Cryogenic sample environment on TOSCA

Richard B E Down, Anibal J Ramirez-Cuesta, Robert A Major, Jeff Keeping, Svemir Rudić and Oleg Kirichek

ISIS Facility, Rutherford Appleton Laboratory, Chilton, Didcot, Oxfordshire, OX11 0QX, United Kingdom

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

Abstract. We have developed a new top-loading cryogenic insert that forms part of the integrated cryogenic sample environment for the Tosca spectrometer at the ISIS Facility. New advanced cryo-coolers and optimised design of the cryogenic insert allowed us to dramatically decrease system cool down time from 20 to 7 hours, sample cooling time from 3 hours to 45 minutes and also reduce the base temperature of the insert from 12 K to 4.5 K in the continuous regime and ~1.5 K in the single shot regime. We have also reduced the number of cold heads from three to two. This lets us save the significant cost of the third cold head maintenance and has cut electricity consumption by ~30%. A much lower base temperature of 4.5 K has opened new opportunities for molecular spectroscopy studies on Tosca.

1. Introduction

Tosca is an indirect geometry spectrometer optimised for the study of molecular vibrations in the solid state [1]. Science on Tosca includes studies of catalysts, hydrogen storage materials, hydrogen bonded systems, advanced materials, biological samples and organic compounds. For the majority of experiments Tosca requires cryogenic sample environment. This is why the cryogenic sample chamber was embedded in the Tosca instrument. The design of the original sample chamber has been based on a dry cryogen free approach [2]. Necessary cooling power has been provided by three closed cycle refrigerators (CCR), which allowed measurements in the range between 12 K and room temperature. However the system suffered from a very slow initial cool-down of more than 20 h and time consuming sample changes and subsequent cooling of around 3 h.

The ISIS cryogenic section together with the Tosca instrument scientists have re-designed the cryogenic chamber in order to optimise the cryogenic performance of the system. Due to much higher efficiency of the newly designed cryostat the number of cryo-coolers has been reduced from three to two, which has resulted in a 30% reduction in electricity consumption. The newly designed cryostat has also demonstrated a much lower base temperature of 4.5 K and significantly reduced system and sample cooling times.

Here we present the design of the optimised cryogenic sample chamber and discuss the system’s performance in a proof of principle measurement.

2. Design of cryogenic sample changer

There are two entirely different cryogen free approaches: one is based on re-condensing of the evaporating helium back to the liquid cryogen based cryostat [3] and another one, is the so-called “dry” cryogenics that does not contain liquid cryogens at all. The last system is constructed around a
closed cycle refrigerator (CCR) and utilises the cooling power produced by the cold head [4, 5]. The Tosca cryogenic sample chamber is a dry cryogen free system. The main advantage of this approach is the absence of liquid cryogens and all of the following consequences: no liquid cryogens top-ups, elimination of asphyxiation risk and a relative independence of helium supply troubles. However there is one significant drawback - the limited cooling power of the CCR. The most powerful Gifford-McMahon (GM) CCRs achieve no more than 1.5 W of cooling power at 4.2 K. In a conventional liquid helium based system, one can easily achieve more than an order of magnitude higher cooling power than in a dry system just by opening a needle-valve and allowing higher flow of liquid helium. The main consequence of this is the long cool-down time of the dry system. However this crucial parameter can be significantly improved by optimisation of the cryogenic design.

The Tosca cryostat was originally designed and manufactured by AS Scientific Ltd and cooling was supplied by three Leybold CCRs. Two Leybold RGD1245 cold heads were used to cool the main copper tube thermally linked to the sample space and a single stage Leybold RGS120 cold head was used to cool the outer radiation shield. The three cold heads gave the cryostat the name “Triplex”. The Triplex system cooled from room temperature to a base temperature of 20 K in approximately 20 hours and had a sample change time of approximately 3 hours.

In the first phase of the Triplex upgrade project the two Leybold 1245 10K cold heads were replaced by two Sumitomo SRDK415 4 K cold heads. In order to reduce the sample change time a new capillary for transferring liquid helium into the bottom of the sample space was also added. The new 4 K cold heads have high cooling power on the first stage which enabled the redundancy of the third single stage CCR. This provided a large saving in running and maintenance costs. The removal of this cold head led to a change of name to “Duplex”. The results of the upgrade lead to an overall improvement with the Duplex achieving a base temperature of 8 K. However the cool-down time was not as good as expected.

**Figure 1.** The new design of the Tosca cryostat: (1) two CCRs; (2) first stages of CCRs; (3) outer vacuum can; (4) infrared radiation shield; (5) cryogenic insert; (6) sample; (7) sample chamber with windows for neutron beam access; (8) CCR’s second stages; (9) shimmed thermal links.
In order to improve the cool-down time it was decided to implement significant changes to the low-temperature part of the cryostat design. The new design presented in figure 1 was based on a modular approach in order to simplify any further repairs or modifications that could be required in the future. The cryogenic insert (5) is surrounded by an infrared radiation shield (4) attached to the first stages (2). The CCRs (1) are placed in the outer vacuum vessel (3). The thermal contact between the cold walls of the insert and the sample (6) is achieved by a small amount of heat-exchange helium gas added into the insert. The pressure of the heat-exchange helium gas is usually around 20 mbar.

The cooling power of CCR’s second stages (8) is delivered to the copper tube of the insert through the shimmed thermal links (9). During preliminary tests of the newly designed cryostat significant temperature gradients were observed across the thermal links which pointed out that there was a “bottle-neck” in the thermal conductivity circuit between the CCR’s second stages and the sample chamber (7).

In order to optimise the thermal conductance of the cryostat components, tests were carried out to compare the efficiency of different thermal linking techniques. Three copper thermal links were manufactured using different fixing techniques presented in figure 2: (A) clamping, (B) clamping with soldering and (C) welding. Each link was then mounted to the bottom of a 10K cold head with a dummy load fixed to the end of the link. Two temperature sensors were mounted to the links. The first was to read the temperature at the bottom of the cold head and the second was to read the temperature of the dummy load. Each thermal link was then cooled to its base temperature and the results of the three tests compared.

![Figure 2](image_url)

Figure 2. Three copper thermal links manufactured using different fixing techniques: (A) clamping; (B) clamping with soldering and (C) copper eutectic welding.

The data clearly showed the difference between the three fixing techniques. Initially clamping and clamping with solder were almost matched with clamping with solder cooling slightly faster. However the soldered links cooling began to slow around 50 K and only reached a base temperature of 33 K in 5 hours. The clamped link continued to cool to a base of 16 K in 4 hours. The copper eutectic welded links initial cooling was also similar yet maintained an almost linear cooling rate until it reached its base temperature of 14 K within 4 hours. With this new information it was decided to use the welded thermal links in the design of the cryostat.

3. System test results and discussion
The system cool-down test results are shown in figure 3a. It is possible to see the temperature of the sample chamber (1) lagging behind the temperature of the second stages of the CCRs (2). In the test the temperatures of both second stages were the same within our temperature resolution. After approximately three hours the temperature of the sample chamber started to catch up with the temperature of CCR’s second stage and they became comparable after ~ 6.5 hours. The cryostat reached the base temperature of 4.5 K from start up in 7 hours, which demonstrates significant
improvement in comparison with the previously achieved 20 hours of cool-down time to a base temperature of 12 K.

In figure 3b we present the cool-down of the sample from room to the base temperature with the cryogenic insert already cold. As it is possible to see, the sample cooling from 300 K to 20 K takes just 45 minutes in comparison with previously achieved 3 hours. If required the sample can be rapidly cooled using a flow of liquid helium through the 4 mm helium transfer tube, that enters the cryostat at the sample space. In this case 4.5 K can be reached in less than 10 minutes. Pumping on the residual liquid helium remaining after completion of the sample cool down gives a temperature of ~ 1.5 K for approximately 3 hours in a single shot regime. However liquid helium assisted cooling turned out to be quite an expensive and inconvenient method, which can be explained by the high helium prices and the necessity of using trained personnel to perform this procedure. These disadvantages outweigh the short sample cooling time and lower base temperature. Therefore the use of liquid helium assisted cooling was restricted to a small number of experiments where a temperature of less than 2 K was essential.

Inelastic neutron scattering spectra of solid parahydrogen (p-H₂) recorded at a temperature of 4 K (black trace) and solid p-H₂ at 26 K (red trace) are presented in figure 4. In both cases the solid p-H₂ was kept under 2500 bar of normal H₂ pressure. The peak at 14.7 meV (117 cm⁻¹; 1 meV = 8.066 cm⁻¹) corresponds to the H₂ free rotor rotation. This transition is optically forbidden because it involves a nuclear-spin flip between ground-state p-H₂ (J = 0) and rotationally excited orthohydrogen, o-H₂ (J=1).
Furthermore, features at 7 and 20 meV are fundamental and combination bands which correspond to the most intense lattice vibrational frequencies in the solid and simultaneous excitation of rotational and lattice vibrational modes, respectively [6]. As the temperature is increased to 26 K the percentage of p-H₂ is reduced; consequently 14.7 meV peak as well as combination band are lower in intensity.

![Graph showing inelastic neutron scattering spectra of solid p-H₂ recorded at temperature of 4 K (black trace) and solid p-H₂ at 26 K (red trace). At both temperatures the solid p-H₂ was kept under 2500 bar of normal H₂ pressure.](image)

**Figure 4.** Inelastic neutron scattering spectra of solid p-H₂ recorded at temperature of 4 K (black trace) and solid p-H₂ at 26 K (red trace). At both temperatures the solid p-H₂ was kept under 2500 bar of normal H₂ pressure.

4. Conclusions

We have developed a new cryogenic insert for the Tosca inelastic neutron scattering spectrometer. New CCRs and optimised design of the cryogenic insert have allowed us to dramatically decrease the initial system cool down time to 7 hours, sample cooling time to 45 minutes and also to reduce the base temperature of the insert to 4.5 K in the continuous regime and 1.5 K in a single shot regime. We have also reduced the number of CCRs from three to two. This enables us to save the significant cost of the third CCR’s maintenance and to cut electricity consumption by ~30%. The base temperature of 4.5 K has opened new realms of low temperature study on Tosca.

Acknowledgements

We are grateful to members of the ISIS Experimental Operation Division involved in the Tosca cryogenics upgrade project. We would like to thank Prof Felix Fernandez-Alonso and Dr Stewart F. Parker for their help with data analysis and comments about the manuscript, as well as the TOSCA user community for their advice.

References

[1] Colognesi D, Celli M, Cilloco E, Newport R J, Parker S F, Rossi-Albertini V, Sacchetti F, Tomkinson J and Zoppi M 2002 TOSCA neutron spectrometer: the final configuration *Appl. Phys. A* 74 S64

[2] Kirichek O 2012 Impact of the cryogen free revolution on neutron scattering laboratories *Modern Physics Letters B* 26 1230006
[3] Kirichek O, Carr P, Johnson C and Atrey M 2005 Nuclear magnetic resonance magnet actively cooled by pulse tube refrigerator 2005 *Review of Scientific Instruments* **76** 055104

[4] Oliver E C, Evans B E, Chowdhury M A H, Major R A, Kirichek O and Bowden Z A 2008 Stress rig for neutron scattering measurements of bulk stress in engineering components at cryogenic temperatures *Meas. Sci. Technol.* **19** 034019

[5] Chapman C R, Evans B E, Dudman M P, Keeping J, Down R B E, Kirichek O and Bowden Z A 2011 Cryogen-free cryostat for neutron scattering sample environment. *Cryogenics* **51** 146

[6] Fernandez-Alonso F, Bermejo F J and Sabouni M L 2011 Molecular Hydrogen in Carbon Nanostructures *Handbook of Nanophysics: Functional Nanomaterials*, ed K D Sattler (Boca Raton, FL, US: CRC Press) chapter 40 pp 40-1