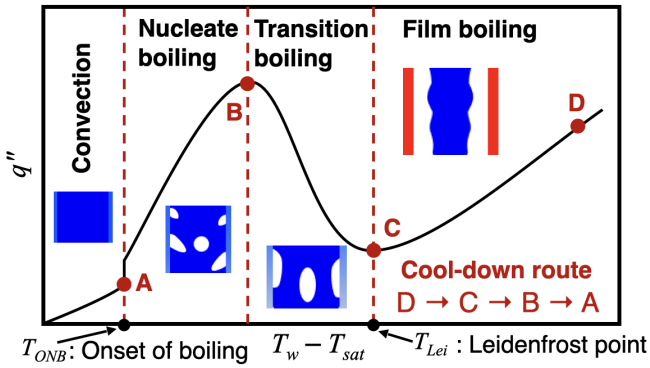


**FIGURE 4**. Boiling curve.

During the cooling of the sample stick, an arbitrary point of interest in the nitrogen transport tube is expected to experience a reverse process order to the one described above, with a heat transfer mechanism from D to A. The heat transfer coefficient ($h$) of a subject submerged in liquid nitrogen can be estimated using correlation (Eq. 1) based on the range of surface superheat, $\Delta T$ (difference between surface temperature and nitrogen saturation temperature), including film boiling, transition boiling, nucleate boiling, and natural convection with all units in SI form [11].

$$
\begin{aligned}
h &= 125 + 0.069 \times \Delta T, && 52\ K \le \Delta T \le 214\ K,\\
h &= 13087.8 - 723.04 \times \Delta T + 13.48 \times \Delta T^2 \\
&\quad - 0.084 \times \Delta T^3, && 19.6\ K \le \Delta T < 52\ K,\\
h &= 82.74 - 131.22 \times \Delta T + 37.64 \times \Delta T^2 \\
&\quad - 1.13 \times \Delta T^3, && 4\ K \le \Delta T < 19.6\ K,\\
h &= 21.945 \times \Delta T, && 0\ K \le \Delta T < 4\ K.
\end{aligned}
\tag{1}
$$

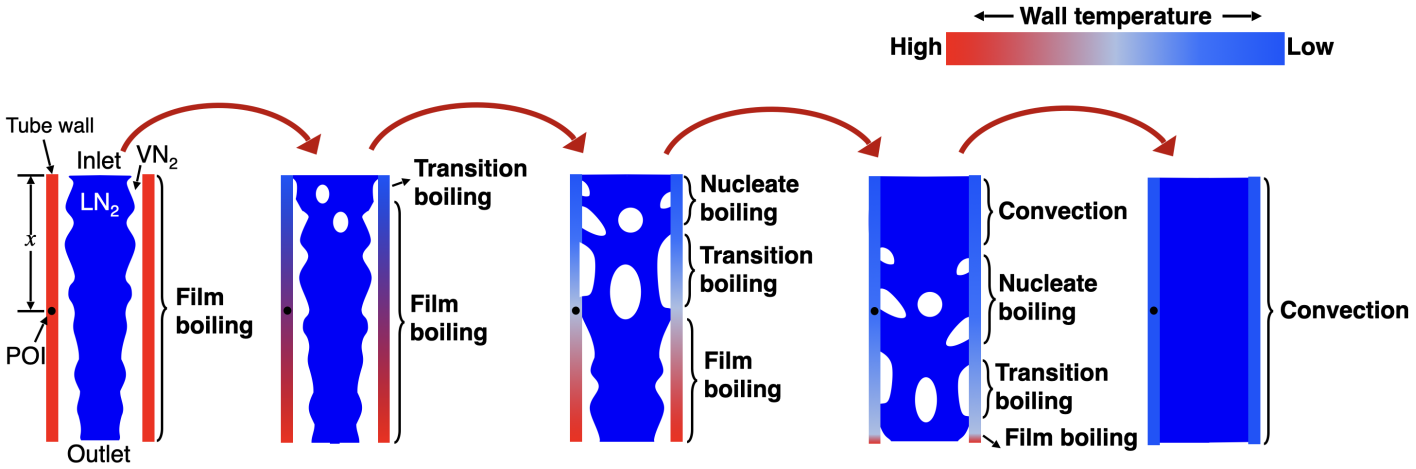


**FIGURE 5**. Expected heat transfer mechanisms in cooling of the cryostick.

Figure 5 provides a general visual overview of the heat transfer mechanism for each stage of nitrogen source tube cool-down. Initially, the entire tube is at room temperature, far beyond the Leidenfrost point of nitrogen, thus film boiling is the only heat transfer mechanism. Subsequently, the tube near the inlet is cooled down first, allowing local transition boiling to occur while the downstream area is still cooled by film boiling. Progressively, the transition boiling covers more and more regions as the nucleate boiling occurs near the inlet. All three mechanisms can coexist for sufficiently long pipes with film boiling remaining downstream, transition boiling at the middle, and nucleate boiling establishing upstream. Eventually, the surface temperature sufficiently reduces for boiling to stop at the inlet, and the heat transfer mechanism becomes single-phase convection. Ultimately, the entire tube is cooled by single-phase convection only.

## 3 RESULTS AND DISCUSSION

### 3.1 Proof-of-Concept Experiments

Preliminary proof-of-concept experiments were run for the discussed concentric tube design. An initial version of cryostick, shown in Figure 6, features an embedded liquid nitrogen transport line fabricated by inserting a 1/4" tube (nitrogen transport line) in a 3/4" tube (sample stick). The length of the sample stick below the ISO flange is 55". This is a full-scale prototype, which is supposed to be implemented directly into a cryostat with a sample space of 4" diameter. The assembly is open to ambient at the top and sealed at the bottom by a stainless tube reducer. The large thermal mass reduced simulates the targeted sample holder (right of Figure 6). The prepared cryostick assembly was placed in a 4" vacuum chamber, sealed by ISO flanges. After evacuating the vacuum chamber to mTorr vacuum level, the liquid nitrogen was supplied to the cryostick transport line from a standard nitrogen Dewar. T-type thermocouples were attached at the neck of the tube reducer (denoted as sample) as well as 6", 9", 12", and 18" above the sample position to measure the cryostick outer wall temperature and its evolution with time. All temperature readings were recorded by NI DAQ modules and the LabVIEW VI program. The measurement errors in this experiment are from T-type thermocouple readings, which is $\pm 1$ °C or $\pm 0.75\%$ whichever is greater. The maximum uncertainty of measurement in the current work is $\pm 1.5$ K. The temperature measurement approach is further verified by measuring liquid nitrogen temperature at atmospheric pressure. A T-type thermocouple was immersed in a liquid nitrogen pool, and the temperature measured from the average of 2.5 min readings (one reading per second) is $77.73 \pm 1.48$ K, in a good agreement with nitrogen's boiling point at atmospheric pressure (77.36 K).

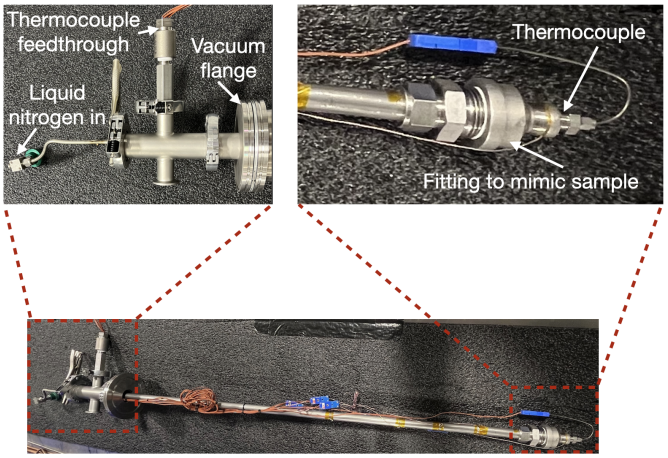


**FIGURE 6**. Fabricated version zero of cryostick for proof-of-concept testing.

Figure 7 shows the preliminary cooling curve of the proposed cryostick with an embedded liquid nitrogen transport line. Starting from room temperature, the simulated sample was cooled down to $84.96 \pm 1.44$ K within two minutes, compared

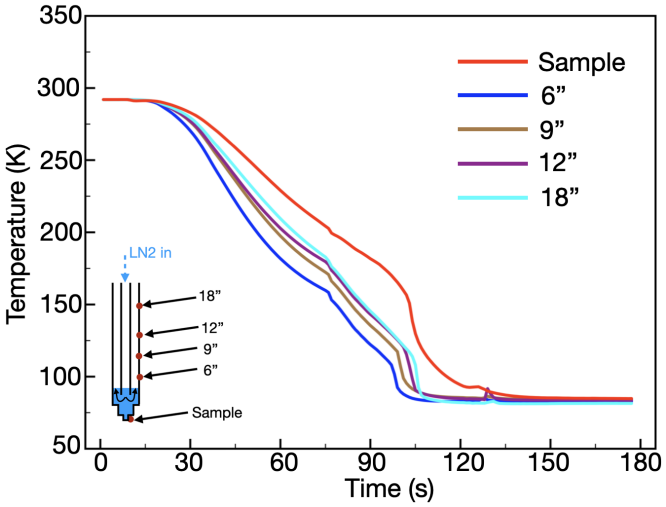


**FIGURE 7**. Cryostick cooling curve.

to almost 2 hours in a typical CCR cryostat. The temperature decreased relatively slowly at the early stage of the cooling due to the low heat transfer coefficient of film boiling, followed by a sudden drop, indicating the transition from film boiling to transition/nucleate boiling. The transition at the sample occurred at ∼160 K and around ∼125 K for 6”, 9”, 12”, and 18” positions above the sample. The higher transition temperature at the sample is due to a large temperature gradient across the thick wall of the transition fitting, where the thermocouple is positioned at the outer wall. Meanwhile, the ∼125 K transition at the thin wall cryostick (small temperature difference between the inner surface and the outer wall) shows a good agreement with literature [12, 13]. Notably, the 6” position shows the fastest cooling rate due to its proximity to liquid nitrogen accumulation at the bottom of the cryostick. The observed behavior confirms the expected evolution of liquid nitrogen inside the rapid cooldown cryostick. Furthermore, the assembly successfully reduced the initial cooldown time from room temperature to ∼80K down to a couple of minutes instead of hours.

### 3.2 Future Work

In the near future, a prototype sample stick compatible with the actual CCR system will be fabricated and tested. To reduce liquid nitrogen consumption and further reduce time, several strategies to enhance film boiling heat transfer and to increase Leidenfrost point will be explored. Fine control of the cooling process, i.e., pressure and flow rate of liquid nitrogen, will also be attempted.

## 4 CONCLUSIONS

Reduction of the cool-down time of the sample in cryostats is of great significance for fast sample change in neutron scattering experiments. Inconveniently, it currently takes almost two hours for current dry cryostats to cool the sample from room temperature down to ∼80 K. A new concept of a sample stick has been presented that embeds liquid nitrogen transport line to achieve fast cooling by involving boiling of liquid nitrogen. The preliminary experiments demonstrate a fast cooling time from room temperature to ∼80 K within <2 min.

## ACKNOWLEDGMENT

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