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MAGNETIC REFRIGERATION FOR LOW-TEMPERATURE APPLICATIONS

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An increasing number of applications require refrigeration at low temperatures ranging from production of liquid helium for medical imaging systems to cooling of infrared sensors on surveillance satellites. Cooling below about 15 K with regenerative refrigerators is difficult because of the decreasing thermal mass of the regenerator compared to that of the working material. In order to overcome this difficulty with helium gas as the working material, a heat exchanger plus a Joule-Thomson or other expander is used. Regenerative magnetic refrigerators with magnetic solids as the working material have the same regenerator problem as gas refrigerators. This problem provides motivation for the development of non-regenerative magnetic refrigerators that span ~4 K to ~20 K. Several laboratories around the world have magnetic-refrigeration programs underway; some are working on 4 - 20 K refrigerators. In the development efforts, particular emphasis has been placed on high reliability and high efficiency. Detailed calculations indicate considerable promise in this area, but several key problems have been identified in each of several possible devices. The principles, the potential, the problems, and the progress towards development of successful 4 - 20 K magnetic refrigerators are discussed.

Key words: Carnot cycle; low temperature; magnetic; non-regenerative; refrigerator; review.

I. INTRODUCTION

Magnetic resonance tomography and magnetic-field-gradient ore separation are two examples of developing commercial applications of superconducting magnets.[1,2] Many other potential users for superconducting magnet systems exist.[3] All of the superconducting magnet systems will require liquid helium or closed-cycle refrigeration near 4 K. Long wavelength infrared (LWIR) sensors need to be cooled to 8 - 10 K to obtain an adequate signal-to-noise ratio for high sensititivity.[4] Small helium liquefiers and some LWIR systems require approximately 1 W of cooling power. At this size, the efficiency of conventional refrigeration systems is typically a few percent of Carnot efficiency for heat rejection near room temprature.[5] Mean periods between failures or major maintenance are several thousand hours.

Regenerative gas refrigerators typically cease operation near 15 K because of decreasing thermal mass of the regenerator material. Hence both regenerative and recuperative gas refrigerators have a recuperative low-temperature stage plus an expander, such as a Joule-Thomson (J-T) device.[6] The efficiency of the bottom heat exchanger and J-T expander operating from 15 - 20 K down to ~4 K is 50-60% of Carnot if carefully designed.[7] The J-T loop suffers from poor reliability and requires a high-pressure compressor for operation.

If a simple, non-regenerative, non-recuperative 20 - 4 K refrigerator with high reliability and high efficiency (greater than 60% of Carnot) could be developed, it would offer significant improvements to existing refrigerator systems by eliminating the J-T loop and its associated problems. Refrigerators based on the magnetocaloric effect that execute a magnetic Carnot cycle between ~15 - 20 K and ~4 K can potentially satisfy these requirements. This conceptual review paper will try to illustrate the development of magnetic refrigerators for the 20 - 4 K temperature region. The reference list is not complete but is representative.

II. PRINCIPLES

A. Materials

All refrigeration processes require an entropy change to absorb heat from a cold thermal source and deliver it to a higher temperature sink. Therefore, the entropy-temperature (S-T) diagrams for the working magnetic materials are essential to understanding the operation of magnetic refrigerators. In the 4 - 20 K region several types of magnetic materials can be used; the most obvious ones are paramagnets. There are many criteria for selection of suitable refrigerants including low lattice specific heat and a large magnetic moment. A survey of existing and potential paramagnetic materials has been published. [8] This and other work [9,10,11,12] have shown that gadolunium gallium garnet (GGG) is an excellent initial choice for 4- 20 K magnetic refrigerator design. Figure 1 shows the S-T curves for GGG as a function of aplied magnetic field.

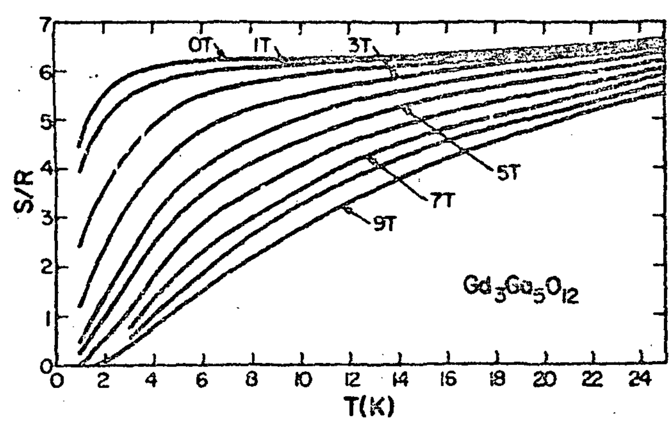

Figure 1: The entropy of gadolinium gallium garnet as a function of temperature and magnetic field.

It is also possible to consider composites of paramagnets such as $DyVO_4$-GGG as suggested by the Grenoble group.[13] Physical mixing of materials allows some freedom in shaping the entropy-temperature diagrams which, in turn, allows cycles other than the Carnot cycle to be considered. Figure 2, taken from reference 13, shows the S-T curves for a 50-50% composition of the materials. As will be explained in the next section, this composite would be unsuitable for a magnetic Carnot cycle but is well suited to a magnetic Ericsson cycle, as was suggested in reference 13. The entropy change at 20 K in the composite is comparable to that in GGG alone, but the parallel nature of the S-T curves is essential for effective regeneration if a good regenerator material is used.

Finally, ferromagnetic materials can also be considered as working materials in this temperature range. An example is EuS whose calculated S-T curves [14] are shown in Fig. 3. Other ferromagnets such as GdRh [15] could be used although rhodium is extremely expensive. GdRh has a Curie temperature near 20 K but lower Curie temperature materials are available. For example, the ErRh-GdRh series has Curie temperatures ranging from ~6 K to ~20 K.[16]

B. Cycles

The only non-regenerative, non-recuperative cycle that exists for use in the 4-20 K range is the magnetic Carnot cycle. This consists of two isothermal stages and two adiabatic stages while magnetizing or or demagnetizing. Characteristics of this cycle are that the magnetic field is continously changing and excellent heat transfer is required during the isothermal parts of the cycle. Because the lattice entropy of many paramagnets becomes comparable to the field-induced entropy change near 20 K, the upper operational limit of the Carnot cycle is near 20 K, depending upon the magnetic field strength and the magnitude of the lattice entropy.

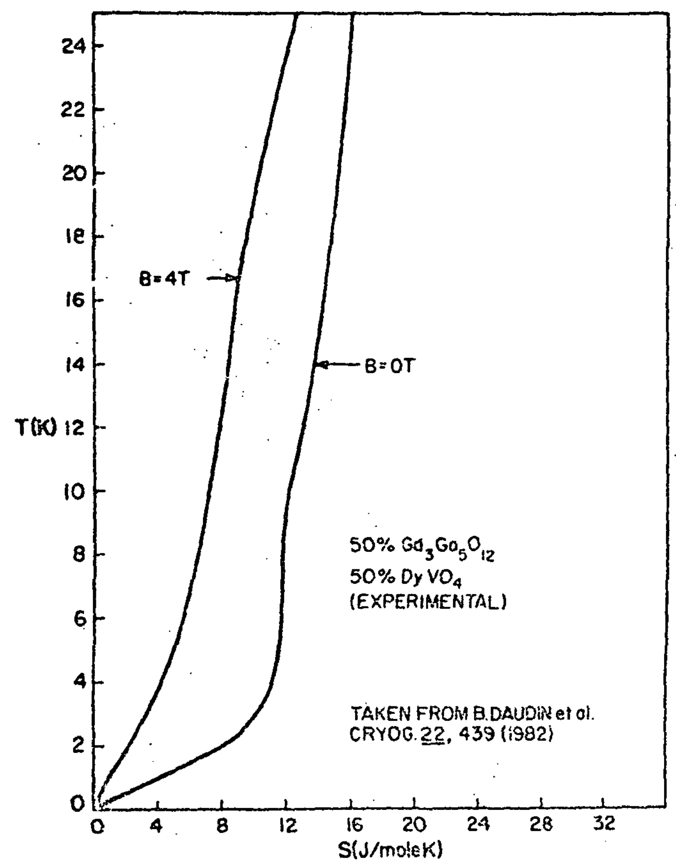

Figure 2: The entropy of 50% gadolinium gallium garnet and 50% Dyprosium Vanadate as a function of temperature and magnetic field.

Other cycles besides the Carnot cycle have possibilities if regeneration or recuperation can be effectively added. The magnetic Stirling cycle (two isomagnetization stages and two isothermals); magnetic Ericsson cycle (two isofield stages and two isothermals) and the magnetic Brayton cycle (two isofield stages and two isentropes are all possible in the 4-20 K range. These cycles are illustrated in Fig. 4 using the entropy temperature diagram of a paramagnetic material. Each cycle has different field-temperature changes required at different parts of the cycle. These requirements will be reflected in the designs of actual refrigerators. The magnetic Stirling cycle using a paramagnet such as GGG is probably a good choice because the amount of regeneration is minimal, as is shown in Fig. 4.

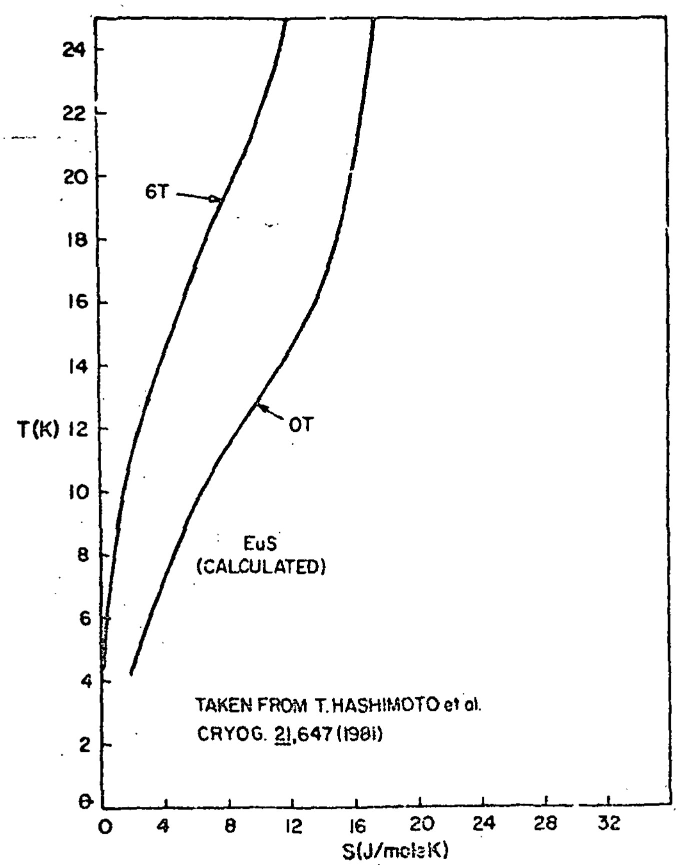

Figure 3: The calculated entropy of europium sulfide as a function of temperature and magnetic field.

Figure 4 also shows that the Brayton and Ericsson cycles are not feasible with a single paramagnetic material because the regenerative heat flows will be completely unbalanced. Composites or ferromagnets will be required for successful implementation of these cycles.

The entropy changes in the various cycles can be obtained from

$$dS = \left(\frac{C_B}{T}\right) dT + \left(\frac{\partial M}{\partial T}\right)_B dB . \qquad (1)$$

Knowledge of C_B and M as a function of B and T allows calculation of the heat and work flows during the idealized cycles. Real cycles, of course, are polytropic and require detailed knowledge of many additional sources of irreversible entropy and heat capacities of thermal addenda in order to be correctly modelled.

III. Potential

A. Designs

A series of ~2 - 4 K magnetic refrigerators have already been built and tested with varying degrees of success.[17,18,19,10] The Grenoble group has reported efficiencies as high as 79% of

Carnot [17] for a 2 - 4 K reciprocating design using GGG. In the 4 - 20 K range there is a large variety of magnetic refrigerator design possibilities. Table I presents an attempt at classification of the possibilities and includes location of groups working on devices of the various types.

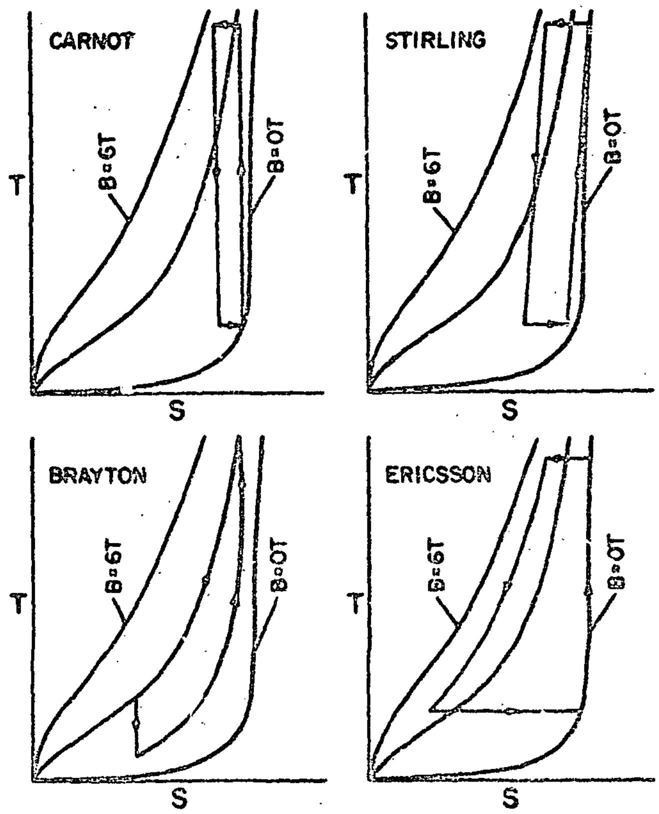

Figure 4: The magnetic cycles illustrated on the entropy-temperature diagram of a paramagnetic material.

Table I. Possible 4 - 20 K Magnetic Refrigerator Designs

	Non-Regenerative Non-Recuperative	Regenerative	Recuperative	Hybrid
Rotational	*LANL, **HAC	Grenoble		
Reciprocating	***J.P.L., Tokyo	LANL		(LANL)
Other	****Tokyo	MIT		(MIT)

*LANL - Los Alamos
**HAC - Hughes Aircraft Co.
***J.P.L. - Jet Propulsion Laboratory
****Tokyo - Japanese groups at Tokyo Institute of Technology, Hatachi-Research Laboratories, and other interacting laboratories.

The majority of devices underdevelopment appear to be non-regenerative, non-recuperative. Both rotating and reciprocating designs have been chosen. The details on most of these devices are not fully known because the various groups have not yet published reports. There are pros and cons to each design. Some of these design choices will become apparent in the discussion of the problems in the next major section.

B. Second-law analysis[21]

The efficiency of a refrigerator executing a thermodynamic cycle between hot and cold temperature, T_H and T_C, respectively, can be written as

$$\eta = \frac{\dot{Q}_C}{\dot{W}_{TOTAL}} \left(\frac{T_H}{T_C} - 1 \right) , \quad (2)$$

where $\dot{Q}_C$ is the reversible cooling power and $\dot{W}_{total}$ is net work flow into the refrigerator. This total work rate includes the pump power, magnet supply power, etc., in addition to the work rate from the refrigerator itself. The refrigerator work rate can be calculated from the Second Law according to

$$\dot{W} = \dot{Q}_c \left(\frac{T_H}{T_c} - 1 \right) + \frac{T_H \int \dot{w}_i \left(\frac{1}{T} - \frac{1}{T_H} \right) dT}{\int dT} + \frac{T_H \int \dot{q}_j \left(\frac{1}{T} - \frac{1}{T_H} \right) dT}{\int dT} + \frac{T_H \int \Delta\dot{S}_{IRR}\, dT}{\int dT} , \quad (3)$$

where $\dot{w}_i$ is the power added externally that the refrigerator must remove, e.g., friction, $\dot{q}_j$ is the power introduced through heat conduction from the surroundings; and $\Delta\dot{S}_{IRR}$ is the rate of irreversible entropy production from different mechanisms. Once a specific design is chosen, a detailed analysis of the device can be done and a projected efficiency obtained. For example, in a Carnot-cycle rotational device designed at Los Alamos, the work rates from each mechanism were calculated and are presented in Table II.

Assuming good heat exchangers, pump efficiencies of 50%, and drive motor efficiency of 90%, the overall projected efficiency for the wheel device was 65% when rotating at 0.2 Hz through a 6-T field to pump 0.67 W from 4.3 K to 15 K. This is an excellent efficiency but only slightly greater than a very well designed J-T loop. However, the low rotational frequency suggests it may have a long lifetime and good reliability.

Table II. WORK RATES FROM VARIOUS MECHANISMS FOR A CARNOT-CYCLE WHEEL REFRIGERATOR

Item		$\dot{W}$ (watts)
$\dot{W}$	reversible	1.64
$\dot{W}_H$	conduction	0.046
$\dot{W}_H$	heat transfer	0.012
$\dot{W}_H$	pressure drop	0.011
$\dot{W}_c$	conduction	0.016
$\dot{W}_c$	heat transfer	0.012
$\dot{W}_c$	pressure drop	0.0001
$\dot{W}$	4-15 K conduction	0.18
$\dot{W}$	friction	0.10
$\dot{W}$	parasitic heat leak	0.10
$\dot{W}_H$	heat exchanger	0.060
$\dot{W}_c$	heat exchanger	0.060

IV. Problems

A. Isothermal Stages

The importance of the need for "isothermal" stages in efficient low-temperature cycles was realized by Jacob [22] some years ago for a Brayton cycle. The isothermal stages are essential for all efficient low temperature cycles for the following reasons. In order to transfer heat to and from magnetic material to a single-phase heat-transfer gas, a temperature difference between the cold magnetic material and the gas (ΔT_C) and hot magnetic material and gas (ΔT_H) must exist. The direct effect of ΔT_C and ΔT_H on efficiency can be calculated from Eqs (2) and (3) by integrating from $T_C - \Delta T_C$ to T_C at the cold stage and T_H to $T + \Delta T_H$ at the hot stage. The total expression is complex, but for only the reversible cooling power component to the efficiency, we obtain

$$\eta = \frac{(T_H - T_C)}{T_C \left[\dfrac{\ln\left(\dfrac{T_C}{T_C - \Delta T_C}\right) \Delta T_H}{\ln\left(\dfrac{T_H + \Delta T_H}{T_H}\right) \Delta T_C} - 1 \right]} \quad (4)$$

For example, between 4 and 20 K, if $\Delta T_C \sim 0.3$ K and $\Delta T_H \sim 1$ K, the efficiency drops from 100% (reversible) to 92.5% from this effect alone. This clearly indicates that magnetic Brayton cycles in the 4-20 K range will be less efficient than the other cycles. While this effect is probably obvious, it is important to remember for operation at very low temperatures.

B. Addenda

The major effect of additional thermal mass from various sources is to reduce the cooling power of a magnetic refrigerator. This is particularly important in non-regenerative designs using the magnetic Carnot cycle because many of the addenda items have temperature-dependent thermal masses that tend to contribute more entropy near 20 K than near 4 K. There are several ways to account for the effects of addenda on the refrigerator, but the total entropy approach is one of the more useful ways. In this approach, the entropy of each item in the refrigerator and that of the magnetic materials are totaled. The change in total entropy caused by the magnetic field is easily obtained.

Some of the addenda items common to many 4 -20 K designs are containers, forms, supports, etc. which must be present to support or contain the paramagnetic material. Because excellent heat transfer between the working material and heat-transfer gas is required, a porous magnetic-material of some geometry along with convected helium gas are used. The porosity of the material can lead to entrained helium gas, which causes an internal thermal load and gas movement during the cycle. The housing around the moving working material doesn't contribute to the addenda once thermal equilibrium is attained, but thermal conduction from hot to cold sections adds a thermal load equivalent to thermal addenda. Figure 5 shows the relative magnitudes of the entropy of the addenda for a particular design. These effects are a severe problem in non-regenerative designs.

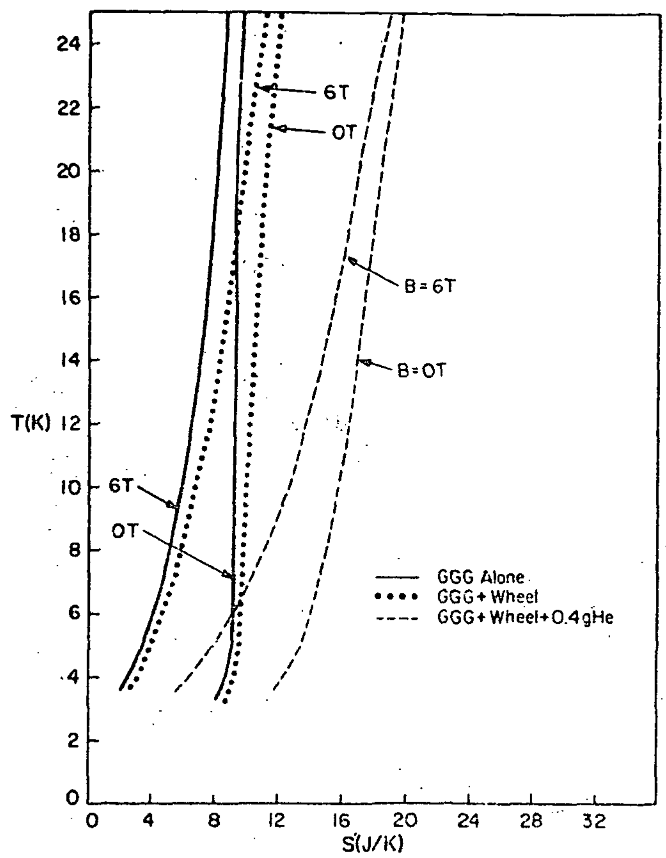

Figure 5: The total entropy of a particular design that shows the effects of various contributions of thermal addenda.

C. <u>Flow Control</u>

In any 4 -20 K refrigerator that uses convective heat transfer between helium gas and the magnetic solid, there will be a flow-control problem. The problem is caused in part by the temperature differences in the refrigerator and in part by the motion of the working material. In a rotating design, either crossflow or counterflow gas motion in sections of the wheel is possible and heat transfer occurs continously. The hot and cold section must be separated by some sealing mechanism, such as clearance or labyrinth seals. Rubbing seals of various designs are possible, but careful attention must be paid to clogging due to small particles from abrasion. Because of the close tolerances in highly effective porous beds, performance could degrade rapidly with clogging. Friction must also be kept to a minimum, especially when it occurs at the cold part of the refrigerator.

Reciprocating designs need flow during the motion of the piston of working material in order to achieve an isothermal stage in a cycle. Clever designs are needed to establish flow through the piston in some fashion and at the same time have isolated hot and cold sections in the refrigerator cylinder.

Pumps are essential to flow control. The pumping power must be fairly small compared to the cooling power in order to attain high efficiency. Efficient, reliable, long-lifetime pumps operating at low temperatures are a real problem area. he small pressure differences and modest volume flow rates are different requirements from most , vious low-temperature pump work [23]

D. External Heat Exchangers

High performance heat exchangers are very important for high efficiency because the temperature differences allowed on each end of the refrigerator must be relatively small according to Eq. 4. The heat exchanger heat-transfer irreversibility for single-coil heat exchangers in a pot of boiling or condensing cryogen, such as liquid hydrogen or helium, can be shown to be given by [24]

$$\frac{\dot{S}_{IRR}}{\dot{m}C_p} = \left(\frac{T_{in} - T_o}{T_o}\right)\left(1-e^{-N_{tu}}\right) + \ln\left(\frac{T_o + \left(T_{in} - T_o\right) e^{-N_{tu}}}{T_{in}}\right) \quad . \tag{5}$$

where $\dot{S}_{IRR}$ is the entropy generation rate, Cp is the fluid thermal mass flow rate, T_{in} is the gas inlet temperature, T_o is the cryogen bath temperature, and N_{tu} is the number of heat transfer units characterizing the heat exchanger. For N_{tu} greater than about 10, Eq. (5) reduces to

$$\dot{S}_{IRR} \cong \frac{1}{2}\ \dot{m}C_p \left(\frac{\Delta T}{T_o}\right)^2 \quad . \tag{6}$$

where ΔT is defined by $T_{in} = T_o + \Delta T$. Equation (6) clearly shows that a small ΔT is important for efficient heat exchangers if the N_{tu} is reasonably high. Exchangers with $N_{tu} > 10$ are readily achieveable.[25]

E. Magnetic Field

1. Forces

The forces between the magnetic material and the magnet can be substantial if high fields and large volumes of material are combined. For example, in a simple axial case where dB/dz might be 70 T/m, 100 cm^3 of saturated GGG in a cylindrical piston will experience a force of ~5400 N (1200 lbs$_f$).Clearly, forces of this magnitude must be carefully considered in any design. Reciprocating devices have the possibility of partially cancelling the forces by using two opposing pistons, but these designs must have supports for the large compressive stress between magnetic pistons. Rotating devices have natural cancellation of forces but have a large load on the drive shaft bearing because the entire wheel is attracted into the magnet.

2. Profile

Reciprocating devices use solenoidal magnets which are rather straight forward but do require some field shaping depending upon the desired cycle. Non-regenerative Carnot cycles require approximately linear field profiles which are readily approximated. Rotating devices allow several possible ways for production of the magnetic field such, as Helmholtz pairs, racetrack coils, and solenoids or bent solenoids. The first two allow axial drive while the latter requires rim drive. The difficulty in all these cases in that the ratio of field at the magnetic material to the field in the magnet windings is large, i.e., 1: 1.2-1.4. This restricts the maxiumum field easily obtainable at the magnetic material when the coils are wound with NbTi superconducting wire.[26] If Nb_3Sn is used to wind the magnets, higher fields are possible but only at higher costs.

V. PROGRESS

There are several magnetic-refrigeration development projects around the world. The comments here are restricted to those with immediate interests in ~4 to ~20 K prototypes.

A. Hughes Aircraft Corporation

As indicated in Table I, HAC has started a magnetic refrigerator development program on low-temperature devices for applications as spacecraft coolers.[27]

B. Jet Propulsion Laboratory

The deep-space communications network uses a series of antennaes with cooled maser amplifiers. The present Gifford-McMahon plus J-T loop coolers have worked for many years, but there is a desire to see if the J-T loop could be replaced by a magnetic stage that would increase efficiency and improve reliability. A report on this project is included in another session of this conference.[28]

C. Massachusetts Institute of Technology

This is a fairly new program working on a magnetic regenerative concept and on hybrid gas-magnetic devices.[29]

D. Grenoble Group

Following the outstanding success of their 2-4 K reciprocating magnetic refrigerator, the French group have started work on a 4-20 K rotational magnetic refrigerator.[30]

E. Tokyo Group

The Japanese group represents several industrial research laboratories in collaboration with Professor Hashimoto's group in the Department of Applied Physics at Tokoyo Insititute of Technology. Their general interest is in low-temperature refrigerators for cryogen liquefaction. They have reported several results on their use of GGG to liquefy helium [31] with a charge/discharge magnet cycle. We understand that they are now modifying their apparatus to operate in a reciprocating mode between ~4 and 20 K.

F. Los Alamos National Laboratory

The 4-20 K rotational refrigerator under development at Los Alamos is part of a broad magnetic refrigeration program. The objectives include basic research to provide a data base for design and prototype development to prove the potential of this technology by constructing and testing working devices. The 4-20 K device is described below in more detail.

1. Description. The rotational concept has several desirable features, such as balanced magnetic forces and continuous refrigeration. The Carnot cycle is also the easiest of the cycles to execute, although the entrained fluid problem must be carefully handled. A magnetic wheel designed to execute a Carnot cycle is shown in Fig. 6. The GGG used in this design is contained in small rectangular compartments on the rim of the wheel. The GGG is in the form of approximately spherical chunks. The wheel is made from stainless steel, and stainless steel screen is used to enclose the inside and outside of each compartment.

The wheel rotates inside a stationary housing that has two duct regions such that helium gas can pass radially through the GGG. The ducts are located at positions where the GGG reaches 20 K as it is magnetized and where the GGG reaches > 4.2 K as it is demagnetized. The external heat exchangers are connected to the ducts and form part of a hermetic system around the wheel that contains the helium gas. Two pumps circulate the helium gas through the wheel and external heat exchangers. The wheel can be driven inside the housing by a magnetic coupling to avoid any cold seals to a rotating shaft. The fabrication tolerances on the wheel and the housing need to be very close, so that clearance seals are formed between the moving wheel and the fixed housing. Because the entire housing contains helium gas, there should be very little movement of helium past these clearance seals.

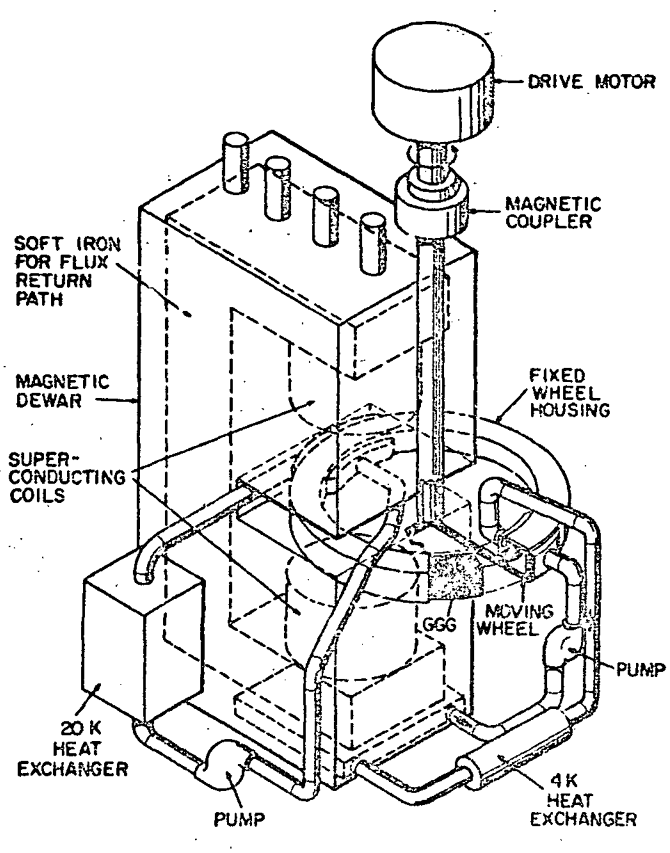

Figure 6: A schematic diagram of a Carnot-cycle, wheel-type magnetic refrigerator for operation between 4.5 and 20 K.

The magnet consists of a pair of solenoids wound on iron cores and supported by an iron yoke for flux return. The resultant c-shaped magnet is designed for a maximum field of ~ 6T in the gap of 2 cm. The leads can be removable so that after the magnet is charged and put in persistent mode, the power supply can be turned down and the leads removed. The magnet is kept at ~ 4.2 K by liquid helium in the dewar surrounding the magnet. Because the whole magnetic refrigerator operates in a vacuum chamber, the liquid helium dewar need only be single-walled and have no superinsulation directly on it.

2. Results

The magnet works well in persistent-mode and produces 6T in the gap where wheel is located. The flow of helium through the wheel is about as expected with respect to the pressure drops, but there is significantly more leakage from duct to duct than expected. There is substantial loading on the central shaft bushing and early versions of the wheel jammed when the field was increased from zero. As an example of the performance, Fig. 7 shows an early curve of the cooling power as a function of temperature span.

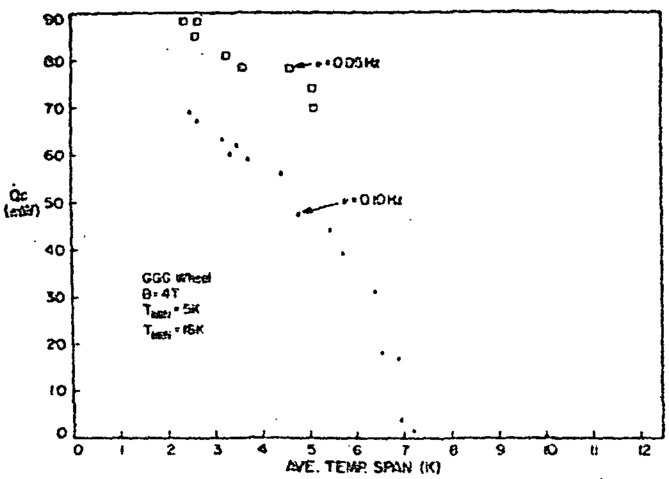

Figure 7: The cooling power as a function of average temperature span for the Carnot-cycle rotational refrigerator.

So far the results indicate that gas movement is the dominant problem, as might have been expected. The efficiency of the overall refrigerator, including drive motor, gear reducers, friction, etc., is low, typically 1% of Carnot but the efficiency of the refrigerator excluding these contributions is ~20% of Carnot. Obviously much more work needs to be done to achieve the design goals but these early results are very encouraging, and we expect they are the beginning of a series of successful devices from the several existing programs.

Acknowledgements

This work was performed under the auspices of the U.S. Department of Energy. Funding support from DOE (BES), DARPA, and NASA (KSC) are gratefully acknowledged.

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