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DILUTION REFRIGERATOR FOR A TWO-LITRE POLARIZED TARGET

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ABSTRACT

A large dilution refrigerator has been designed and constructed for cooling a polarized proton target of two-litre volume. The main features of the refrigerator are: maximum cooling power 1.3 W at 0.55 K with 0.23 mole/s ^3He circulation, base temperature below 50 mK, and direct horizontal access to the mixing chamber, which has a microwave blockage permitting separation of opposite polarized halves of the target.

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1. INTRODUCTION

In the following we briefly describe the design, construction, and results of tests of a dilution refrigerator, which is by far the largest one so far operated.

The refrigerator was built for cooling a large polarized target¹⁾. The discussion regarding the choice between dilution refrigeration and ³He evaporation refrigeration will be given in detail in a forthcoming publication²⁾. Here we only note that the advantage lies in the better heat contact in between the hydrocarbon target material and the dilute solution, and in the good convection heat transfer in the dilute solution itself, rather than in the lower temperature that can be reached with a dilution refrigerator. Even in small targets, important improvements in the ultimate polarization have been obtained, when changing from ³He to dilution refrigeration. In targets with large dimensions, this effect can be expected to be even more important.

For optimum design of a dilution refrigerator, the maximum desired cooling power must first be specified. With a suitable choice of the target material, dynamic polarization of protons can be obtained with less than 1 mW/g power per unit mass of the target. The design of the present target started at one-litre volume, and was later increased to two litres. With the density of frozen alcohols, about 0.8 g/cm³, and a filling factor for small frozen beads of about 0.65, about 1.1 W cooling power is then required. In order to have some margin in the design, the aim was set at 2.5 W.

Extensive tests were carried out to find the best target material which would require as little power as possible and have the best possible hydrogen content. The conclusion of the study³⁾ was that pentanol-5% water doped with 4×10^{19} spins/ml EHBA-Cr(V)⁴⁾ would be best suited for the job.

The dilution refrigerator is connected to the end flange of a horizontal superconducting solenoid dewar. The solenoid⁵⁾ is fabricated at Rutherford Laboratory and the dewar at Liverpool University.

2. DILUTION REFRIGERATOR

The horizontal dilution refrigerator was designed for a maximum ^3He circulation speed of 0.5 mol/s. At the moment, the ^3He pumps limit this speed to 0.23 mol/s, which however, is sufficient for cooling the present target. A simplified diagram of the refrigerator is shown in Fig. 1.

The mixing chamber is constructed of 0.25 mm thick mylar film, and has a 72 mm inner diameter and 1020 mm length. Two ^3He inlet tubes have outlet holes distributed along the length inside the mixing chamber. The target, at present in two perforated mylar cartridges of 400 mm length, is centred within the mixing chamber by epoxy/fibre-glass laminate stiffening rings. These rings are held by three thin-wall stainless-steel tubes and nine BeCu semi-rigid coaxial cables (which are used for polarization measurement). This supporting structure is pre-strained so that the 1300 g weight of the target brings its axis along the axis of the holder. The structure is attached to the end of a vacuum can, which is sealed by rubber O-ring at room temperature, and a special indium seal at the level of the still at the moment of loading the target. This loading operation must be done avoiding overheating of the target material above 120 K. The indium seal is therefore first tightened by rapid press clamps and then adjusted by four nuts and torque wrenches.

In the target holder, eight of the coaxial cables are each terminated by a loop of 2 mm cupronickel thin-wall tube; these loops are the inductances of series-resonant circuits used for NMR measurement of the polarization of the target. Between the two target cartridges there are four baffles, which should allow sufficient microwave isolation between the two sides of the target for polarizing them in opposite directions. The target support structure is covered with a semi-circular ^3He trap, which prevents the concentrated ^3He phase from escaping from the mixing chamber.

The heat exchanger of the dilution refrigerator passes in a 7 cm^2 section groove, formed by inserting machined epoxy/fibre-glass rings in an annular channel which connects the mixing chamber and the still. The heat exchanger itself is 1 m long and consists of four segments of flat cupronickel tube, with sintered $44 \text{ }\mu\text{m}$ Cu powder both inside and outside. The total mass of the sintered powder is about 400 g, giving approximately 7.5 m^2 total heat-exchange surface in both streams. In order to minimize the transverse heat resistance of the helium in the pores of the sintered powder, the maximum thickness of the sintered layer is only 1 mm, and average thickness about 0.7 mm: this consideration is important in refrigerators optimized for running also at the high end of the temperature range.

The detailed design of the heat exchanger is described elsewhere⁶). The dimensions of the sintered exchanger were chosen so that there would be little viscous heating at low temperatures, but the axial conduction might prevent reaching very low temperatures (say, below 30 mK). At high temperatures, when the optimized ^3He flow rate exceeds 0.1 mol/s, the flow of ^3He in the concentrated stream is expected to become turbulent, which improves heat transfer but also increases frictional heating. The frictional heating, however, remains totally negligible with respect to the heat exchange at temperatures above 0.1 K. In the dilute stream the osmotic pressure drop, determined from the mass diffusion coefficient of ^3He in ^4He , leads to the choice of a stream cross-sectional area as large as 7 cm^2 .

The still was designed to boil 0.5 mol/s ^3He from the dilute solution. This requires approximately 16 W heating power, half of which was originally planned to come from the recondensing ^3He . In the first tests, however, it turned out that this recondensation system might be the likely cause of observed instabilities, and the expansion needle valve, originally in between two heat exchangers in the still, was placed before both of these exchangers. As some other changes were done simultaneously, it cannot be stated that this was the only cause of instability.

The boiling correlations in the still, which have never been measured, were estimated from the test results of an earlier still design⁷⁾. Using these, the still exchanger at maximum flow rate should have an efficiency in excess of 90%, provided that the extended outer surface of the exchanger is a good boiling surface. The fins outside the tubular exchanger were formed by wrapping a copper wire spring around the tube and soft-soldering these together⁸⁾.

The electric heater in the still consists of vertical stainless-steel strips having 9000 cm² boiling surface. This is believed to be large enough to stay below the critical boiling flux. At our largest flow rates, no sign of film boiling was observed, up to the maximum power of 15 W in the tests.

3. ³He CONDENSATION AND CIRCULATION

A simplified diagram (Fig. 2) shows the ³He and ⁴He circuits of the refrigerator system. As described above, we had originally planned to avoid condensation of the circulated ³He in a pumped ⁴He bath condenser so as to save liquid-helium consumption and to avoid buying large pumps for ⁴He. We had (fortunately, in retrospect) foreseen an evaporator heat exchanger with an adjustable ⁴He needle valve for continuous flow regulation, in order to be able to speed up the initial condensation of the ³He-⁴He mixture in the refrigerator during cool-down. Following the ³He stream in Fig. 2, we first pass through a heat-exchanger block consisting of a helix of the ³He tube in an annular low-pressure passage of ⁴He. Below this, we then have a tube-in-tube heat exchanger, in which the ³He is cooled to near ⁴He bath temperature. In this bath there is a double helix exchanger, which should bring the ³He temperature down to 3.0-3.3 K, and partly recondense the fluid. Although the phase diagram of concentrated ³He-⁴He mixtures is completely unknown, it seems plausible that at least a small fraction of ⁴He-rich liquid phase will be formed in the evaporator. The helium mixture is then further condensed in the following heat exchanger, which recovers enthalpy from the cold low-pressure ³He pumped out of the still. This exchanger has a pressure drop of 10 μ m Hg; the resulting heat-exchange efficiency is in the range 50-70% according to numerical

estimates. The ^3He is then expanded in a needle valve and further cooled in the dilution refrigerator, as described in the preceding section.

The ^4He is continuously transferred from a 500 l or 1000 l dewar. In the final layout, this dewar will be connected directly to a cold box, which can deliver 100 l/h of liquid helium. In the tests, this dewar was filled off-line, necessitating daily dewar changes.

The vapour due to transfer losses is separated in a vessel, where ^4He enters through a diffuser. A sintered brass membrane helps the gravity to separate the vapour from the liquid phase. The main purpose of this separator is to prevent vapour from entering the ^4He needle valve, which might cause serious instability in the condensation of ^3He . The vapour of the separator is used for cooling three thermal screens surrounding the ^4He part and the still of the dilution refrigerator, and also the microwave cavity which surrounds the mixing chamber and main heat exchanger of the dilution refrigerator.

The ^3He is pumped out from the refrigerator at a temperature below 4 K; no attempt was made to recover the enthalpy of this low-pressure stream, because no solution exists for making efficient exchange without too large a pressure drop, considering exchangers of a reasonable size. The ^3He is compressed in a series of eight Root's blowers*) featuring internal gas cooling, interstage water-cooled exchangers, sealed rotors for the electric motors, and purification of the ^3He leaking from the cylinder through a labyrinth seal and gear-oil volume back to the pump inlet. This purification is accomplished with molecular sieve traps. The pumps can exhaust the ^3He at 4 bar pressure; their designed maximum mass flow-rate is 0.5 mol/s, limited by the available electric motor power. The effective pumping speed at the inlet is 13000 m³/h.

The residual oil vapours are removed in an activated charcoal filter; possible residual air after start-up is absorbed in another activated charcoal trap cooled to liquid nitrogen temperature.

*) Manufacturer: Pfeiffer, Wetzlar, Fed. Rep. Germany.

The ^4He from the evaporator is pumped with six double-head membrane compressors in parallel.

4. INSTRUMENTATION

The temperature in the refrigerator system is measured using calibrated germanium, carbon, and platinum resistance thermometers. The resistance values are read with four-wire and two-wire automatic resistance bridges^{*)}.

The ^3He flow and counter-current ^4He flow are measured with thermal mass flowmeters^{**)}. At present, the flow of ^4He is controlled with the cold needle valve between the separator and evaporator. An electric motor closes or opens the valve according to the reading of a carbon resistor thermometer in the coaxial tube counter-current heat exchanger above the evaporator. Interlock circuits make it possible to have other inputs to the command of the needle valve: ^3He condensation pressure or the evaporator pressure, sensed by steel membrane transducers; evaporator temperature; $^3\text{He}/^4\text{He}$ flow ratio, etc.

In case of excessive condensation pressure, the helium on the closed circuit of the dilution refrigerator is sent to storage vessels through an hermetic pressure release valve.

Additional interlock circuits protect the pumps and refrigerator from overload situations, switching off the microwaves applied to the target and the still heater.

5. TEST RESULTS

In a series of cryogenic tests, we have reached 1.3 W cooling power at 0.55 K with 0.23 mol/s ^3He flow, 0.2 W at 0.2 K and 0.2 mol/s, and an ultimate

^{*)} Manufacturer: Instruments for Technology, Espoo, Finland.

^{**)} Manufacturer: Hastings-Teledyne, Hampton, Virginia, USA.

low temperature of 50 mK at 0.03 mol/s. In these tests the pump performance limited the ^3He circulation to 0.23 mol/s continuously, preventing a comparison of the cooling performance with theoretical predictions. At 0.2 K, however, the ^3He flow could be optimized. The power calculated from the heat exchanger parameters is 150 mW, with optimum ^3He speed of 0.14 mol/s at 0.2 K. Taking into account that the calculation is done with somewhat pessimistic assumptions, the agreement can be considered as satisfactory, and we may conclude that the main exchanger is performing according to specifications. Further tests will show whether the design maximum power of 2.5 W at 0.5 K can be reached; the present target, however, does not seem to need more power than is now available.

For stable operation at the maximum flow rates, the ^4He counter-current must be maintained 50% higher than the ^3He circulation speed. We have doubts about the efficiency of the heat exchanger between room temperature and the evaporator. However, with the cold-box supply of liquid helium this does not limit the performance of the refrigerator, even at the maximum design flow of 0.5 mol/s.

It was estimated that the heat leak to the still would be below 1 W, caused mainly by conduction through the rather solid mechanical support structures connecting the still to the back plate of the refrigerator. This leak was determined as 0.6 W, considering the minimum stable ^3He flow rate of 20 millimol/s which can be maintained continuously.

In two successful polarization test runs it was shown that a rather uniform temperature can be maintained in the target volume under high power, judging from the polarization rise-time and final value. It was, however, noted that there was a significant difference in the polarization rise-time between targets of 5 cm and 6 cm diameter. The polarization tests will be reported in detail in a forthcoming publication²⁾.

The design asymptotic low-temperature cooling power of the refrigerator is $1900 (T/K)^4$ W. Supposing that the ultimate low temperature is determined by residual heat leaks (and not by heat-exchanger parameters), the residual heat leak

gets a value of 12 mW. To explain such a large heat leak, we note that the mixing chamber and the heat-exchanger are surrounded by a 4.2 K copper microwave cavity, which already gives about 1 mW radiation heat load to the mixing chamber. Local hot areas in the cavity, or residual helium heat-exchange gas, could explain the difference. The target holder is also estimated to be able to bring a few milliwatts to the mixing chamber, owing to the nine coaxial cables, which are thermalized with titanium rings at three places along the heat exchanger. The differential thermal dilatation of these rings within the stainless-steel tube of the target holder causes thermalization of the outer conductor of the coaxial semi-rigid cables. The inner and outer conductors of these cables are both made of beryllium copper. The insulation between them is made of PTFE, the shrinkage of which may cause poor thermalization of the insulation and of the inner conductor. This 12 mW heat leak is tolerable in the present application of the refrigerator, and it could probably be made smaller by proper shielding of the mixing chamber and improved thermal anchoring of the target-holder insert.

6. CONCLUSIONS

Our test results show that it is feasible to construct dilution refrigerators with ^3He flow speeds of over 0.2 mol/s, having heat absorption capability in excess of 1 W. We feel that we are now rather close to the limit of the technology: the ^3He pump performance is limited by the power output of sealed-rotor electric motors, and simple compact heat exchangers cannot easily cope with much larger flow rates because of the large pressure drop, which is unavoidable if we want to obtain a reasonable efficiency. Although solutions may exist for overcoming these difficulties, the technology must be developed to a stage which allows long-term stable running in a demanding accelerator laboratory environment.

A critical performance parameter of the system in a long-term operation is the purity of the ^3He after compression and purification. Minor impurity concentrations, in the range 0.01 ppm, would cause blockage of the refrigerator heat exchangers in about one week. Although we do not have accurate means of measuring the present

impurity concentration, pressure rise after warm-up of the liquid-nitrogen-cooled trap makes it possible to calculate that the impurity concentration is below 0.001 ppm at the inlet to the heat-exchanger.

We also conclude that the loading system of the refrigerator is working according to specifications. Little target material is lost during the loading and unloading operations, and no unwanted temperature rise has been seen in the two polarization tests that have been done so far.

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Figure captions

- Fig. 1 : Schematic side view of the dilution refrigerator. The thermal screens are omitted for clarity.
- Fig. 2 : The ^3He and ^4He circuits of the dilution refrigerator (somewhat simplified). The ^4He flow through the thermal screens and microwave cavity is omitted for clarity.

Fig. 1

Fig. 2