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Operational test of bonded magnetocaloric plates

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ABSTRACT

Bonded plates made by hot pressing $La_{0.85}Ce_{0.15}Fe_{11.25}Mn_{0.25}Si_{1.5}H_y$ particles and resin have been tested as active magnetic regenerators in a small scale magnetocaloric device. Firstly, the plates were carefully characterised magnetically and thermally. The plates were prepared with 5 wt% resin, and from density measurements it was found that the volume ratio of the magnetocaloric material in the plates was 0.53, due to the resin and porosity. The best operating conditions for the plate regenerator were determined at which a temperature span of 6.4 K was measured along the plates.

Test opérationnel de plaques magnétocaloriques liées

Mots clés : Magnétocalorique ; Réfrigérateur magnétique ; Régénérateur ; Liant résineux

1. Introduction

For the past decade significant progress has been made in bringing magnetic refrigeration towards commercialisation. New magnetocaloric materials have been developed and magnetocaloric demonstration devices become ever more powerful and efficient (Kitanovski et al., 2015). Most of these devices have employed the so called active magnetic regenerator (AMR) cycle as a method of utilising the magnetocaloric effect in a device. Here the temperature of a porous regenerator structure is increased and decreased by applying and removing a

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Nomenclature

Abbreviations

AMR	active magnetic regenerator
DSC	differential scanning calorimeter
EDS	energy-dispersive X-ray spectroscopy
Gd	gadolinium
PPMS	physical properties measurement system
SEM	scanning electron microscope
TTO	thermal transport option
VSM	vibration sample magnetometer

Variables

c	specific heat
c_H	specific heat at constant magnetic field
m	mass
T_{hot}	hot end temperature
ΔS_M	magnetic isothermal entropy change
ΔT_{ad}	adiabatic temperature change
$\Delta T_{effective}$	effective adiabatic temperature change
ΔT_{span}	temperature span
ϕ	utilisation

Subscripts

f	fluid
MCM	magnetocaloric material
resin	resin binder material
s	solid

magnetic field, while a heat transfer fluid is reciprocated back and forth in order to build up a temperature gradient. Keys to utilising the AMR concept are firstly to have a magnetocaloric material, or a cascade of such, with high magnetocaloric effect, and secondly, to shape this material in a way that there is good thermal contact between solid and fluid, allowing an effective heat transfer as, e.g., discussed in Ferreira et al. (2014).

Many magnetocaloric materials have been studied and considered for application in devices (Smith et al., 2012). Recently a lot of research has been done on materials that undergo a first order magnetic phase transition, so-called first order materials, as they have a very high magnetic entropy change ΔS_M around the phase transition. An important example of this class of materials is the intermetallic $La(Fe,Si)_{13}H_y$, which has been studied with a number of different elements doped into the Fe and Si sites (Zhang et al., 2013). In general this material series has a high magnetocaloric effect, a tunable Curie temperature and only a small amount of undesirable thermal hysteresis. The disadvantage of these materials is the lack of structural stability due to the volume change at the phase transition (Lyubina et al., 2010).

Recently, different strategies for shaping these and similar magnetocaloric materials into the desired shapes have been proposed and tested. Polymer bonding and pressing to plates has been demonstrated for first order materials (Radulov et al., 2015; Skokov et al., 2014) and second order materials of the same type (Pulko et al., 2015). Other methods of producing structures have also been presented, such as injection moulding (Lanzarini et al., 2015), hot pressing with Cu (Liu et al., 2015) and extrusion into monoliths (Pryds et al., 2011). Actual AMR performance in a test device has only been reported for the second order bonded plates (Pulko et al., 2015). However, porous particle based regenerators where the particles are bonded for stability have been presented, most notably by Jacobs et al. (2014) using $La(Fe,Si)_{13}H_y$.

Here we demonstrate in a small versatile AMR device the performance of plates of first order materials bonded with resin and cured during pressing.

2. Experimental

A $La_{0.85}Ce_{0.15}Fe_{11.25}Mn_{0.25}Si_{1.5}$ ingot was prepared in an induction furnace. The purity of the raw materials was at least 99.9 wt%. The annealed ingots were crushed into particles less than 0.35 mm in size for the hydrogenation process. These were annealed in a high purity hydrogen atmosphere until saturation. The structure of the hydrides was confirmed by x-ray powder diffraction. The hydrogen concentration y of $La_{0.85}Ce_{0.15}Fe_{11.25}Mn_{0.25}Si_{1.5}H_y$ (referred to as LaFeSiH in the following) was estimated to be 0.2 wt% by the inert gas pulse infrared absorption method. When vacuum annealing under 350 °C for 2 hours a sharp decrease of the Curie temperature was observed due to the escape of H.

2.1. Processing the plates

The resulting powder was mixed with a phenolic resin system in a mass ratio of 20:1. The mixed powders were pressed into plate shape under pressure of 20 MPa and then solidified at 150 °C for 10 minutes, as shown in Fig. 1. The plates were prepared in the size 25 mm by 40 mm, in order to fit into the regenerator test device, with a thickness of 0.5 mm. Measuring the specific heat shows that the Curie temperature does not change during the resin curing process, indicating that the LaFeSiH is unaffected by this treatment.

2.2. Characterisation of the plates

Thermal conductivity was measured at 300 K using a Thermal Transport Option (TTO) on a Quantum Design physical property measurement system (PPMS). The density of the plates was measured using an AccuPyc 1340 helium pycnometer repeating each measurement 10 times, and the density of the resin was measured with a Pentapyc5200e Auto Density Analyzer. The magnetic properties were analysed in a LakeShore 7407 vibrating sample magnetometer (VSM). The sample was measured in the temperature range 270 K–306 K at applied fields up to 1.5 T. The specific heat of the sample was measured using a custom built differential scanning calorimeter (DSC), allowing the applied field to be varied in the range 0–1.5 T at a fixed orientation relative to the sample (Jeppesen et al., 2008). A scanning electron microscope (SEM), Hitachi TM3000, equipped with a Bruker Quantax Energy-dispersive X-ray spectroscopy detector (EDS) was used to analyse the microstructure of a piece of one of the plates.