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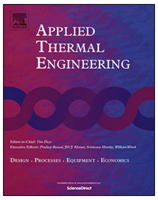

Research Paper

Optimization of the physical properties of magnetocaloric materials for solid state magnetic refrigeration

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HIGHLIGHTS

- The influence of the physical properties on solid magnetic refrigerators is studied.
- An increase of density and heat capacity widens the temperature span.
- The operating frequency increases with the thermal conductivity.
- The adiabatic change of temperature has profound implications on the refrigerator.
- The maximization of the temperature span leads to smaller operating frequencies.

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ABSTRACT

We have investigated numerically the dependence of the temperature span and optimum operating frequency of solid state magnetic refrigerators on the most important physical properties of magnetocaloric materials. We concluded that the temperature span depends inversely on the thermal conductivity of the magnetocaloric material and is proportional to the square root of its density, heat capacity and maximum adiabatic temperature variation upon the application/removal of a magnetic field (ΔT_{ad}^{m}). Also, and contrary to general belief, the increase of the thermal conductivity is not the only factor able to increase the optimum operating frequency. Reducing the density, heat capacity, ΔT_{ad}^{m} or broadening of the $\Delta T_{ad}(T)$ curve can result in larger operating frequencies. Our results show that a compromise between the maximum achievable temperature span and optimum operating frequency must be reached, as the increase of one reduces the other.

1. Introduction

Refrigeration is a key technology for our present welfare. Indeed, refrigeration made possible the conservation of fresh food for long periods of time and to tolerate the extremely high temperatures faced in certain regions of the globe. Up to now, the vapor compression technology has been essentially the only marketed refrigeration system. Although vapor compression technology has become the perfect paradigm of a technological monopoly, it has significant disadvantages, such as the low efficiency or the use of gases that substantially increase the refrigerator size and has nocive effects on the environment [1]. Such drawbacks can be overcome by other cooling mechanisms. Generally speaking, two alternative technologies have been rising for the last two decade. One of them is thermoelectricity [2,3]. This technology has a large cooling power but its efficiency is too low to compete with vapor compression refrigerators [4]. Efforts have been made to find better material properties to increase the overall performance. In that respect, a dimensionless figure of merit ZT was introduced for thermoelectrics [2]:

$$ZT = \frac{\lambda S^2 T}{k}, \tag{1}$$

where λ is the electric conductivity, S the Seeback coefficient, T the temperature and k the thermal conductivity. Systems having a ZT far above 1 might affect the dominance of vapor compression technology [5]. The other refrigeration technology that can overcome

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the issues of vapor compression refrigerators is magnetic refrigeration (MR) [6,7]. This technology relies on the magnetocaloric effect and, in contrast to thermoelectrics, shows extremely high efficiencies, which can reach 30%–60% of the Carnot cycle [1,6,8]. Efforts have been made to find cheap magnetocaloric materials (MCMs) that can be used in room temperature MR applications [9]. $Gd_5Si_xGe_{1-x}$ [10,11] and $La(Fe_{1-x}Si_x)_{13}$ [12] are systems where the Curie temperature can be easily shifted by changing the stoichiometry of the compound [13–15]. However, good combinations of physical properties of MCMs for MR have still to be worked out [16]. In that respect, it is surprising that, contrarily to the case of thermoelectrics, no figure of merit involving the proper functioning of magnetic refrigerators was so far proposed to guide the search for high performance MCMs for MR applications, in particular for solid state magnetic refrigerators.

To find a clear route for the synthesis of MCMs for solid state magnetic refrigeration, we have numerically investigated the influence of their most important physical parameters, namely the thermal conductivity k, heat capacity C_p, maximum adiabatic temperature variation upon the application or removal of a magnetic field ΔT_{ad}^{m} and broadening σ of the $\Delta T_{ad}(T)$ curve, on the maximum achievable temperature span and optimum operating frequency of solid state active magnetic regenerative refrigerators (AMRR). Our results can be easily expanded to conventional magnetic refrigerators.

2. Numerical model

We have here chosen to simulate a cooling system similar to the solid state refrigerators used in Ref. [17]. These systems can be considered as ideal AMRR systems as cascades of nearly perfect Brayton cycles are used. The only two differences that distinguish ideal systems from this group of refrigerators are the following. First, solid state refrigerators operate at a finite frequency, while the period of an ideal Brayton cycle is infinite. Second, although the hot end is a reservoir at temperature T_0, the cold end is a thermally insulated element and not a reservoir as in the ideal Brayton cycle. These two differences are of paramount importance for the present investigation since real refrigerators must take into account the time it takes to reach the stationary state (influence of a non-zero frequency) and the determination of the temperature span can only be achieved if at least one of the two ends is thermally insulated. In these solid state systems, the pumping of the fluid is replaced by a switching of thermal contact between the MCM and both cold and hot ends [18]. However, one should note that the results obtained for solid state magnetic refrigerators can be generalized to include fluidic heat exchange systems, as occurring in conventional magnetic refrigeration. Despite possible changes in the operating frequency we do not expect large discrepancies on the most suited MCM physical properties when fluidic heat exchange is considered. Moreover, such substitution is useful as possible numerical errors coming from the numerical methods for simulating heat convection are totally avoided [19].

The first stage of the refrigeration process starts with the cascade demagnetization of the MCM, from the hot end to the cold end. This approach results on the active magnetic regenerative mechanism. At this stage, the magnetocaloric material is completely insulated. The second stage occurs after the removal of the magnetic field. In this stage, the MCM (at a low temperature) is in contact with the cold end. A small contact time avoids the unwanted temperature increase of the MCM by the cold end (cf. Ref [17].). The final stage begins with the application of a magnetic field to the whole MCM. The MCM is now in thermal contact with the hot end, while the thermal contact with the cold end is removed.

3. Simulation

The numerical simulation was performed considering a one-dimensional finite difference model. Only heat conduction was taken into account [20]:

$$\frac{dT}{dt} - \frac{k}{\rho C_p}\frac{d^2T}{dx^2} = 0, \tag{2}$$

where t is the time, x the position, ρ the density and C_p the specific heat. Although Eq. (2) has a simple analytical solution, the present system can only be solved numerically, since it has a finite frequency, where the temperature changes according to ΔT_{ad} at the beginning of each cycle. Moreover, the contact of the MCM with the cold and hot ends changes in each cycle. The discretization, boundary conditions, and the simulation of the magnetocaloric effect are described in Refs. [17,21].

Overall, the discretization of the temperature in terms of space and time was performed according to the following formula:

$$T_{t,x} = T_{t-\Delta t,x} + \frac{\Delta t}{\alpha \Delta x^2}\left[T_{t-\Delta t,x-\Delta x} - 2T_{t-\Delta t,x} + T_{t-\Delta t,x+\Delta x}\right], \tag{3}$$

where $\alpha = \frac{\rho C_p}{k}$ is the characteristic temperature variation time, Δt is the time step and Δx is the distance between two consecutive points. The final temperature that we used for the analysis was the temperature of the middle of the cold material when the system attains the stationary state. The 1D simulated system is constituted by a MCM with 10 cm and a Cu cold material with 2 cm. The used number of points was 120 (100 for the MCM and 20 for the cold material). The temperature variation, due to the magnetocaloric effect, occurs instantaneously between the H-application and removal:

$$T_{n\frac{1}{v}+\Delta t,x} = T_{n\frac{1}{v},x} \pm \Delta T_{ad}^{a/r}\left(T_{n\frac{1}{v},x}\right), \tag{4}$$

where v is the operating frequency of the refrigerator, n is the cycle number, and $+\Delta T_{ad}^{a}$ and $-\Delta T_{ad}^{r}$ represent the adiabatic temperature variation for H-application and H-removal, respectively. Insulation was taken into account by considering the space derivative at the boundary to be zero:

$$\left(\frac{\partial T}{\partial x}\right)_{x=x_i} = 0. \tag{5}$$

In this work, the cold end is thermally insulated and the hot end has a fixed temperature of $T = 297$ K. $T = 297$ K was also the initial temperature of the system.

Our model was validated using three different criteria. First, we certified that the net heat exchange between the MCM and the cold end approaches zero near the stationary state. Second, we confirmed that the solution does not depend on the initial temperature of the MCM, but only on the working temperature (temperature at the hot end). Finally, we have verified that doubling the number of discretization points of the system leads to negligible deviations from the results.

One has now to define how to measure the performance of magnetic refrigerators. In that respect, we know that the cooling power and efficiency are important parameters. However, the cooling power will, generally speaking, determine the time it takes to reach the stationary state. Moreover, the inherent high efficiency is not the limiting parameter when compared to thermoelectrics or vapor compression refrigerators. On the other hand, the temperature span determines if the refrigerator is competitive [22]. We have hence used the temperature span as the key value for the performance of present magnetic refrigerator systems. The temperature span was