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MAGNETIC REFRIGERATORS FOR USE AT ROOM TEMPERATURE AND BELOW<sup>+</sup>

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Résumé.- Des réfrigérateurs magnétiques à cycle de Carnot sont capables de pomper de la chaleur de façon efficace et peu onéreuse depuis la température de l'hélium liquide jusqu'à celle de l'hydrogène liquide. Aux températures plus élevées, jusqu'à la température ambiante, on peut concevoir des systèmes ferromagnétiques à cycle de Stirling. A basse température les paramagnétiques absorbent des centaines de Joules au litre par désaimantation, tandis qu'aux températures plus élevées les ferromagnétiques absorbent des dizaines de milliers de Joules au litre par désaimantation. On a construit quatre réfrigérateurs magnétiques, mais aucun d'eux n'est économiquement intéressant. Cependant des considérations fondamentales indiquent que les réfrigérateurs magnétiques pourraient remplacer éventuellement les réfrigérateurs à fluide, au moins au-dessous de 80 K environ, où les chaleurs spécifiques de réseau peuvent être rendues petites.

Abstract.- Magnetic Carnot cycle refrigerators should be capable of pumping heat efficiently and inexpensively from liquid helium temperatures to liquid hydrogen temperatures. At higher temperatures, up to room temperature, Stirling cycle devices using ferromagnets are feasible. At low temperatures paramagnets absorb hundreds of joules per liter upon demagnetization; at higher temperatures, ferromagnets absorb tens of kilojoules per liter upon demagnetization. Four magnetic refrigerators have been built, but no economically viable unit is in operation. However, fundamental consideration indicates that magnetic refrigerators should eventually replace gas refrigerators, at least below about 80 K where lattice specific heats can be kept small.

1. INTRODUCTION TO MAGNETIC REFRIGERATION.- More efficient and economical refrigeration would make low temperatures more accessible to researchers and to engineers. Gradually, large scale applications of low temperature phenomena, especially superconductivity, are being introduced into modern technology. Many superconducting magnets are used in particle accelerators and in particle experiments, and large magnets are being used in plasma and fusion research. Magnetically levitated trains, superconducting power transmission lines, superconducting motors and generators and other applications are currently being tested. All of these require refrigeration in the 2 K to 12 K regions.

Oxygen separation for steel production requires huge quantities of refrigeration near 80 K. Large refrigeration capacity at 20 K might be required if hydrogen succeeds as an alternative fuel.

The object of this paper is to show that modern magnetic materials and magnets provide a basis to allow the replacement of gas cycle refrigerators by more economical and efficient magnetic cycle refrigerators, at least below about 80 K. A great deal of diligent and creative hardware development is needed before that goal will be reached, however.

1.1. The Principles of Magnetic Refrigeration.- Application of a magnetic field to paramagnetic materials at low temperatures and ferromagnetic materials near their Curie temperatures causes them to warm up; alternatively, heat is expelled from such materials if the temperature is held constant during the field application. Conversely, removal of the field will cool the material or, at constant temperature, allow absorption of heat by the material. For temperatures at and below room temperature, the temperature changes can be of the order of 10-20 K if fields of about 7 T are applied to an appropriately chosen material.

The principle of a magnetic refrigerator can be illustrated with the conventional Carnot-cycle device shown in figure 1. With thermal switch TS1 closed, thermal switch TS2 is opened, and a magnetic field is applied to the paramagnetic or ferromagnetic working material (WM). The field aligns the magnetic spins in the working material, decreasing the randomness (i.e., entropy S) of the spin system. The spin system is now in good thermal contact with the fixed temperature of the heat reservoir (HR), and heat will flow out of WM into HR. Next, TS1 is opened while TS2 remains open, and the magnetic field is partially removed; the spin system becomes partially randomized, requiring energy and thus cooling WM to the temperature of the heat

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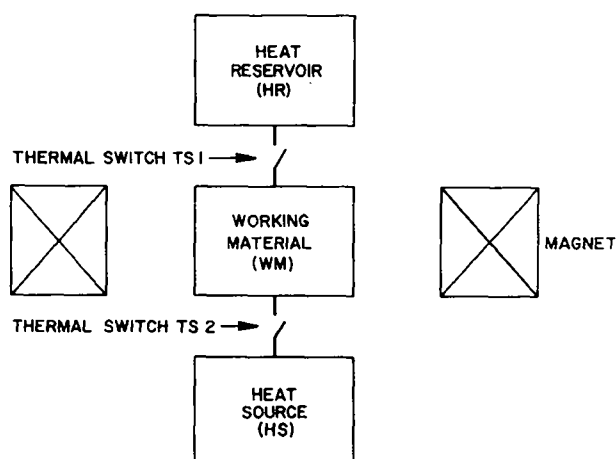


Fig. 1 : Schematic diagram of conventional magnetic Carnot refrigeration principles.

source (HS). TS2 is then closed while the magnetic field is decreased to zero, completing the spin-randomization process and allowing heat to be absorbed from HS. TS2 is then opened and a small magnetic field applied, so that WM warms to the temperature of HR. The cycle can be repeated if TS1 is closed as the full field is again applied to WM.

1.2. The Basic Concepts of Room Temperature Magnetic Refrigeration.- The magnetic Stirling-cycle refrigerator takes a ferromagnet, cools it in a magnet then removes it from the magnet, requiring a large input of work. The ferromagnet further cools upon demagnetization, allowing it to absorb heat. The ferromagnet is then heated and inserted into the magnet. The ferromagnet further warms, expelling heat at the higher temperature. Magnetic heat engines use the reverse of this cycle.

The remainder of this paper will be devoted to recent developments in the low temperature adiabatic demagnetization refrigerators working on a magnetic Carnot cycle and in the higher temperature magnetic refrigerators working on a magnetic Stirling cycle. All the details of magnetic refrigerators will involve the entropy concept and entropy calculations. Magnetic devices, like all high efficiency refrigerators and heat engines, are best approached in terms of entropy, which is approximately conserved in high efficiency devices.

1.3. Actual Entropy-Temperature Diagram Illustrating the Cycles.- Figure 2 illustrates the Carnot cycle that could be executed by low temperature magnetic refrigerators like the one shown in

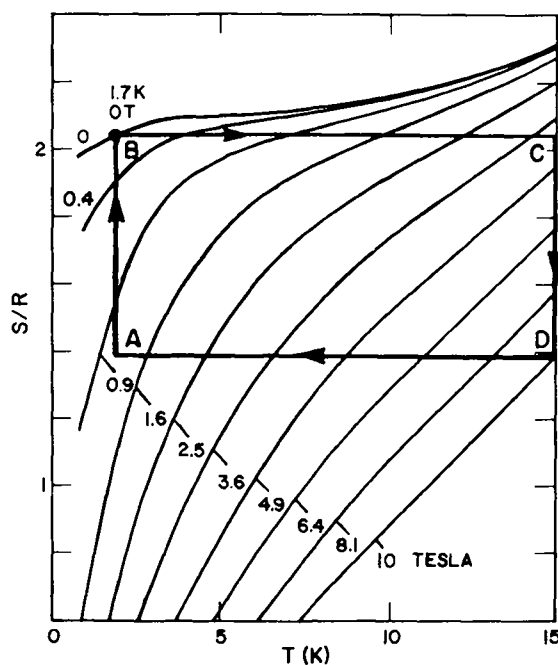


Fig. 2 : Thermodynamic cycle executed by  $Gd_2(SO_4)_3 \cdot 8H_2O$  in a magnetic Carnot cycle. Entropy per gram ion of  $Gd^{+++}$  versus temperature for various applied fields is obtained from reference /1/. Utilization of  $Gd_2(SO_4)_3$  or  $GdPO_4$  would reduce the magnetic field requirements and allow heat expulsion at higher temperatures without any appreciable performance deterioration.

figure 1. The material illustrated is the paramagnet  $Gd_2(SO_4)_3 \cdot 8H_2O$ .

From a heat source at 1.7 K, this material would absorb  $Q = T\Delta S = 9.9$  J/mol. each cycle ( $\Delta S$  is the entropy change at temperature  $T = 1.85$  K, as shown in figure 2, and  $R$  is the gas constant). This represents 82 J/l of  $Gd_2(SO_4)_3 \cdot 8H_2O$  each cycle. This number is expected to be two or three times as large for more dense /1/  $Gd_2(SO_4)_3$  and  $GdPO_4$ ; however, measurements are needed to establish the shape of the zero field entropy curve near 2 K in these materials. Such work is currently underway at Los Alamos Scientific Laboratory (LASL).

Figure 3 illustrates the Stirling cycle executed by higher temperature magnetic refrigerators or heat engines. The material illustrated is the ferromagnet gadolinium metal. It is necessary to use a ferromagnet for refrigeration above 10 or 20 K because it is impossible to provide enough magnetic field to make  $\mu H \approx kT$  as is required to remove entropy in a paramagnet. Here  $\mu$  is the magnetic moment of the ion,  $H$  is the applied field and  $k$  is the Boltzmann constant. In a ferromagnet near

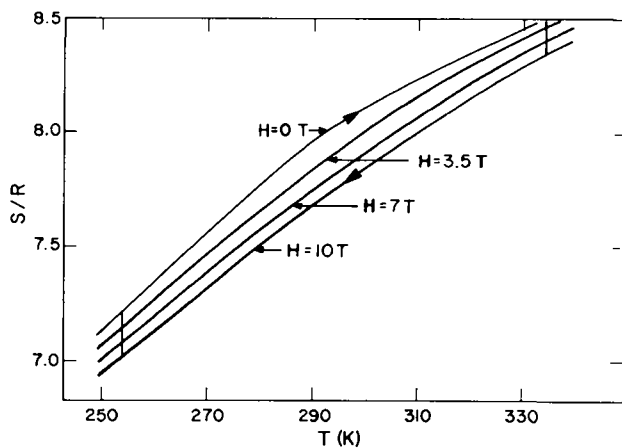


Fig. 3 : Experimental and calculated entropy for Gd metal. The experimental data of reference /2/ at 0 and 7 T are within 0.01 of the calculated results. The results are obtained from a simple molecular-field calculation using measured spin  $J = 7/2$ ,  $g$  factor  $g = 2$ , Curie temperature = 293 K and lattice and electronic molar specific-heat sum = 3.5 R. No free parameters are available for this calculation. A similar calculation allows an accurate evaluation of the magnetocaloric effect in iron metal near its Curie temperature.

the Curie point there are large ferromagnetic spin-spin interactions. These interactions supplement the very much smaller  $\mu H$  interaction and reduce the entropy in an applied field. From a heat source at 254 K, this material (figure 3) could absorb  $Q = 422$  J/mol. or 21 kJ/l. Section 2 of this paper will show some devices which provide for the execution of a Stirling cycle by the gadolinium.

During the course of the measurement of the temperature-entropy curves of figures 2 and 3 and in measurements on numerous other materials, tests were made to determine the reversibility of the magnetic heating and cooling process /1,2/. High field magnetization and demagnetization was found to return the material to the original temperature although the temperature excursions were very large. Thus, no entropy creating irreversibilities could be detected in these tests.

## 2. SOME APPROACHES TO ACTUAL DEVICES.-

2.1. Two-Switch Refrigerator of Figure 1.- The units built so far /3,4,5/ are limited to operation below 1 K because of the character of the superconducting switches which they use. Refrigerators /6/ have been proposed which use magneto-resistive switches /7/, such as single crystals of beryllium, with switching ratios of 1000 at temperatures as high as 15 K. These would pump several watts from

2 K to 10 K with 70 % of Carnot efficiency using a Carnot cycle.

Better performance could be obtained if heat were carried to and from the paramagnetic working material by a liquid made to flow through a porous paramagnetic salt. Higher temperature operation would be feasible since switch limitations would not enter. Such a device would probably be very similar to reciprocating units discussed below.

2.2 Reciprocating Units.- The Stirling cycle reciprocating refrigerators, figure 4, proposed by Van Geuns /8/ have been developed by Brown /2/ and Barclay et al. /9/. Brown's latest results /10/

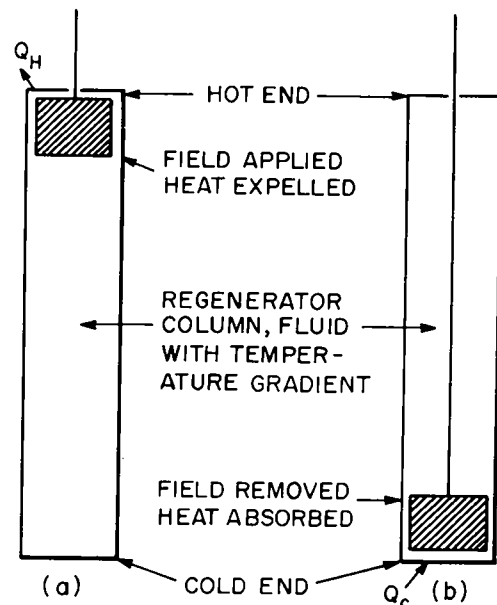


Fig. 4 : Operation of reciprocating Stirling cycle (figure 3) refrigerator. Heat  $Q_H$  is expelled (a) through the magnetization of the working material at a high temperature. The working material is then moved down through the refrigerator column to the cold end. Heat is expelled into the large thermal capacity fluid in the process. Now, the field is removed from the working material /6/, absorbing heat  $Q_C$  and providing the required refrigeration. The cycle is completed when the working material is moved back up through the regenerator column, absorbing the previously deposited heat from the column. Note that there are three primary objects in this refrigerator, the working material, the regenerator column, and the magnet (not shown). Any two of these can be moved in order to accomplish the cycle shown. (At this stage of superconducting magnet development, it is probably impractical to attempt cycling the magnet on-and-off at the required rate).

show the production of an 80 K gradient (centered near room temperature) in the column. The rate of gradient development, following initial startup of

the gadolinium refrigerator shows 34 W capacity operating at about 0.05 Hz.

Barclay's device, using  $\text{Gd}_2(\text{SO}_4)_3 \cdot 8\text{H}_2\text{O}$  as a working material, pumped heat into a 4.2 K bath. The projected performance was an 8 W capacity from a heat source at 2 K at 1 Hz operation.

2.3. Rotating Devices.- Figure 5 illustrates the operation of the magnetic Stirling-cycle wheel as a refrigerator /11/. At the lower right-hand side, the fluid at  $T^H$  enters the porous wheel; in a conceptual device with perfect heat transfer, the wheel is also at  $T^H$  at this point. The fluid flows through the porous wheel, in contact with the wheel which was at temperature  $T^C$  after leaving the high-field region. It exchanges heat with the wheel,

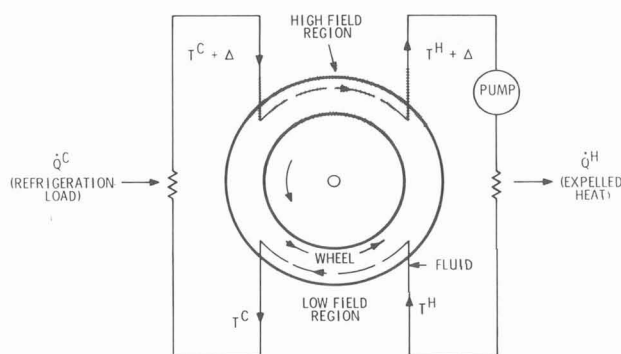


Fig. 5 : Stirling-cycle wheel in refrigerator mode. Ideal heat transfer with  $T^H$ ,  $T^C$ ,  $T^H + \Delta$  and  $T^C + \Delta$  being the temperature of the wheel and fluid at the points where the fluid enters or leaves the wheel;  $\Delta$  is the inherent temperature change of the working material upon entering and leaving the field. See text for description of operation.

exiting the wheel at temperature  $T^C$ , the same temperature as the wheel (again in the case of perfect heat exchange and flow balance). The fluid warms by an amount  $\Delta$ , picking up heat  $\dot{Q}^C$  from the low-temperature heat source and reenters the wheel at temperature  $T = T^C + \Delta$ . In exchange with the wheel, which was at temperature  $T^H + \Delta$  after entering the high-field region, the fluid warms to  $T^H + \Delta$ . It deposits the heat  $\dot{Q}^H$  in the high-temperature heat sink, completing the cycle as it reenters the wheel at temperature  $T^H$ . Note that in this idealized system no heat flows across any large temperature gradient and there is no entropy production as would be the case if the fluid entered or left the wheel at a temperature appreciably different from the wheel temperature at that point.

Note that the high and low temperature extremes of this cycle are adiabats instead of the isotherms of figure 2.

A miniature version of such a device /12/, with gadolinium in the form of 0.5-mm sheets and using the field of a permanent magnet, was built and tested at Los Alamos. Because of excessive flow leakage and mechanical friction caused by the force of the magnet on the sheets, it was able to carry only a few watts over a few K temperature droop. However, the device demonstrated a Stirling rather than Carnot cycle. A more sturdy 15 cm-diameter wheel designed to pump 1 kW is currently under construction at Los Alamos /13,14/ for the Electric Power Research Institute (EPRI). It uses a porous matrix composed of fine lathe turnings of gadolinium metal. Water is the heat transfer fluid.

Figure 6 shows a low temperature rotating refrigerator /15/ operating on a Carnot cycle similar to that shown in figure 2. On the upper left-

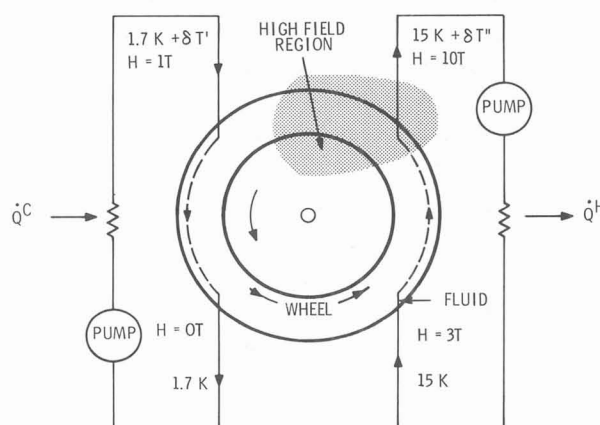


Fig. 6 : Schematic of Carnot wheel refrigerator. The paramagnetic material forming the rim of the wheel rotates in a counterclockwise direction. Supercritical helium is cooled a small amount,  $\delta T'$  by the demagnetizing material on the left-hand side and is warmed a small amount  $\delta T''$  by the magnetizing material on the right-hand side. Power  $\dot{Q}^C$  is absorbed from the load and  $\dot{Q}^H$  is deposited in the upper stage refrigerator. Magnetic field (H) and temperatures are indicated. In an actual device, the pumps would be located at the highest possible temperatures. The magnitude of  $\delta T'$  is chosen to suit the particular refrigeration requirement;  $\delta T''$  is chosen to suit the particular upper stage refrigerator.

hand side of figure 6 and point A of the cycles in figure 2, supercritical helium enters the porous wheel and is forced to flow in heat exchange with the moving wheel. (Reference /15/ gives one exam-

ple of how the heat exchange might be implemented). The wheel absorbs heat from the helium as it is demagnetized to point B in the cycle shown in figure 2 ; the helium cools to 1.7 K. The helium then absorbs heat  $\dot{Q}_C^C$  from the load and reenters the wheel heat-exchanger area at a slightly warmer temperature. Similarly, in the lower right-hand side of figure 6, supercritical helium (at essentially the same pressure as the helium in the left-hand side) enters the porous wheel at 15 K ; this corresponds to point C in the cycle in figure 2. The wheel deposits heat in the helium as the material is magnetized to point D, while the fluid leaves the wheel at a slightly warmer temperature. This heat  $\dot{Q}^H$  is deposited externally, completing the cycle, with the net result that work is used to rotate the wheel and heat is absorbed at a low temperature and expelled at a high temperature. Note in figure 6 that the wheel is adiabatically magnetized as it goes from the lower left-hand to the lower right-hand side and adiabatically demagnetized as it goes from the upper right-hand to the upper left-hand side. These are the horizontal sections of the cycles shown in figure 2.

A more primitive type of rotating refrigerator has been built at Los Alamos /1/ ; it works also on a Carnot cycle but without using forced flow through a porous wheel. In steady state operation it carries 0.2 W from an electrical heater in a superfluid helium bath at 2.1 K into a boiling helium bath at 4 K. The refrigerator has maximum capacity at 0.25 Hz.

3. FUNDAMENTAL LIMITATIONS ON MAGNETIC REFRIGERATORS.- The Magnetic refrigeration process itself is essentially reversible. In a refrigerator that is mechanically optimized (i.e., minimum leakage around fluid seals, minimum mechanical friction, minimum heat leak, etc.) performance will be limited by heat transfer considerations. The question then becomes, how fast can heat be absorbed and expelled by the working material without excessive entropy production associated with heat flowing across a finite temperature difference  $\Delta T$  and heat transfer fluid flowing across a finite pressure drop  $\Delta P$  ?

3.1 Allowable Temperature Drops and Pressure Drops.- Consider a refrigerator absorbing heat  $Q_C$  and entropy  $S_C$  each cycle at the cold temperature

$T_C$ . The work input  $W$  allows the expulsion of heat  $Q_H$  and entropy  $S_H$  at the hotter temperature  $T_H$ . Inside the real irreversible refrigerator there is a net entropy produced  $\Delta S$  after the refrigerator has returned to its initial state upon completion of one cycle. Because

$$S_H = S_C + \Delta S \quad (1)$$

and from the First law

$$W = Q_H - Q_C \quad (2)$$

we can deduce that

$$W = (T_H - T_C)S_C + T_H\Delta S \quad (3)$$

Thus, the work in the irreversible refrigerator, compared to the work  $W_R$  in a reversible refrigerator where  $\Delta S = 0$ , is

$$\frac{W}{W_R} = 1 + \frac{\Delta S T_H}{S_C(T_H - T_C)} \quad (4)$$

The relationship (4) can be used to estimate the allowable heat transfer fluid  $\Delta P$  and fluid-magnetic material  $\Delta T$  in a refrigerator. In a mechanically optimized refrigerator,

$$\Delta S_P = \Delta PV/\bar{T} \quad (5)$$

is the entropy net production associated with the conversion of work from a 100 % efficient pump in frictional heat at mean temperature  $\bar{T}$ . The volume of fluid flowing through each cycle is  $V$ . It can be shown that

$$\Delta S_T \approx Q\Delta T/T^2 \quad (6)$$

is the net entropy production associated with an amount of heat  $Q$  flowing across a temperature difference  $\Delta T$  at a mean temperature  $\bar{T}$ . The sum of  $\Delta S$  from (5) and (6) when entered into (4) must not lead to excessive work  $W$ , or the refrigerator is very inefficient. Note that in a real refrigerator, if  $\Delta S$  is too large, this simple approach fails because the working material no longer follows a cycle approaching the ideal cycle of figures 2 or 3 ; (6) underestimates the performance deterioration associated with  $\Delta S$ .

This simple entropy calculation is useful for magnetic refrigerator design [or magnetic heat engine design with the appropriate changes in (4)]. In section 3.2. we will examine one particular refrigerator.

3.2. Pressure Drops and Temperature Differences in a Particular Refrigerator.- As an example of the use of (4), (5) and (6) in refrigerator/heat

engine evaluation, consider heat exchange by water flowing through a porous gadolinium matrix in the rotating refrigerator at room temperature. Using the subdivision approach of figure 7, we can arrange to keep the flow velocity of the water through the matrix to  $10^{-2}$  m/s at a rotation rate of 1 Hz /14/. From figure 3, in a field of 10 T,  $S_C=1.7$  J/mol-K. In 40-mesh 70 % porous gadolinium filings

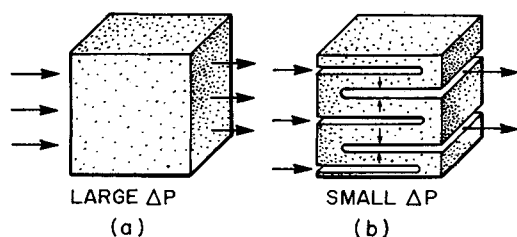


Fig. 7 : The approach required to minimize temperature and pressure drops in flow through porous refrigerator working materials. Heat transfer is a very weak function of flow velocity  $v$  while  $\Delta P$  is a strong function of  $v$  /15/. Consider the arrangement of (b) with a total of 8 slits (5 are shown in the figure), and with particles a factor of 3 smaller than in (a). The heat transfer is unchanged, but  $\Delta P$  is a factor of about 7 lower. Practical considerations limit the number of channels which can be cut into the material. Too many channels make the structure mechanically weak or there is too much heat conduction from channel to channel through the working material. Note that properly oriented channels do have the important benefit of making eddy current losses manageable even if the working material is an electrical conductor in a changing magnetic field.

having a thickness of  $11 \mu\text{m}$ ,  $\Delta P = 2.9 \times 10^6 \text{ N/m}^2$  for water moving at  $10^{-2}$  m/s through 1 m of material (This can be deduced from the measurements of reference /14/). The energy dissipation of water flowing in a 1-m cube at  $10^{-2} \text{ m}^3/\text{s}$  during a 1-s cycle is  $Q = 2.9 \times 10^6 \times 10^{-2} = 2.9 \times 10^4 \text{ J/m}^3$ . Because 1  $\text{m}^3$  contains about  $9.5 \times 10^3$  moles, this is a 3 J/mol. or an entropy creation of  $\Delta S = Q/T = 0.01 \text{ J/mole-K}$ .

To calculate heat flow entropy creation from (6), heat transfer estimates in reference /15/ based on experimental data are used. The heat transfer coefficient per unit volume in the gadolinium filings is estimated as  $1500 \times 10^6 \text{ W/m}^3\text{-K}$  or  $1.8 \times 10^5 \text{ W/mole-K}$ . In one cycle we must transfer about 7.300 J/mol. (from figure 3). At 1 Hz  $\Delta T$  is thus  $7.300/1.8 \times 10^5 = 0.041 \text{ K}$  or  $\Delta S = 7.300 \times 0.041/(294)^2 = 0.0035$ . In (4)

$$W/W_R = 1 + \frac{(0.01 + 0.0035) v 334}{1.7 (334-254)} = 1 + 0.033 v \quad (7)$$

(The net entropy creation per cycle is proportional to the frequency  $v$  since  $\Delta P$  and  $\Delta T$  are proportional to  $v$ ). Thus at  $v = 10 \text{ Hz}$  the refrigerator requires 33 % more work than a Carnot refrigerator.

The above calculation illustrates the principle problem with magnetic refrigeration near room temperature. The heat transferred is much greater than the heat pumped. However, the large fluid-solid heat transfer available in a porous matrix makes this acceptable.

4. CONCLUSIONS.- Magnetic refrigerators and magnetic heat engines are in their infancy. To date, no practical unit has been built. However, fundamental considerations like those of the preceding section show that magnetic refrigerator/heat engine operation at rates of 10 Hz with high efficiencies are feasible. Economic evaluations, not reported here, show that for above a few hertz magnetic refrigerators/heat engines are economically viable near room temperature. They also show that at low temperatures, operation somewhat below 1 Hz will result in viable refrigerators.

However, the approach to the design and construction of magnetic units is not clear at this point. Reciprocating and rotating units have been tested. The reciprocating units seem to have the advantage of simplicity. They also have the advantage of being able to operate with a more porous working material where pressure drops are much smaller /16/. They have the disadvantage of requiring a larger volume of fluid flow, as a large amount of fluid is needed to provide a very large thermal capacity compared to the working material. At this stage of development, there is still ample opportunity for creative new approaches to magnetic refrigerator concepts.

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