Journal of Applied Physics

Magnetic refrigeration: Single and multimaterial active magnetic regenerator experiments

M.-A. Richard, A. M. Rowe, and R. Chahine

Citation: J. Appl. Phys. 95, 2146 (2004); doi: 10.1063/1.1643200
View online: http://dx.doi.org/10.1063/1.1643200
View Table of Contents: http://jap.aip.org/resource/1/JAPIAU/v95/i4
Published by the American Institute of Physics.

Magnetic refrigeration: Single and multimaterial active magnetic regenerator experiments

M.-A. Richard
Institut de Recherche sur l'Hydrogène, Université du Québec à Trois-Rivières, 3351 Boulevard des Forges, P.O. Box 500, Trois-Rivières, Québec G9A 5H7, Canada

A. M. Rowe
Department of Mechanical Engineering, University of Victoria, P.O. Box 3055 STN CSC, Victoria, British Columbia V8W 3P6, Canada

R. Chahine[a)]
Institut de Recherche sur l'Hydrogène, Université du Québec à Trois-Rivières, 3351 Boulevard des Forges, P.O. Box 500, Trois-Rivières, Québec G9A 5H7, Canada

(Received 24 September 2003; accepted 1 December 2003)

An investigation of active magnetic regenerators (AMRs) has been performed near room temperature using helium as a heat transfer fluid and a magnetic field of 2 T. To evaluate the impact of the operating conditions, the performance of two reciprocating 90 g gadolinium packed bed regenerators was mapped as a function of the fluid flux and cycle frequency. In addition, two multilayer regenerators of similar mass and dimensions, composed of a layer of gadolinium and a layer of a gadolinium–terbium alloy, were tested and compared to the performance of the Gd-only regenerators. The multilayer regenerators produced a larger temperature span and cooling power compared to the single material regenerators of equivalent mass and geometry (temperature spans of about 20 and 16 K, respectively). Theses results validate the concept of a multilayer AMR, provide useful data for magnetic refrigerator design, and provide better understanding of active magnetic refrigeration. [DOI: 10.1063/1.1643200]

INTRODUCTION

Magnetic refrigeration is based on the magnetocaloric effect (MCE). Near the phase transition of magnetic materials (i.e., the Curie temperature), adiabatic application of a magnetic field reduces the magnetic entropy by ordering the magnetic moments. This results in an increase in the temperature of the magnetic material. The MCE is defined in terms of an adiabatic change in temperature or isothermal change in magnetic entropy. This phenomenon is practically reversible for some magnetic materials; thus, adiabatic removal of the field reverts the magnetic entropy back to its original state and cools the material accordingly. This reversibility combined with the ability to create devices with inherent work recovery makes magnetic refrigeration a potentially more efficient process than gas compression and expansion. To produce refrigeration, the magnetic material is coupled with a heat transfer fluid. While the refrigerant is kept magnetized, the fluid transfers energy to a hot heat sink. Removal of the field reduces the refrigerant temperature to a point lower than it was prior to magnetization. Thus, the material can refrigerate a cold source coupled to the heat transfer fluid. With the magnetic material in the form of a porous solid and a benign substance like water or helium as the heat transfer fluid, magnetic refrigerators can be compact and do not affect the ozone layer or increase the greenhouse effect.

Magnetic refrigerators using a simple magnetic Brayton refrigeration cycle already exist for the ultralow temperature regime (<4 K) but only operate over a small temperature span. The MCE is usually on the order of 2–3 K/T, and is not large enough to allow this simple cycle to produce temperature spans much larger than 10 K (depending upon the strength of the magnetic field applied). For higher temperatures and larger temperature spans, the active magnetic regenerative cycle (AMR) is used.[1] In this cycle, the magnetic material is not only the working material but also a thermal regenerator with the temperature across the porous bed ranging from that of the heat source to that of the heat sink. Magnetic refrigeration technologies for high temperature ranges needed for liquefaction of gases are still under development and require performance improvement. Near room-temperature AMR refrigerator prototypes have been demonstrated using superconducting magnets (4–7 T) and have generated relatively high cooling powers of 600 (Ref. 2) and 100 W.[3] Recently, devices using permanent magnets (around 0.5–2 T) have been tested, but the cooling power is generally lower because of the lower field applied. Prototypes using Gd-only regenerators[4–6] and $Gd_{1-x}Dy_x$ alloy ($x=0.11,0.13,0.16$) regenerators[7] have also been developed. Since the MCE is at a maximum near the transition temperature of magnetic materials, it has been suggested that a way to improve refrigerator performance would be to arrange different refrigerants in layers ordered according to transition temperature.[8]

The governing differential equations that describe magnetic refrigeration cycles are highly nonlinear. Thus, analyti-

[a)]Author to whom correspondence should be addressed; electronic mail: richard_chahine@uqtr.ca

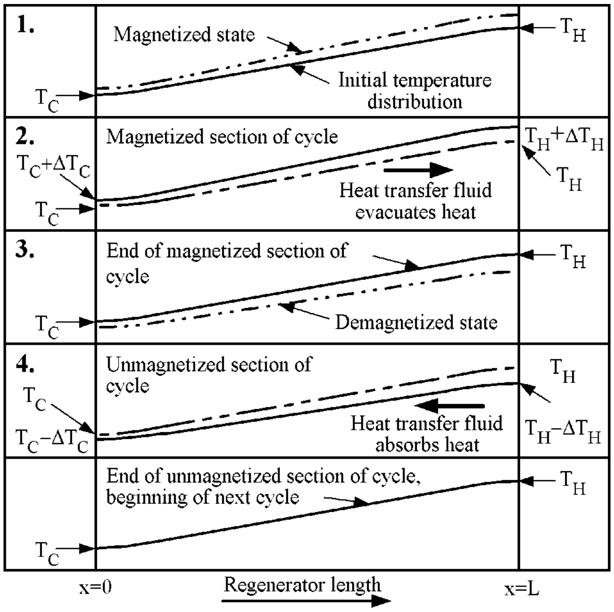

FIG. 1. Diagram of the temperature vs the position of AMR operation.

cal relationships are limited and numerical models often poorly duplicate experimental results. Experimental investigations of active magnetic regenerators are needed to further our understanding of AMR cycles and to provide useful information for magnetic refrigerator design. Although the use of multilayer regenerators instead of single material regenerators is felt to be a way of improving MR performance, the idea requires experimental verification and much more study. The objectives of this work were first to experimentally study the performance of single material regenerators under different operating conditions and, second, to compare single material to multimaterial active magnetic regenerators.

ACTIVE MAGNETIC REGENERATIVE CYCLE

In the AMR cycle, the magnetic refrigerant also acts as a heat storage medium and participates in the cycle much like a regenerator. Going from cold source to hot sink, each section of the regenerator experiences its own refrigeration cycle with the net work at each section dependent on the local MCE value. The cycle is illustrated in Fig. 1 and consists of the following steps.

(1) The regenerator is adiabatically magnetized, causing an increase in temperature throughout the bed determined by the local MCE value.
(2) Fluid flows through the magnetized regenerator from the cold to the hot end. The fluid absorbs heat from the bed and delivers a heat load to the warm sink through a heat exchanger at the hot side of the bed.
(3) The regenerator is adiabatically demagnetized, causing the temperature everywhere in the bed to decrease by the local MCE value.
(4) Fluid flows through the regenerator in the reverse direction, from the hot to the cold end. The fluid is cooled by the bed and can absorb a heat load at the cold section through a heat exchanger. The cycle is then repeated.

Over a cycle the cooling power is determined by the product of the thermal mass and the difference in temperature of the fluid passing through the cold heat exchanger. During magnetization, the magnetic forces draw the regenerator into the field. The magnetization increases when the regenerator is cooled under high field and, thus, larger force must be applied to move the regenerator out of the magnetic field. The work performed to compensate for the difference between these forces constitutes the net theoretical work input in the cycle. In a good design, the forces applied to demagnetize the refrigerant can be partially balanced by the forces produced when magnetizing another quantity of refrigerant.

To transport heat through the entire regenerator, the magnetic material must have an adequate MCE over its length. Because the MCE decreases quickly for most materials as the temperature moves away from the phase transition region, combining many layers of different materials in one regenerator was suggested[8] and modeled[9] as a way to maintain a higher MCE over the entire temperature span, thereby improving performance. Hence, an appropriate multimaterial regenerator should produce a larger temperature span and more cooling power, which are essential for large temperature range devices such as hydrogen liquefiers. To the knowledge of the authors, the only published attempt to experimentally compare a multimaterial layered AMR to a single material regenerator was done in 1990 at the David Taylor Research Center.[10] Using a 7 T field, a 24 K temperature span was obtained with an active magnetic regenerator composed of three equal layers of Gd, a Gd and Tb mixture, and Tb. This was significantly less than the 50 K these researchers obtained using gadolinium in the same apparatus.[11] To overcome these lower than expected results, they suggested establishing an initial temperature gradient in the regenerator and using materials that have closer Curie temperatures. The results may also be attributed to nonoptimal operating conditions and/or arrangement of material in the regenerators.

EXPERIMENTS

Gadolinium, a magnetic refrigerant with a Curie temperature near 293 K, is a well-studied material that produces a relatively large MCE. It was selected as the refrigerant for a single material regenerator and for the first layer of a multimaterial regenerator. A gadolinium–terbium alloy, $Gd_{0.74}Tb_{0.26}$, with a Curie temperature of 278 K, was selected for the second layer of the multimaterial regenerator. The Gd and Tb are commercial grade with purity of 99.9% for both materials. Figure 2 shows Gd (Ref. 12) and $Gd_{0.74}Tb_{0.26}$ (Ref. 13) MCE curves in terms of the adiabatic change in temperature taken from the literature.

Buttons of each material were milled into flakes approximately $120\times700\times1000\ \mu m^3$. Spherical particles would be preferred for testing, but the cost precluded that geometry from being used. The flakes were inserted into cylindrical phenolic shells and bonded with a thin coating of epoxy to form a monolithic layer. The amount of epoxy layer was minimized to avoid negative impacts on the porosity and

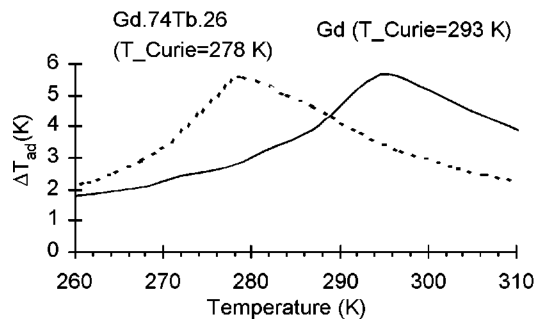

FIG. 2. Gd and $Gd_{0.74}Tb_{0.26}$ adiabatic temperature change curves.

thermal properties of the bed.[14] The regenerator layers had diameter and length of 2.5 cm, mass of approximately 45 g, and porosity of 55%–60%, depending on the material. A sample of Gd was tested before and after processing to see if the isothermal change in entropy was affected. Figure 3 shows the measured magnetic entropy change for a field of 2 T. The difference is within the precision of the measurements, thus the regenerator fabrication method does not significantly affect the MCE of the material.

Experiments were conducted with an active magnetic regenerator test apparatus (AMRTA) built as a tool to dynamically characterize the behavior of magnetic regenerators.[15] A schematic of the system is shown in Fig. 4. It was designed to test two reciprocating cylindrical regenerators moving relative to a 2 T magnetic field generated by a conduction-cooled NbTi solenoid. Layering is along the axes of the regenerators. (Note that Fig. 4 represents the magnet moving relative to the regenerators to simplify the drawing.) Heaters between the regenerators act as heat loads. On the warm ends, heat exchangers that are water cooled act as hot heat sinks (295 K). The heat transfer fluid, helium, is blown through the regenerators by a displacer. Using gas as the heat transfer fluid eases the procedure of changing refrigerants and helium was chosen in view of future low temperature tests. The drawback is that helium has a low volumetric heat capacity in comparison with liquids and limits the thermal flux of fluid, which limits the operating range of the device. The whole system is confined in a vacuum chamber to minimize convective heat leakage to the cold sections. The maximum operating pressure for helium is 10 atm, corresponding to mass flow of approximately 0.4 g per half cycle due to the displacer volume. The operating frequency can be set between 0.2 and 1 Hz. The temperature is measured at the regenerator ends using standard platinum resistance temperature detectors (RTDs).

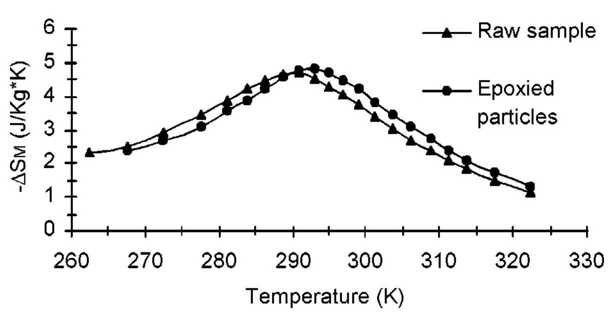

FIG. 3. Comparison of the MCE of Gd raw material and epoxied particles in terms of a isothermal change in entropy.

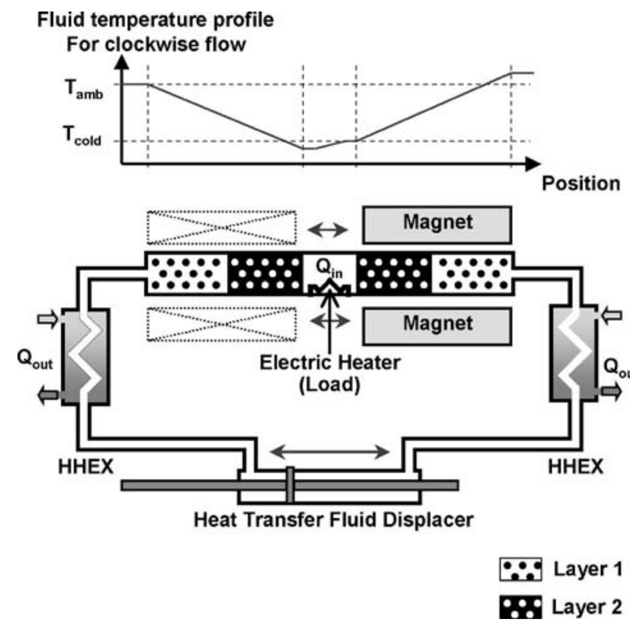

FIG. 4. Schematic of the active magnetic regenerator test apparatus.

RESULTS

For the tests, each regenerator was made up of two individual monolithic beds stacked together. Two single material regenerators composed of two 45 g Gd layers and then two multilayer regenerators, each composed of a 45 g Gd puck on the warm side and a 40 g $Gd_{0.74}Tb_{0.26}$ puck on the cold side, were tested. A similar range of operating parameters was used for both regenerators. Because the heat transfer fluid is helium and the cooling power is relatively low the system takes approximately 1.5 h or 3000–5000 cycles to reach steady state. In the following, performances are compared based on the temperature span achieved.

One aspect examined in the single material regenerator test was the impact of the ratio of the fluid thermal capacity to the refrigerant thermal capacity. This ratio is called utilization (Φ), defined as $\Phi = C_{p\_\mathrm{fluid}} m_{\mathrm{fluid}} / C_{\mathrm{ref}} m_{\mathrm{ref}}$. One way to vary Φ is to change the fluid density by varying the pressure. For a passive thermal regenerator, small utilization is preferred to obtain high thermal efficiency. However, in magnetic refrigeration, the refrigerant is also the working material and a significant amount of heat must be exchanged in order to produce refrigeration. Presently, there is no analytical relationship that quantitatively links utilization and performance. Figure 5 shows that, in the apparatus operating range, increasing the heat transfer fluid utilization positively impacts system performance. Therefore, to obtain peak performance with similar geometry and operating conditions, the utilization should be increased. This shows that, in the present experimental temperature range, helium may not be the optimal heat transfer fluid due to its low thermal mass.

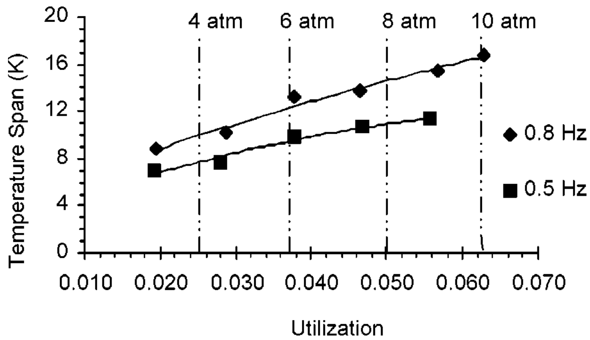

FIG. 5. Temperature span as a function of utilization for two 90 g Gd regenerators with no cooling load applied.

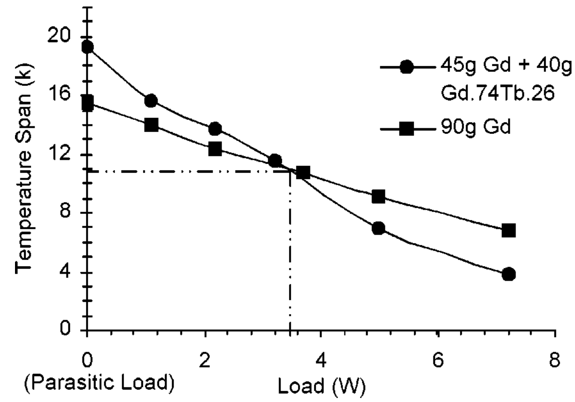

FIG. 7. Temperature span as a function of the cooling power for 0.8 Hz frequency and helium pressure of 10 atm for a multilayer and a single layer configuration.

Increasing the pressure or volume of gas displaced would increase the thermal capacity flux. Water could also be used in this temperature range.

Increasing the operating frequency, i.e., the number of thermodynamic cycles, increases the rate of work performed on the material, and thus the cooling power. In practice, high frequency allows less magnetic material and, therefore, overall apparatus size. The results presented in Fig. 6 show that frequency positively impacts the performance. However, it is expected that frictional and other parasitic loss will eventually limit the benefits of higher frequency.

One of the primary objectives of the experiments was to examine and prove the multilayer concept. Figure 7 shows a comparison of the performance of the two different active magnetic regenerators with similar mass, size, particles, and operating conditions. One can see a temperature span near 20 K ($T_{\mathrm{cold}} = 277\,\mathrm{K}$) obtained with two Gd/$Gd_{0.74}Tb_{0.26}$ multilayer regenerators. Also shown is the performance of equivalently sized regenerators made up of two layers of Gd with a maximum temperature span of about 16 K ($T_{\mathrm{cold}} = 281\,\mathrm{K}$).

Producing a larger temperature span implies that the AMR generates more cooling power. For instance, for a temperature span of 14 K, the multilayer regenerator is capable of approximately 2 W of cooling power compared to 1 W from the Gd-only regenerator. The multilayer AMR temperature span starts to exceed that of the single material when the cooling power is lower than 3.4 W which corresponds to a temperature span of 11 K and a cold temperature of 284 K. These results demonstrate that a multimaterial AMR can produce a larger temperature span and more cooling power than a single material AMR. These experiments also suggest that finding a rule as to how best use a second regenerator layer of another material is needed. More data and analysis are needed before this will be possible.

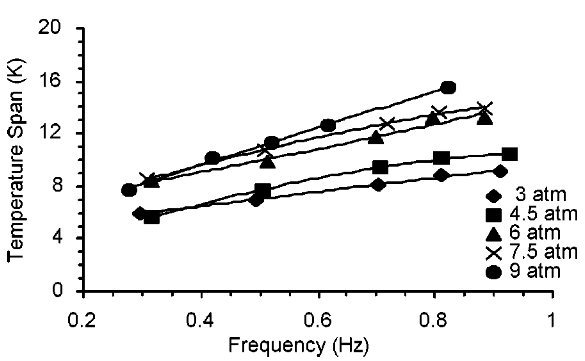

FIG. 6. Temperature span as a function of the frequency for two 90 g Gd regenerators with no cooling load applied.

CONCLUSIONS

Gd and $Gd_{0.74}Tb_{0.26}$ active magnetic regenerators were experimentally studied in an AMR cycle in the near-room temperature range. Increasing the fluid capacity ratio and the operating frequency positively impact the temperature span and cooling power within the current operating range of the device. The multimaterial regenerator was found to produce a higher temperature span and more cooling power than the single material regenerator under certain operating conditions. This experimental study opens the way to a better understanding of the multimaterial layered regenerator and could lead to the refinement of theoretical models. Being able to produce a larger temperature span and more cooling power using regenerators composed of more than one material is a major milestone in magnetic refrigerator development.

ACKNOWLEDGMENTS

The support of Natural Resources Canada and of the Ministère des Resources Naturelles du Québec is greatly appreciated. Special thanks to Professor John A. Barclay and Professor Tapan K. Bose for having initiated this work and to Yi Zhuo for isothermal entropy change measurements of Gd and $Gd_{0.74}Tb_{0.26}$ samples.

[1] J. A. Barclay, *Proceedings of 2nd Conference of Refrigeration for Cryogenic Sensor and Electronic Systems*, 1982.
[2] C. Zimm, A. Jastrab, A. Sternberg, V. Percharsky, K. Gschneigner, Jr., M. Osborne, and I. Anderson, Adv. Cryog. Eng. **43**, 1759 (1998).
[3] N. Hirano, S. Nagaya, M. Takahashi, T. Kuriyama, K. Ito, and S. Nomura, Adv. Cryog. Eng. **47**, 1027 (2002).
[4] C. Zimm, A. Sternberg, S. Russek, A. Boeder, and K. King, *Proceedings of IMECE'03 No. 42704*, Washington, D.C., 16–21 November, 2003.
[5] W. Wu, American Physical Society Conference, Austin, TX, 2003, abstract K7.004.
[6] P. Clot, D. Viallet, F. Allab, A. Kedous-Lebouc, J. M. Fournier, and J. P. Yonnet, IEEE Trans. Magn. **39**, 3349 (2003).
[7] N. Hirano, American Physical Society Conference, Austin, TX, 2003, ab-

stract K7.002.
[8] J. A. Barclay and W. A. Steyert, Jr., U.S. Patent No. 4,332,135 (filed June1982).
[9] S. R. Schuricht, A. J. DeGregoria, and C. B. Zimm, *Proceedings of the 7th International Cryocooler Conference* (Plenum, New York, 1993), p. 614.
[10] G. Green, J. Chafe, J. Stevens, and J. Humphrey, Adv. Cryog. Eng. **35**, 1165 (1990).
[11] G. Green, G. Patton, J. Stevens, and J. Humphrey, Rep. No. DTNSRDC-87/032, David Taylor Naval Ship Research and Development Center (1987).
[12] K. A. Gschneidner, Jr. and V. K. Pecharsky, Mater. Sci. Eng., A **287**, 301 (2000).
[13] S. Y. Dan'kov, A. M. Tishin, V. K. Pecharsky, and K. A. Gschneidner, Jr., Phys. Rev. B **57**, 3478 (1998).
[14] T. W. Wysokinski, X. Xu, and J. A. Barclay, Cryogenics **42**, 691 (2002).
[15] A. M. Rowe and J. A. Barclay, Adv. Cryog. Eng. **47**, 995 (2002).