Cryogen free cryostat for neutron scattering experiments

O Kirichek, R B E Down, P Manuel, J Keeping and Z A Bowden
ISIS Facility, STFC, Rutherford Appleton Laboratory, Harwell Oxford Campus, Chilton, Didcot, OX11 0QX, UK

E-mail: oleg.kirichek@stfc.ac.uk

Abstract. Most very low temperature (below 1K) experiments at advanced neutron facilities are based on dilution and $^3$He refrigerator inserts used with Orange cryostats, or similar systems. However recent increases in the cost of liquid helium caused by global helium supply problems, has raised significant concern about the affordability of such cryostats. Here we present the design and test results of a cryogen free top-loading cryostat with a standard KelvinoxVT® dilution refrigerator insert which provides sample environment for neutron scattering experiments in the temperature range 35 mK – 300 K. The dilution refrigerator insert operates in a continuous regime. The cooling time of the insert is similar to one operated in the Orange cryostat. The main performance criteria such as base temperature, cooling power, and circulation rate are compatible with the technical specification of a standard dilution refrigerator. In fact the system offers operating parameters very similar to those of an Orange cryostat, but without the complication of cryogens. The first scientific results obtained in ultra-low temperature neutron scattering experiment with this system are also going to be discussed.

1. Introduction
Low temperature (LT) sample environment below 1K at neutron scattering facilities is usually provided by dilution and $^3$He refrigerator inserts set in a top-loading Orange cryostat [1] or similar system. The demand for LT sample environment has dramatically increased in the last decade and in the case of the ISIS facility [2] has exceeded eighty LT experiments per year. This large scale usage of cryogenic equipment requires significant resources and can create a number of problems including health and safety issues and the considerable cost of the required cryogens. This last problem has become more significant due to increases in liquid helium costs caused by global helium supply problems [3].

The latest progress in cryo-cooler technology offers a new generation of cryogenic systems in which the cryogen consumption can be significantly reduced and in some cases completely eliminated. These systems also offer the advantage of operational simplicity, require less space than conventional cryogen-cooled systems and can significantly improve user safety. The most successful representative of the cryo-cooler family today is the pulse tube refrigerator (PTR). The unique feature of the PTR is the absence of any cold moving parts, which considerably reduces noise and vibrations generated by
the cooler [4, 5]. It also increases the reliability of the cold head because no expensive high-precision seals are required and the cold head can be operated without any service inspection.

Here we discuss the operational experience and first scientific results obtained in a neutron beam line experiment where ultra-low temperature sample environment has been provided by a dilution refrigerator insert accommodated in the newly developed 50 mm diameter top-loading cryogen-free cryostat [6]. The cryostat provides sample environment in the temperature range of 1.4 – 300K with an exceptionally high cooling power of 0.23W @ 1.9K. Employment of an Oxford Instruments’ KelvinoxVT® dilution refrigerator insert loaded into the cryostat’s variable temperature insert (VTI) allowed us to perform neutron diffraction measurements at a base temperature of 36 mK. This system has been developed through an ISIS and Oxford Instruments collaborative project.

![Figure 1. Cooling of the cryostat down to base temperature: (a) time dependence of temperature at the VTI exchanger; (a) time dependence of the mixing chamber temperature.](image)

2. System design

The design of the system is based on the idea of combining a top-loading cryogen-free system described in Ref. [4] with a helium condensation loop [7], which has been successfully realized in the first prototype described in [8]. The major changes in design of the second prototype [6] used in the experiment presented in this paper are the replacement of the cryostat’s top-loading insert with a standard Oxford Instruments VTI and the inclusion of a PTR second stage heat exchanger developed by ISIS to provide greater efficiency at high flow rates preferred for beam line operation. The detailed description of the system design, performance and test results can be found in Ref. [6].

Most of the modern dilution refrigerators used in neutron scattering experiments are based on the Grenoble design which incorporates sintered silver heat exchangers [9-11]. A relatively small number of neutron scattering experiments that require very low temperature in combination with high cooling power use a powerful cryogen free dilution refrigerators built around a PTR cryo-cooler [2, 12, 13]. Yet predominantly the sample environment below 1K is required for small samples. For this purpose compact dilution refrigerator inserts are usually used with the VTI of a cryostat. These systems consume cooling power produced at the VTI heat exchanger. In our case the procedure of loading and cooling of a standard KelvinoxVT® dilution insert was similar to the one employed in conventional
cryostats. The cool down procedure consisted of three stages. Stage one was pre-cooling of the insert with heat-exchange helium gas in the refrigerator’s vacuum jacket from room temperature down to ~ 2K. After that the VTI was warmed up to 5K and exchange gas was pumped out for approximately one hour. Once good vacuum in the vacuum jacket was achieved, the VTI heater was switched off and dilution refrigerator insert started helium mixture condensation and, eventually, circulation in an automated regime.

![Figure 2](image.png)

Figure 2. Time dependence of the mixing chamber temperature. (a) time dependence of temperature at the VTI exchanger; (b) time dependence of the mixing chamber temperature. Point A - neutron beam is switched off. Point B - neutron beam is back to its previous value (accelerator proton beam current 42 μA).

3. System operation

Cooling of the refrigerator down to base temperature is shown in Figure 1, where (a) represents the behavior of temperature at the VTI exchanger and (b) time dependence of the mixing chamber temperature. The refrigerator has been operated in a fully automated mode. The total cool down time of the dilution insert starting from inserting the refrigerator in the VTI was similar to the typical time taken for a conventional cryogen based top-loading cryostat.

During the first overnight base temperature run the system demonstrated remarkably stable performance. In Figure 2 we present: (a) time dependence of the VTI heat-exchanger temperature and (b) mixing chamber temperature. This high stability can be explained by the absence of time dependent processes such as changes of liquid levels in the helium and nitrogen vessels present in conventional cryostat operation. In the middle of the plot (point A) it is possible to see a step-like drop in the base temperature caused by switching the neutron beam off. After some time (point B) the neutron beam was back and the mixing chamber temperature returned to its previous value. Based on the technical specification of a standard KelvinoxVT® insert we would estimate the heating power of the neutron beam at a level of a few microwatts.
4. Experimental results
The first neutron scattering experiment where ultra-low temperature sample environment was provided by the cryostat described here has been conducted on WISH [9] at the ISIS Facility, STFC Rutherford Appleton Laboratory, UK. WISH is a high count rate, long-wavelength diffractometer with remarkably low neutron scattering background.

The diffraction pattern shown in Fig. 3 is obtained from a powder sample of Sr$_2$Ir$_{0.5}$Rh$_{0.5}$O$_4$, prepared by Dr D. Prabhakaran (Oxford University). It is an Anderson localized insulator thought to display an unusual magnetic order at 0.6K. There is an increasing interest in 5d oxides due to the richness of phases of matter they display, owing to similar energy scales of $U$, spin orbit coupling and other magnetic interactions in competition. The undoped Sr$_2$IrO$_4$ has been particularly studied over the past few years [10].

WISH provides excellent quality neutron powder diffraction capabilities to study magnetic ordering. As the ordered phase occurs at ~ 0.6K, a dilution fridge is required to cool the sample significantly below this temperature. The high-quality polycrystalline powder sample has been sealed in the thin-wall vanadium can with helium gas added as a heat-exchange media. This approach has been proven to be an efficient way for providing temperature equilibrium across the sample and vanadium can [16]. The diffraction data obtained in the experiment demonstrate very good angular coverage and data quality (low noise and background). Another important achievement is the high-level temperature stability at the refrigerator’s base temperature which remained ± 0.1 mK during the whole measurement.

5. Conclusions
The ISIS facility carries on an internal development program intended to gradually substitute all conventional cryogenic systems with cryogen free systems preferably based on PTR. Here we discuss operational experience and first scientific results obtained in a neutron beam line experiment where very low temperature sample environment has been provided by a KelvinoxVT$^\text{®}$ dilution refrigerator insert accommodated in a newly developed top-loading cryogen-free cryostat. The system demonstrated high long time stability performance with a base temperature of 36 mK. High resolution neutron diffraction data have been obtained in the experiment. From a user perspective, the system
offers operating parameters very similar to those of standard Orange cryostats but without the complication of cryogens.

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