

Millikelvin Cooling by Expansion of ^3He in ^4He

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ABSTRACT

In *Cryogenics Vol. 96* (2018) pp.83-89, a technique is described which enables continuous cooling in space to sub-kelvin using two expansion units which operate in counter-phase. Each unit needs two external coolers. In the present work attention is paid to the cooling chain between the expansion units and a 5 K platform cooled by a pulse-tube refrigerator, for example. It will be shown that only two (and not four) Joule-Thomson (JT) coolers, each with the same input of liquid ^3He at 2 K and 0.2 bar, are needed to cool the expansion units. These two JT coolers are alternatively switched between the units. No heat switches are needed to thermally connect/disconnect the JT coolers to the expansion cells. In addition to these two JT coolers, one more JT cooler is needed to close the gap between the platform at 5 K and the liquid at 2 K.

INTRODUCTION

A technique [1] is described which enables cooling to sub-kelvin levels in space using expansion of ^3He in superfluid ^4He . In this paper this work is extended by taking into account some aspects of the cooling chain and suggesting some simplifications.

At the moment the preferred technology to cool to around 50 mK in space is Adiabatic Demagnetization Refrigeration (ADR)[2,3,4,5]. In ADR superconducting magnets with magnetic fields of one and even three tesla are needed which are periodically charged and discharged. They contribute to weight, volume (including a shield for the stray magnetic fields which can be as heavy as 5.5 kg), and energy demand. The technique proposed in this paper does not use a large magnetic field. As such it could be an alternative for ADR. The Lynx [2] and Origins missions [3] both need 6 μW at 50 mK. In the present paper a cooling power of 10 μW at 50 mK is chosen as an example and it will be assumed that a 5 K platform is available e.g. provided by a multistage pulse-tube refrigerator. [2]

THE CYCLE

A cooling unit consists of a reservoir (R), containing pure superfluid ^4He , and an expansion cell (E), containing a ^3He - ^4He mixture, which are connected by a superleak. Figure 1 illustrates how the temperature T_E and ^3He concentration x_E in E make a loop in the T_E - x_E diagram. The concentration x_E is defined as $x_E = n_{3E}/(n_{3E}+n_{4E})$ with n_{3E} and n_{4E} the amount of moles of ^3He and ^4He in E respectively. As discussed in [1] during **a**→**b**→**c** ^4He is extracted from E by heating R. During **c**→**d** more ^4He is extracted keeping T_E constant while still heating R and externally cooling E so that T_E remains constant. During **d**→**e**→**f** ^4He is injected adiabatically in E by external cooling of R. During **f**→**a** more ^4He is injected by further cooling R. At the same time E cools the application. Together **a**→**b**→**c**→**d** form the compression phase (x_E increases) and **d**→**e**→**f**→**a** the expansion

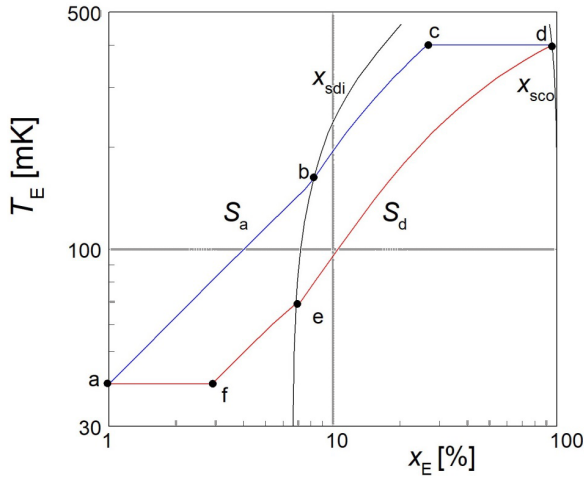


Figure 1. The cycle in the T_E - x_E diagram. This figure is chosen for a low temperature of 40 mK and a high temperature of 400 mK to allow some margin for unforeseen thermal resistances.

phase (x_E decreases). The parts of the cycle $a \rightarrow b \rightarrow c$ and $d \rightarrow e \rightarrow f$ are isentropic and $c \rightarrow d$ and $f \rightarrow a$ are isothermal. So, in the ideal case, the cycle is a Carnot cycle. The technique has been studied extensively already 25 years ago by Voncken et al. for the purpose of reaching very low temperatures in the dilute phase. [6]

Figure 2 shows the T_E - T_R dependence. It can be seen that T_R varies between 0.9 and 1.75 K for this cycle.

For a cycle time τ and a cooling power Q_L at temperature T_L in E we need an amount of ^3He in E given by

$$n_{3E} = \frac{\tau Q_L}{T_L(S_{mf} - S_{ma})} \tag{1}$$

with S_{mf} and S_{ma} the entropies per mole ^3He in **f** and **a** respectively. The thermodynamic data of the ^3He - ^4He mixtures for temperatures below 250 mK are obtained from [7]; above 250 mK from [8]. In particular from Table 6 of [8] at 500 mK $S_{mf} = S_{md} = 8.7 \text{ J}/(\text{molK})$. With S_{mf} we can find the ^3He concentration in point **f**: $x_f = 2.9\%$ from [7]. In point **a**: $S_{ma} = 14.7 \text{ J}/(\text{molK})$.

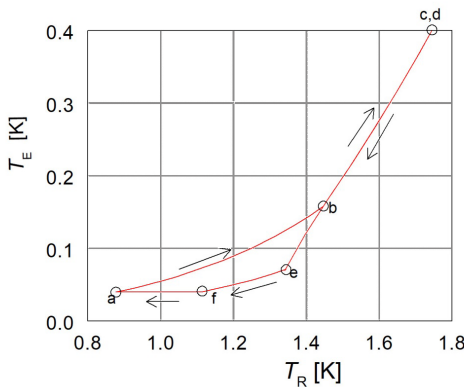


Figure 2. The cycle in the T_E - T_R diagram. During $b \rightarrow c \rightarrow d \rightarrow e$ the fountain pressure in R is equal to the osmotic pressure of the saturated dilute phase.

If $\tau=1$ hour and $Q_L=10 \mu\text{W}$ we need $n_{^3\text{He}} = 0.12$ mol. With a molar volume of pure liquid ^3He of $36.8 \text{ cm}^3/\text{mol}$ this takes a volume of 4.4 cm^3 which is the volume of the liquid in E in point **d**. In a 1% mixture (point **a**) the volume is 0.332 liter which is the minimum volume of E. We need $n_4=99n_3$ mol ^4He to make a 1% mixture, so $n_4=11.2$ mol. This has a liquid volume of 0.328 liter which is the minimum volume R should have.

The sizes, mentioned in this section are proportional to the cycle time. In that sense the numbers can be considered as per cycle hour.

In order to generate *continuous* cooling the cooler consists of two units that work in opposite phase. In an E-R combination heat is pumped from 50 mK to 500 mK. External cooling is needed of R during expansion (**d**→**e**→**f**→**a**) and of E during compression (**c**→**d**). Figure 3 is a schematic drawing of the proposed system. In Figure 3 the liquids are distributed as they would on earth. In microgravity the liquids will be distributed quite differently[1]. It shows the situation that the application is cooled by the left unit where ^4He is injected in E. The parts of the circuit, indicated by dotted lines, carry no flow. In the meantime the right unit is recovered.

The external cooling system uses ^3He as the working fluid. The reservoirs are cooled by a JT cooler with a temperature of 0.9 K. The expansion cells are cooled by a JT cooler at 500 mK. The flows in the cooling circuits are directed by switching valves which could be piezoelectric bimorphs [9] that can be regulated by the applied voltage. These switches are in series with the JT expansion valves. It is interesting to investigate whether the two valves can be combined into one with the option of having adjustable JT valves. So the two JT coolers are alternatively switched between the respective reservoirs and expansion cells.

The heat switches between E and the application are superconducting heat switches which are switched by magnetic fields in the 10 mT range. [10]

JT COOLING

The flow in the JT coolers is driven by sorption pumps to be discussed later. The ^3He vapor pressure at 500 mK is 20.5 Pa. The ^3He vapor pressure at the minimum temperature of R (0.9 K) is 694 Pa. Usually a heat exchanger between the hot and cold flow channels is an important component

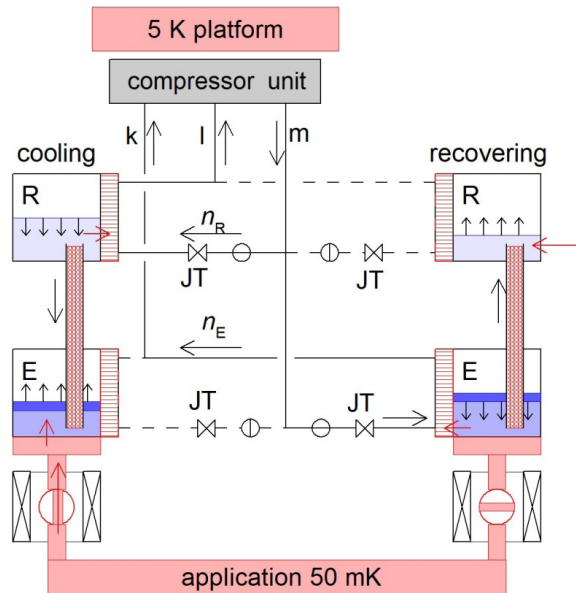


Figure 3. System layout. Left: the ER combination is cooling the application. Right: the combination is recovering. Red arrows are heat flows, black arrows are helium flows. In the tubes, indicated by dotted lines, the flow is zero. The compressor unit is described in Figure 5. The labels k, l, and m refer to Figures 4 and 5.

of JT coolers, but in these cases heat exchangers are not really necessary. Furthermore the pressures are so low that very wide channels would be needed which would prevent good heat exchange. Therefore we will consider the system without exchangers here.

First we look at the cooling requirements of E. The cooling cycle of Figure 1 is a Carnot cycle so the ratio between the adsorbed heat at 50 mK and the heat rejected 500 mK is equal to the ratio of the two temperatures; in this case a factor ten. As we aim for a cooling power of 10 μ W at 50 mK the heat rate to be absorbed at 500 mK is $Q_E=100 \mu$ W.

With H_m the molar enthalpy of pure ^3He Figure 4 is a H_m -T diagram, obtained from [11], showing the relevant isobars and the coexistence region. It will be assumed that the compressor unit delivers liquid ^3He at a pressure of 0.2 bar and a temperature of 2 K (point m in Figures 4 and 5). The cooling power of the ^3He flow rate n_E in the JT-cooling circuit for E is given by

$$Q_E = n_E(H_{mg}(0.5K) - H_{ml}(2K)) \quad (2)$$

The enthalpy of the saturated liquid at 2 K (point **m**) is $H_{ml}(2K)=9.89$ J/mol which is also the enthalpy of the liquid/gas mixture leaving the JT valve. The enthalpy of saturated ^3He gas at 500 mK (point **k**) is $H_{mg}(0.5K)=30.55$ J/mol. With Equation 2 this means that $n_E = 4.84 \mu\text{mol/s}$.

Now we look at the cooling requirements of R which is at 1.84 K when it is full (Figure 1, point **d**). The molar internal energy of pure ^4He at 1.84 K is 3.8 J/mol. In point **a**, R is empty. To remove the internal energy of 11.2 mol at 1.84 K in 1 hour asks for an average cooling power of $Q_R = 12$ mW.

The molar enthalpy of the liquid/gas mixture leaving the JT valve is the same as in point **m** of Figure 4 (9.89 J/mol). Now suppose that the pressure at the exit of the reservoir cooling channel is just below the saturated vapor pressure of ^3He at 0.9 K (696 Pa) but for convenience we will take 700 Pa. The enthalpy of saturated ^3He gas at 0.9 K (point **l**) is $H_{mg}(0.9K)=37.9$ J/mol. A heat load of 12 mW needs a ^3He flow rate in the JT cooling circuit of $n_R = Q_R / (H_{mg}(0.9K) - H_{ml}(2K)) = 428 \mu\text{mol/s}$.

There are liquid-gas mixtures leaving the JT valves. This can be beneficial for the heat exchange between the object to be cooled and the cold fluid e.g. if the object is covered by a porous layer (such as sintered copper powder) that attracts the liquid by the capillary force.

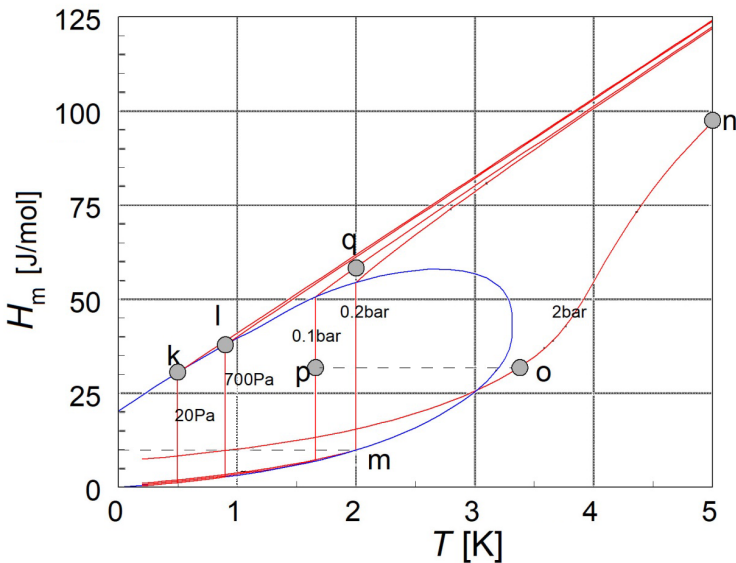


Figure 4. H_m -T diagram with the relevant isobars. The labels k-q refer to Figure 5.

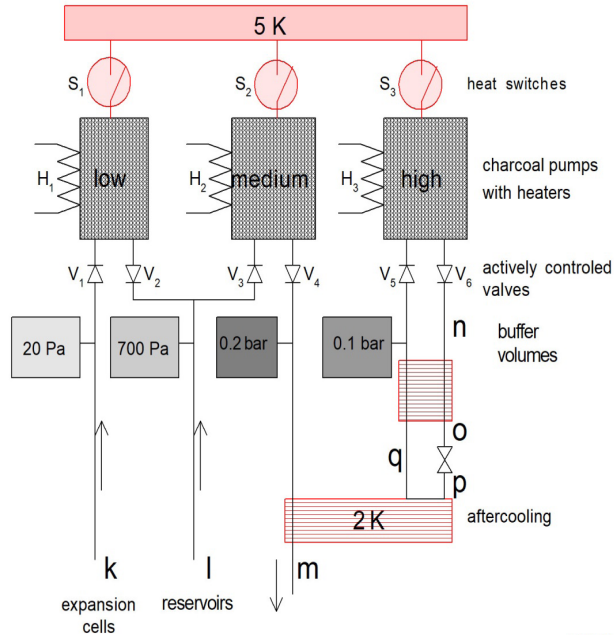


Figure 5. Adsorption-pump layout. The labels k-q refer to Figure 4. Not shown is that the ^3He , leaving the charcoal pumps at $V_{2,4,6}$ is cooled by the 5 K platform.

COMPRESSOR UNIT

The gases are compressed by sorption compressors which are discussed here in more detail. There are three adsorption compressors, shown in Figures 5, which run through cycles which are described in Table 1. Unfortunately the data about the adsorption properties of ^3He on adsorbates is scarce. However, there is evidence that the adsorption properties of ^3He and ^4He are almost the same. First of all Daunt *et al.* [12] measured the adsorption properties of ^3He and ^4He on zeolite. It turns out that the adsorption properties of ^3He and ^4He , if expressed in mol per gram zeolite, almost coincide. Duband *et al.* made a similar observation [13]. So we will base our numerical estimations on the measurement of the adsorption properties of ^4He in particular by [14]. The cycles are indicated in Figure 6. The resulting numbers are given in Table 2.

The aftercooler should have a temperature that is so low that the helium leaving V_4 liquefies at a pressure of 0.2 bar. That means that the pressure at point **p** in Figures 4 and 5 should be below 0.2 bar. Here is chosen for a pressure of 0.1 bar with a corresponding temperature of 1.7 K which is well below the required 2 K.

The buffer volumes, indicated in Figure 5, reduce the pressure buildup during the times that the suction valves $V_{1,3,5}$ are closed. The size of these volumes depends on how much pressure oscillations can be tolerated. E.g. the volume of the gas flow of $4.84 \mu\text{mol/s}$ in the 20 Pa channel in one

Table 1. Valve, heater, heat-switch positions, and temperature evolution during the cycles given in Figure 6. T_a is the temperature of the adsorber.

period	$V_{1,3,5}$	$V_{2,4,6}$	$H_{1,2,3}$	$S_{1,2,3}$	T_a
adsorption	open	Closed	Off	closed	decreasing
pressure increase	closed	Closed	On	open	increasing
exhaust	closed	Open	On	open	increasing
pressure decrease	closed	Closed	Off	closed	decreasing

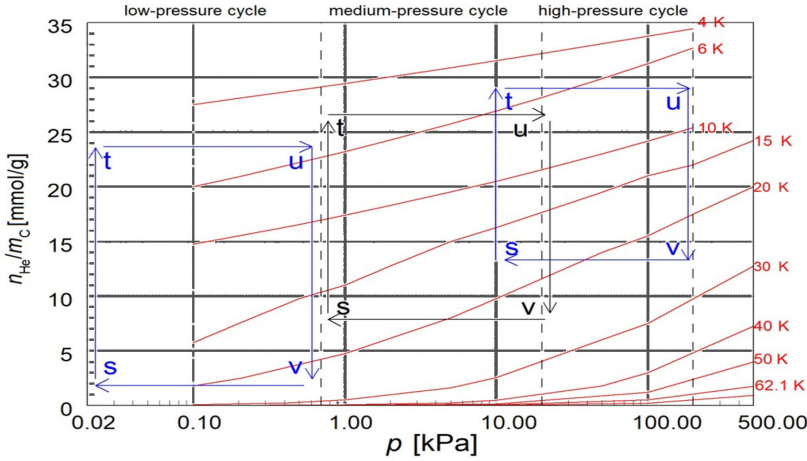


Figure 6. Adsorption isotherms of Barnebey-Cheney [14]. The three pumping/compression cycles are shown. In this example a platform temperature (points t) of 5 K and exhaust temperatures of 25 K (points v) were chosen. Strictly speaking these curves should include not just the adsorbed helium but also the helium in the free volume of the cell.

Table 2. Estimated values of the compressor properties as given in Figure 6 based on [14]. The differences between adsorption (s→t) and release (u→v) adsorption rates are about 20 mmol ³He per gram charcoal.

	s	t	u	v
low pressure				
T [K]	15	5	5.5	25
p [kPa]	0.02	0.02	0.70	0.70
μ [mmol/g]	2	24	24	2
medium pressure				
T [K]	18	5	6.5	25
p [kPa]	0.7	0.7	20	20
μ [mmol/g]	8	27	27	8
high pressure				
T [K]	17	5	8	25
p [kPa]	10	10	200	200
μ [mmol/g]	13	30	30	13

hour at 0.5 K is 3.6 liter and the flow of 368 $\mu\text{mol/s}$ at 700 Pa and 2 K gives a volume of 32 liter. If the volumes of the buffers would be too big it is an option to store the helium as a liquid instead of as a gas. Another option is to synchronize the cycles of the charcoal pumps with the expansion cycles and close the suction valves when the demand for cooling by E or R is small or zero. One can avoid the buffers altogether by splitting the sorption compressors in two compressor units that work in opposite phase.

The amount of ³He determined by flow rate n_E has to be stored in the absorber of the low-pressure stage of the pumping system. If the cycle time of the charcoal pump is 1 hour this corresponds with 17.4 mmol of ³He. With 20 mmol ³He per gram charcoal per cycle we would need about one gram of charcoal. For the medium-pressure circuit in one hour we need to adsorb $3600 \times (4.84 + 428) \times 10^{-6} = 1.56$ mol. With 20 mmol/g per cycle we would need $1560/20 = 78$ gram of charcoal.

The high-pressure cycle needs to cool a flow $n_E + n_R$ at 20 kPa from 5 K to liquid at 2 K. This requires a cooling power of $(n_E + n_R)(H_m(5\text{K}, 20\text{kPa}) - H_m(2\text{K}, 20\text{kPa})) = (4.84 + 428) \times 10^{-6} \times (121.9 - 9.9) = 48$ mW. The maximum cooling power at 2 K in the high-pressure circuit is equal to the enthalpy difference between 0.1 and 2 bar at 5 K [15] which is $121.9 - 97.2 = 24.7$ J/mol. So the minimum

Table 3. Some estimated quantities based on a one hour cycle time.

Property	amount
^3He circulation rate in low-pressure channel	4.84 $\mu\text{mol/s}$
^3He circulation rate in medium-pressure channel	428 $\mu\text{mol/s}$
^3He circulation rate in high-pressure channel	1.94 mmol/s
charcoal in low-pressure compressor	1 gram
charcoal in medium-pressure compressor	78 gram
charcoal in high-pressure compressor	351 gram
amount of ^3He in one expansion cell	0.12 mol
amount ^3He in compressors	8.5 mol

circulation rate in the high-pressure circuit is $48/24.7 = 1.94$ mmol/s; in 1 hour 7 mol. With an effective adsorption capacity of about 20 mmol/g this requires 351 gram charcoal. With an apparent density of 382 kg/m^3 [14] this takes a volume of 910 cm^3 .

DISCUSSION

Adsorption pumps

The three adsorption pumps operate between the same temperature levels (5 K and 25 K). If their operation cycles are also synchronized it is an option to put all three in one casing with only one heat switch.

Generally speaking one should keep the recovery times (during $t \rightarrow u \rightarrow v \rightarrow s$) as short as possible. In this respect it is interesting that graphite has a finite electrical resistivity so that it can be warmed up by sending an electrical current through it so that the graphite heats itself during $t \rightarrow u \rightarrow v$ and we don't have to deal with the thermal penetration depth as in the case the graphite is heated from outside.

Adsorber

The adsorption energies of carbon and zeolite are very high. This leads to a significant heat load during the adsorption and high temperatures are needed to desorb the helium. In our example this is 25 K. First heat has to be supplied to warm the container with its content up to 25 K and then the hot gas and the container have to be cooled to 5 K. Thus the high adsorption energies lead to a significant reduction of the system efficiency. Furthermore it extends the cycle time which in turn affects the amounts of charcoal and helium needed and the size of the system. It may be worth finding adsorbers with lower adsorption energies. It may be hard to find solids with a smaller Van der Waals attraction than graphite or zeolite. So one might consider coating the adsorber with a monolayer of argon, neon, or ^4He by adding a specific amount of one of these gases to the ^3He in the pumping units so that a monolayer is formed on the adsorber.

Thermal buffer

It is possible to obtain continuous cooling with only one E-R unit if a thermal buffer, with a high heat capacity, is put in between the application and E. The buffer has to stay below 50 mK during a complete cycle of the expansion unit. A heat switch between the buffer and the expansion cell thermally connects E with the buffer during the cooling period of the cycle and disconnects it during the recovery period. During the cooling period the cooler removes all the heat flowing into the buffer during one complete cycle. As a result of the oscillating temperature of the buffer the temperature of the application also varies. If that is a problem some control is needed to keep the application temperature constant which reduces the efficiency of the system.

If the cooling and recovery periods both take half an hour we have to deal with a heat load of $10 \mu\text{W}$ during half an hour which corresponds with 18 mJ. E.g. FFA (FeNH_4 alum) has a molar

heat capacity of $C_m = 5$ J/molK in the 50 mK range [16]. With a molar weight of 0.48 kg/mol this gives a specific heat capacity of FFA of 10.4 J/kgK. If we allow a temperature increase of 10 mK it can adsorb 0.1 J/kg. In order to adsorb 18 mJ, we would need 0.17 kg. With a density of 1710 kg/m³ this has a volume of 0.1 liter.

Instead of a thermal buffer one can also think of a demagnetization unit just like in the final stage of an ADR. As this operates around 50 mK it requires magnetic fields of only 0.1 T with modest shielding (or compensating coils) and small energy demand and volume. As such the system would be a hybrid system with ADR and expansion units.

ACKNOWLEDGMENT

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