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INITIAL RESULTS OF A TEST-BED MAGNETIC REFRIGERATION MACHINE WITH PRACTICAL RUNNING CONDITIONS

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ABSTRACT

For a direct characterization of magnetocaloric materials, the measurements are generally made in adiabatic conditions. In such conditions, the initial temperature of the material is stabilized and then the magnetic field is changed from 0 T to a given finite value. However, in the majority of magnetic refrigeration systems, magnetocaloric materials are magnetized and demagnetized in non adiabatic conditions. Furthermore, almost nobody pays attention to the measurement of ΔT in actual conditions encountered in magnetic systems. At present a test-bed modelled and designed in cooperation with Cooltech Applications, to characterize magnetocaloric materials in practical conditions, was realized in the Institute of the Thermal Sciences and Engineering (IGT, Yverdon-les-Bains, Switzerland). This system allows the measurement of temperature differences and forces acting on magnetocaloric materials as a function of temperature and frequency, during magnetization-demagnetisation cycles, with a magnetic source having an induction field of about 2 Tesla. In this article initial results of forces and temperature differences obtained with some magnetocaloric materials are reported.

1. INTRODUCTION

The impact of synthetic refrigerants on the environment as well as the legal safety obligations drive the refrigeration industry to seek for new ways for completely phasing out greenhouse gases or for decreasing their charge in numerous installations. In order to be free from synthetic refrigerants, industries are continuously searching for environmentally friendly and suitable new technologies that will enable high energy savings, therefore reducing indirect CO₂ emissions. During the last fifteen years, both, namely the load reduction of the refrigerants in the installations and the use of natural, non-flammable, environmentally friendly refrigerants have been the preferred options by many end users such as Nestlé, Unilever, Coca Cola, etc.

Research on future refrigeration technologies orients itself on the indirect cooling technology as e.g. Phase Change Slurry (PCS), CO₂ vapour compressor technology, thermo-electric refrigeration, thermo-acoustic refrigeration and magnetic refrigeration (MR). Since 1997, when Pecharsky and Gschneidner discovered a giant magnetocaloric effect in Gd₅(Ge_{1-x}Si_x)₄ [1], intensive investigations have been concentrated on magnetic cooling at room temperature. Based on the magnetocaloric effect (MCE), magnetic cooling is considered in recent years as a serious alternative to replace the common gas-compression/expansion method due to high efficiency and environmental concerns [2]. An important MCE is considered to be the most important requirement for such an application. It is therefore strongly desirable to find new materials with a large MCE, in particular at induction fields close to 2 T and close to room temperature. Since then, there have been a large number of studies on systems, as well as magnetic materials, in particular to find out their suitability as potential refrigerants [3-7]. In addition, the measurement methods, as well as apparatuses for the characterization of magnetocaloric properties, are important research topics [8-10]. In order to realize magnetic cooling, it is important for magnetic materials to exhibit a large entropy change ΔS . However, the calculation of this property, generally based on the Maxwell's relation and isothermal magnetization data, has been the subject of controversy and long discussions [11-14]. This quantity alone does not fully characte-

size the effectiveness of a material for a refrigeration application. Another important parameter is the adiabatic temperature change ΔT_{ad} .

For the measurement of a magnetocaloric material's temperature change, different methods have been proposed in the literature [8]. ΔT can be determined directly or indirectly. For a direct determination of ΔT , the material is subjected to a magnetic field change and its temperature variation is measured generally by a system of thermocouples. For an indirect determination of ΔT (calorimetric method), the adiabatic temperature change is determined by measuring the specific heat as a function of temperature at various magnetic field strengths. However, based on adiabatic magnetization measurements Levitin *et al.* [15] proposed another method to determine ΔT_{ad} . This technique compares the magnetization field dependence performed under isothermal and adiabatic conditions. Due to the magnetocaloric effect (temperature increase), the adiabatic magnetization curve intersects the magnetization isotherms. The intersection point is then used to determine the field dependence of a sample's temperature when adiabatically magnetized, and, as a consequence, $\Delta T_{ad}(T, H)$. Various measurement methods can be found in reference [8].

In almost all described methods the measurements are performed in adiabatic conditions. Practically, the majority of the magnetic refrigeration systems operate in non adiabatic conditions. Moreover, for a magnetic cooling machine, the coefficient of performance (COP) or the efficiency is defined by the ratio of the produced cooling energy and the consumed energy during a magnetization-demagnetization cycle. In such a system, the magnetic field is created without electricity consumption when the magnetic source is a permanent magnet. Only mechanical energy is required to move the magnetocaloric material in and out of the magnetic field. This energy depends on the force and the torque induced by the interaction between the active material and the magnet. These quantities are a function of the material's magneto-thermal properties and the magnetic field. So, in addition to the magnetocaloric properties, the applied forces on the magnetocaloric materials in a magnetic refrigerator can influence strongly the efficiency of the latter.

In this paper, we present a simple apparatus for the characterization of magnetocaloric materials close to room temperature under practical operation conditions (distant or close to the adiabatic condition, 2 T...etc). The obtained results with different materials indicate that the developed setup is suitable for quick measurements of the magnetocaloric effect and determination of the transition temperature (Curie temperature T_C). Besides this, the magnetic forces acting on the material can also be measured.

2. EXISTING MAGNETIC COOLING SYSTEMS

By the introduction of international regulations two axes of research were strengthened. The first one focusses on the development of new alloys and the second is related to system's developments. After the year 2000 several demonstration systems were developed (see e.g. proceedings of the 2nd International Conference of the IIF/IIR on Magnetic Refrigeration at Room Temperature, April 2007, Slovenia) in the USA, China, France, Canada, Switzerland, etc. A list of magnetic refrigeration prototypes is given in table 1.

3. EXPERIMENTAL TEST FACILITY

The design and the building of the presented setup were motivated by the following criteria:

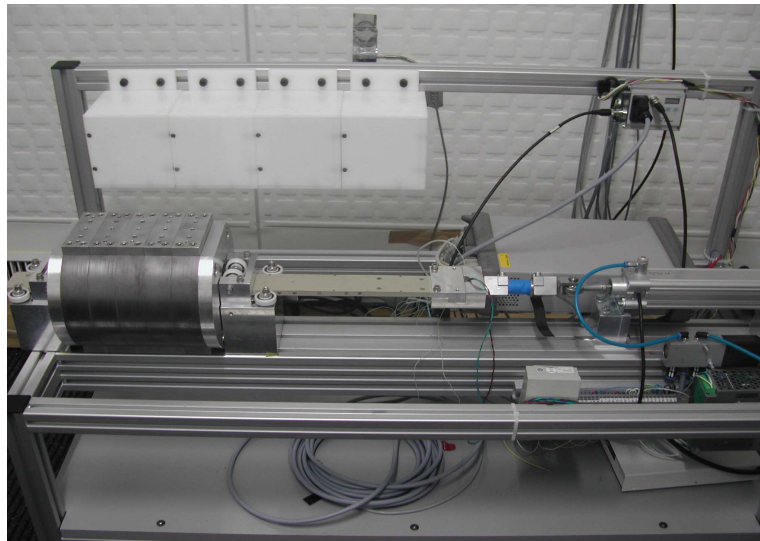
1. Simultaneous measurements of the magnetocaloric effect as well as of the transition temperature of different magnetocaloric materials.
2. Evaluation of the forces acting on the materials and the study of different phenomena related to their internal state, like hysteresis and demagnetization effect, etc.
3. Properties determined close to and at room temperature.
4. The measurements must be performed in real conditions that are encountered in a magnetic refrigeration system in a practical application.

Table 1. Existing magnetic refrigerators are listed in the table beneath (based on information distributed at the 2nd International Conference of the IIF/IIR on Magnetic Refrigeration at Room Temperature, 11-13 April 2007, Portoroz, Slovenia).

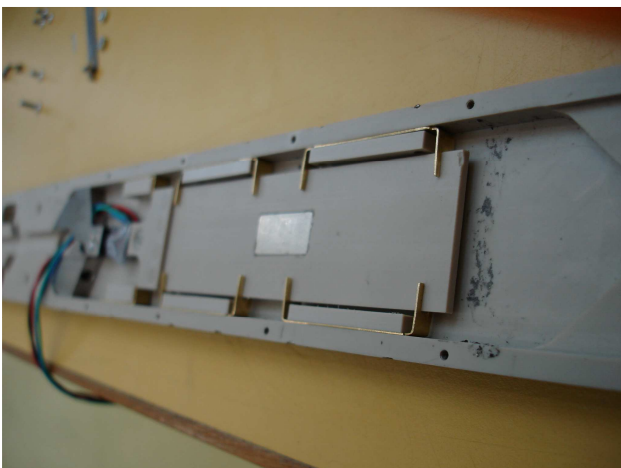
Institute Country	Type Year	Max cooling power (W)	Max ΔT (K)	Magnetic Field (T)	Regenerator
Ames Laboratory Astronautics Corp., USA	Reciprocating 1997	600	10	Superconducting magnet: 5 T	Tested with Gd spheres
Barcelona Spain	Rotary 2000		5	Permanent magnet: 0.95 T	Tested with Gd foil
Chubu Electric Japan	Reciprocating 2000	100	21	Superconducting magnet: 4 T	Tested with Gd spheres
University of Victoria, Canada	Reciprocating 2001	2	14	Superconducting magnet: 2 T	Tested with Gd and ...
Astronautics Corporation, USA	Rotary 2001	50	25	Permanent magnet: 1.5 T	Tested with Gd spheres, layered bed
Sichuan Inst China	Reciprocating 2002	40	23	Permanent magnet: 1.4 T	Tested with Gd powder and $Gd_5(Si_{1.985}Ge_{1.985}Ga_{0.03})$ powder
Chubu Electric Japan	Reciprocating	40	27	Permanent magnet: 0.6 T	
Laboratoire Electrotechnique Grenoble France	Reciprocating	4	4	Permanent magnet: 0.8 T	Tested with Gd foil
Cooltech France	Rotary	95	16	Permanent magnet: 1 T	Tested with Gd foil
Georges Washington University, USA	Reciprocating 2005		5	Permanent magnet: 2 T	Tested with Gd foil
University of Victoria, Canada	Reciprocating 2005	15	50	Superconducting magnet: 2 T	
University of Applied Sciences of Western Switzerland	Rotary 2006	100	3	Permanent magnet: 0.8 T	Tested with Gd spheres
University of Applied Sciences of Western Switzerland	Reciprocating 2007	100	7	Permanent magnet: 2 T	Tested with Gd foil

The test facility is shown in figure 1. In this apparatus the sample is placed and fixed in a ‘material holder’. The ‘material holder’ is moved in and out of the magnetic field by a pneumatic linear piston. The apparatus runs automatically controlled by a Personal Computer. The temperature, the temperature change and the force acting on the material under experimental investigation are observed online. The frequency of the periodic motion and the ambient temperature can also be controlled. A determination of the temperature of the sample is performed, if the ambient temperature is in the range of 278 K to 310 K. It is also possible to

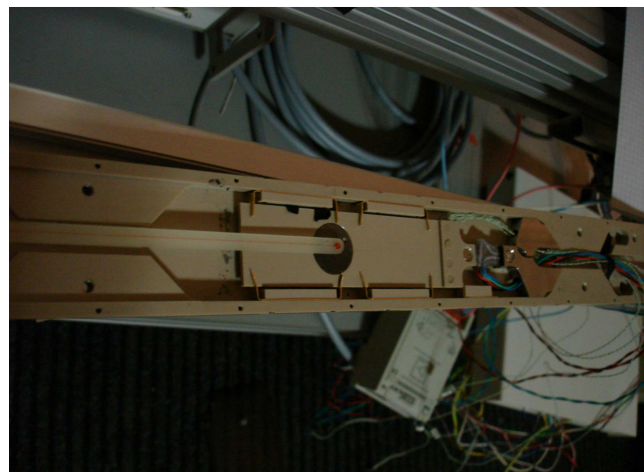
directly heat or cool the ‘material holder’ with locally applied pulsed air, allowing now an extended temperature range of 274 K to 330 K. The temperature distribution is evaluated by a thermocouple probe with a response time lower than 10 ms, and the magnetic field induced by the magnetic circuit is determined by a probe based on the Hall Effect.



a) The test facility.



b) Small permanent magnet (2x10x20 mm).



c) Disc of gadolinium (20 mm diameter and 2 mm thickness) with Hall probe.

Figure 1: Test facility with a magnetic field source of approximately 2 Tesla.

4. RESULTS AND DISCUSSION

For magnetic cooling requirements, several types of magnetic flux sources were reported; Lee *et al.* [16] and Vasile *et al.* [17]. For the here described test bed, neodymium iron boron (NdFeB) permanent magnets, which have a highest remanence of ~ 1.45 T, were chosen to build the magnetic source. It is shown in figure 1. This latter was adopted to generate a magnetic field of approximately 2 Tesla (in the empty gap). The field source’s air gap has a cross section of 10 mm x 50 mm and a length of 200 mm. Figure 2 presents the magnetic field measured in the centre of the gap along its length with a Hall probe. As shown in figure 2, the magnetic field is relatively homogeneous inside the magnetic source and presents a maximal value in its centre of about 2 Tesla.

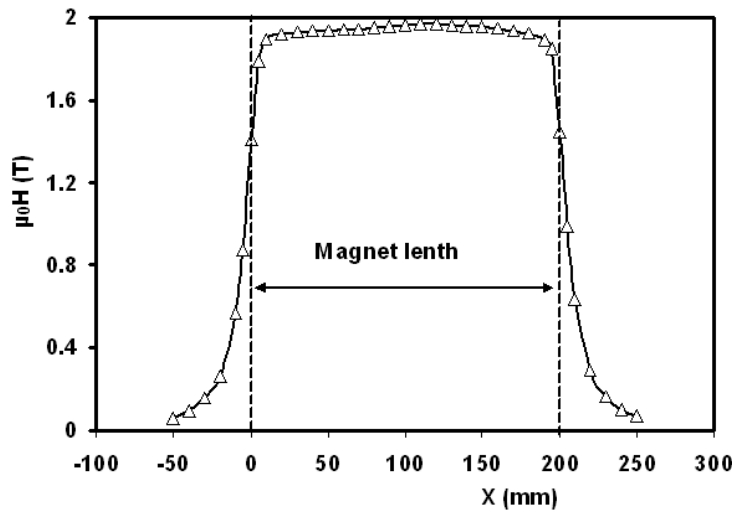


Figure 2: Magnetic field distribution along the axis of the magnet.

Using the test facility, we have performed direct measurements of the temperature change as function of temperature for pure Gd and the compound $\text{LaFe}_{11}\text{Co}_{0.9}\text{Si}_{1.1}$. The pure Gd is serving as a comparative reference. In order to compare the results obtained by using the here presented test bed and those made in adiabatic conditions, we have performed the measurement of ΔT_{ad} for $\text{LaFe}_{11}\text{Co}_{0.9}\text{Si}_{1.1}$ in the Baotou Research Institute of Rare Earths in China. Besides, it is worth noting that the initial temperature of the sample (plotted in figures 3-4) corresponds to the temperature just before the jump caused by the magnetization or demagnetization.

For Gd (figure 3 a), the normalised temperature change measured in the test-bed during magnetization at Curie temperature is found to be about 2 K/T instead of 2.7 and 2.9 K/T obtained respectively in Gd by Huang *et al.* [10] and Gschneidner *et al.* [18] in adiabatic conditions. This difference can be attributed essentially to the demagnetisation effect due to the shape of our sample. We note that our measurements were performed on a sheet of gadolinium with a thickness of 0.3 mm. So, in this case, the application of a perpendicular magnetic field to the surface of the sheet induces a demagnetisation field, cancelling out a part of the applied field. This reduces the magnetic field inside the magnetocaloric material and as consequence decreases the magnetization and the magnetocaloric effect. The demagnetization effect explains also the large deviation observed in figure 3b between obtained experimental data and theoretical calculations of the MCE based on the mean field theory (MFT). In their recent paper Bahl and Nielsen [19] reported on the effect of demagnetisation on the magnetocaloric properties of gadolinium. The adiabatic temperature change of different gadolinium sheets upon application of a magnetic field was determined for various values of applied field and sample orientation. The authors have observed a significant dependence of the MCE on the sample orientation. Furthermore, taking into account the demagnetisation factor, our measurements of the temperature change are in agreement with that obtained for an adiabatic process. The observed deviation between magnetization and demagnetization curves can be explained by the hysteresis effect. It is true that, the equilibrium conditions, heat leaks, the rate of the magnetic field change as well as the Eddy current can influence the magnitude of MCE when the material is magnetized or demagnetized. However, according to different measurements performed for several materials, it seems that the observed difference between figure 3-a curves can be attributed essentially to the irreversibility of MCE, as a consequence of the hysteresis. Looking at figure 3-a, the deviation between MCE curves during magnetization and demagnetization process is large in ferromagnetic state (lower temperatures), while this difference is smaller in paramagnetic region. This, is justified by the fact that in paramagnetic state, the magnetization of gadolinium evolves almost linearly (slightly) with the magnetic field which makes the transformation more reversible leading as consequence to a reversible MCE. The same behavior can be seen clearly in $\text{LaFe}_{11}\text{Co}_{0.9}\text{Si}_{1.1}$ [20]. This problem as well as the demagnetization effect will be treated in detail in a forthcoming communication.

The measured temperature changes for $\text{LaFe}_{11}\text{Co}_{0.9}\text{Si}_{1.1}$ compounds performed in adiabatic and non adiabatic conditions are given in figure 4. For $\text{LaFe}_{11}\text{Co}_{0.9}\text{Si}_{1.1}$ the obtained maximum ΔT (at Curie temperature) is

about 2.15 K, for a magnetic source with an induction field of 1.96 T, which is equivalent to the variation of the temperature change with a rate of about 1 K/T (normalised ΔT). For a magnetic field varying from 0 to 1.48 T, the maximal adiabatic temperature change is about 1.5 K (~ 1 K/T) which is in good agreement with that obtained by using our developed apparatus. However, from the curve in figure 4, the Curie temperature corresponding to the maximum point (test bed measurements) is found to be 293 K for $\text{LaFe}_{11}\text{Co}_{0.9}\text{Si}_{1.1}$. This value agrees closely with that obtained according to thermomagnetic curves collected at lower magnetic fields where $T_C=294$ K [21].

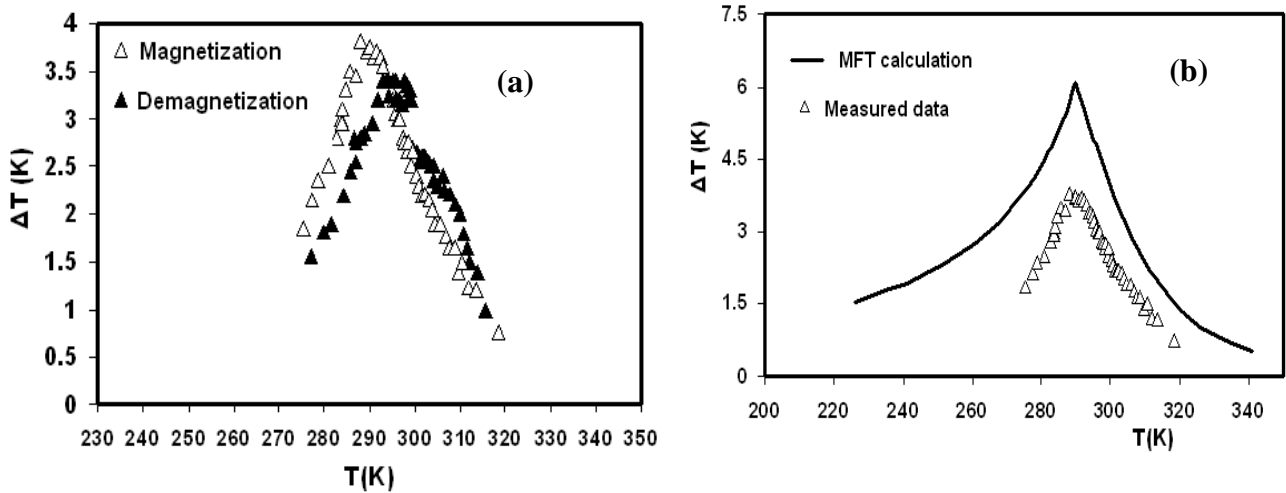


Figure 3: Temperature changes performed in the test bed for pure Gd under non-adiabatic conditions with 1.96 T in a magnetization and a demagnetization process (a), and a comparison between the temperature change obtained (here only during magnetization) using the test bed and that calculated based on the mean field theory (MFT) at the same magnetic field of 1.96 T (b).

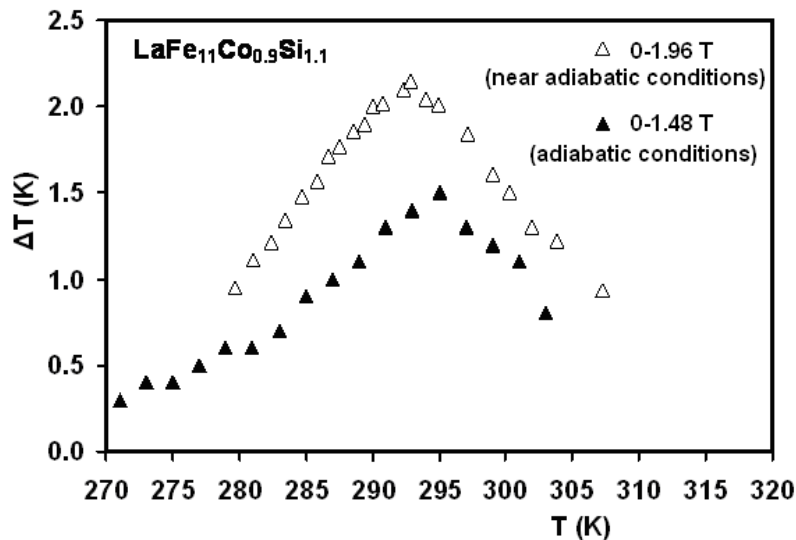


Figure 4: Temperature change performed in the test bed for $\text{LaFe}_{11}\text{Co}_{0.9}\text{Si}_{1.1}$, if a magnetic field change of 1.96 T is applied. For comparison ΔT_{ad} measured in the Baotou Research Institute of Rare Earths in China for 1.48 T is also shown.

The coefficient of performance of a magnetic refrigerator is defined as follows:

$$COP = \frac{Q_C}{W_{tot}}, \quad (1)$$

where Q_C is the absorbed heat from the cold source and W_{tot} is the total work input. W_{tot} is given by:

$$W_{tot} = W_{mag} + W_{pump}, \quad (2)$$

where W_{pump} denotes the fluid pumping work and W_{mag} is the energy absorbed by the motor that drives the refrigerant material in and out the magnetic field region. The magnetic work is given by:

$$W_{mag} = \int_{cycle} \vec{F} d\vec{l}, \quad (3)$$

where \vec{F} is the magnetic force and $d\vec{l}$ represents the displacement of the MCM bed along the thermodynamic cycle. Equations 1 to 3 show the importance of magnetic forces and their relation on the efficiency. Notify the fact that the magnetic work constitutes a large part of the full absorbed energy by the cooling system. Furthermore, the evaluation of these forces is of great interest. A sensor was inserted in the here described test bed to measure the magnetic forces acting on different magnetocaloric materials as a function of temperature, frequency, etc.

Generally, the acting magnetic force applied by a magnetic field source B on a volume dv of magnetocaloric material with magnetization M can be expressed by:

$$\vec{F}(T, B) = \int_v (\vec{M} \cdot \text{grad}) \vec{B} dv. \quad (4)$$

This equation shows that a magnetic force is created when the material is moved in an inhomogeneous magnetic field. Figure 5 shows the observed acting force and magnetic field for measurements performed at room temperature on a small permanent magnet of size 2 x 10 x 20 mm during a magnetization and demagnetization process. The observed magnetic field is measured by a Hall probe placed on the measured material's (magnet's) surface, which explains its high value compared to that of the magnetic source (~1.96 T). Looking at figure 5-a, the applied force to the tested material appears in the region closest to the entrance of the magnetic source, where a field directional derivative exists. Furthermore, when the material is inside the magnetic source, the magnetic force becomes zero due to the uniformity of the magnetic field, which is seen in figure 2.

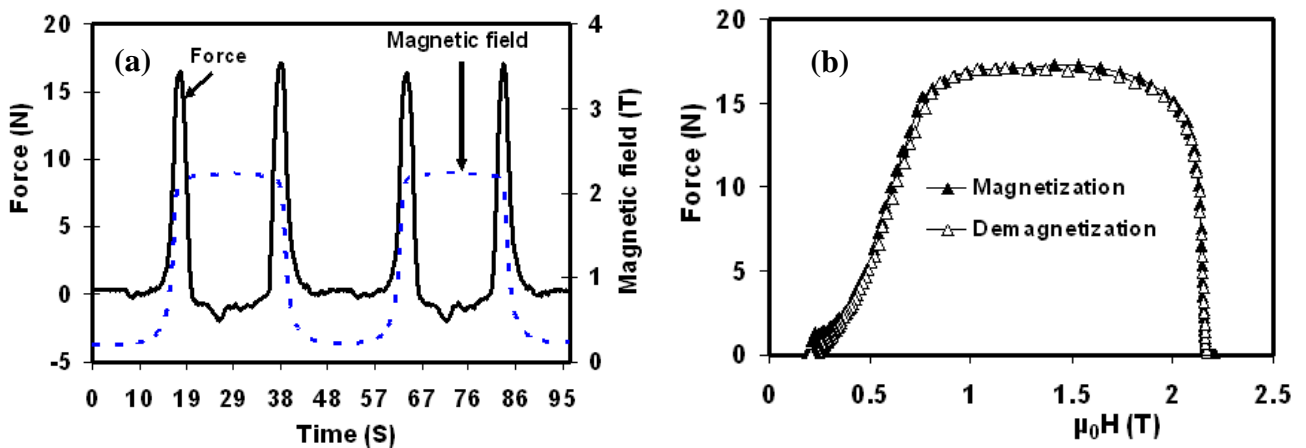


Figure 5: Force and magnetic field time dependence of a magnet (2 x 10 x 20 mm) during the magnetization and demagnetization process (a). Force versus the magnetic field of the magnet is given in (b).

Figure 6 presents the magnetic field dependence of the applied force on a disc of gadolinium (20 mm diameter and 2 mm thickness). For Gd a significant deviation between the curves corresponding to magnetization and demagnetization is observed. On the other hand, for the magnet this phenomenon is absent (see figure 5 b). The observed difference is attributed essentially to the magnetocaloric effect exhibited by Gd under an applied field of 1.96 T, which modifies the thermal and the magnetization profile. However, in the case of Gd the maximal value of the applied force compares well ($\sim 7\%$) with that calculated using a finite element method by applying the software Flux3D.

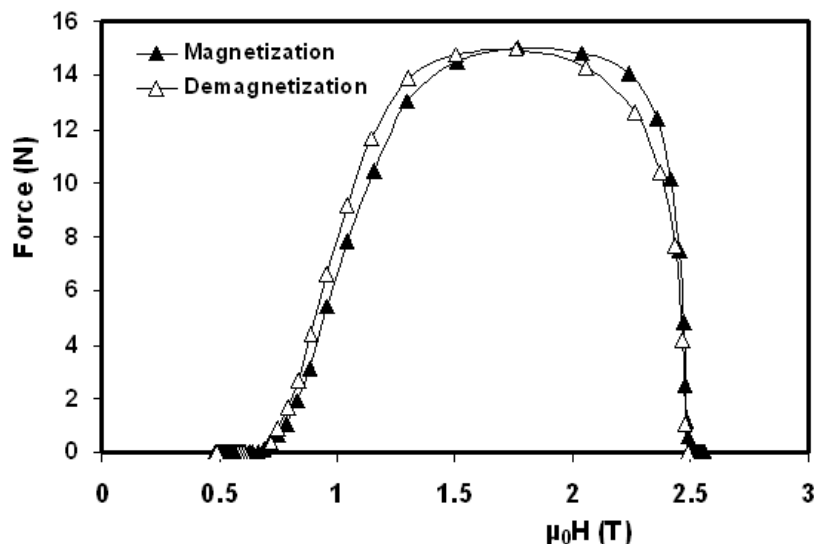


Figure 6: Acting magnetic force on a disc of Gd (20 mm diameter and 2 mm thickness) during a magnetization and demagnetisation process.

5. CONCLUSION AND OUTLOOK

A simple apparatus for the characterization of magnetocaloric materials close to or at room temperature in practical conditions was developed. With this a large number of samples can be tested simultaneously. A magnetic field source based on NdFeB magnets was constructed to generate a large magnetic field of about 2 T. The results of preliminary measurements on some magnetocaloric materials have been presented to demonstrate the performances of the setup. The obtained temperature change for some materials like Gd and $\text{LaFe}_{11}\text{Co}_{0.9}\text{Si}_{1.1}$ correspond well with that performed in adiabatic conditions, showing that the developed device can be used for a fast characterisation of the Curie temperature and adiabatic temperature change of potential magnetocaloric materials close to or at ambient temperature. The acting magnetic forces resulting on the interaction between a magnetocaloric material and the magnetic source can be also determined. This allows us to select materials, so as to minimize forces and to increase the efficiency of potential magnetocaloric refrigeration systems.

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