Magnetism and magnetocaloric effect in LaFe11.9-xCoxSi1.1

Hansen, Britt Rosendahl; Kuhn, Luise Theil; Bahl, Christian; Ancona-Torres, Carlos Eugenio; Katter, Matthias

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Magnetism and magnetocaloric effect in $LaFe_{11.9-x}Co_xSi_{1.1}$

Britt Rosendahl Hansen [(a)], L. Theil Kuhn [(a)], C.R.H. Bahl [(a)], C. Ancona Torres [(a)], M. Katter [(b)].

[(a)] Fuel Cells and Solid State Chemistry Division, Risø DTU, National Laboratory for Sustainable Energy
[(B)] Vacuumschmelze GmbH & Co. KG

Technical University of Denmark
Risø DTU
National Laboratory for Sustainable Energy
MagCool project

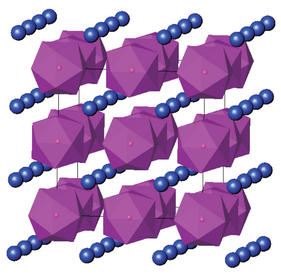
Seven samples of $LaFe_{11.9-x}Co_xSi_{1.1}$ were characterized. The materials are interesting for magnetic refrigeration at room temperature.

Samples:

No.	Composition	T_c (K)
1	$LaFe_{11.25}Co_{0.65}Si_{1.1}$	255.9
2	$LaFe_{11.14}Co_{0.76}S_{1.1}$	267.2
3	$LaFe_{11.05}Co_{0.85}Si_{1.1}$	282.1
4	$LaFe_{10.92}Co_{0.98}Si_{1.1}$	293.5
5	$LaFe_{10.77}Co_{1.13}Si_{1.1}$	311.6
6	$LaFe_{10.61}Co_{1.29}Si_{1.1}$	327.8
7	$LaFe_{10.45}Co_{1.45}Si_{1.1}$	347.3

Magnetism:
itinerant electron ferromagnetism
and localized moments on Fe and Co atoms

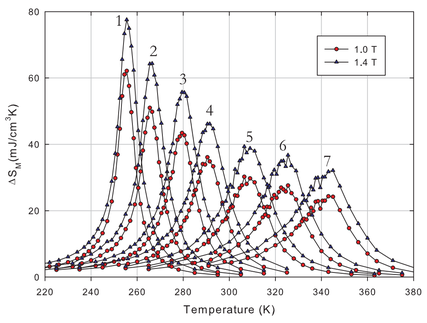
Crystal structure: cubic

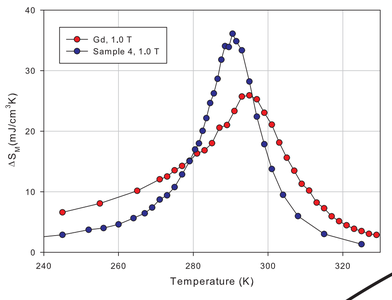


The 13 atoms of Fe, Co and Si form icosahedra (purple) around a central atom, while La sits interstitially (blue).

Magnetic entropy change, ΔS_M

When compared to the magnetocaloric material, Gd, $La(Fe,Co,Si)_{13}$ is seen to have a larger ΔS_M. The peak in ΔS_M is slimmer making the range of operation narrower, but this is remedied by the possibility for a composite material.



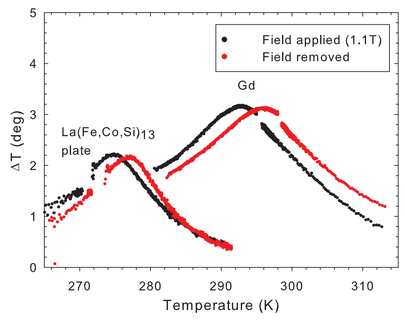


The magnetic entropy change is given by

$$\Delta S_M(T, H) = \int_{H_1}^{H_2} (\partial M/\partial T)_{H,P}\, dH$$

and calculated from a series of initialcurves.

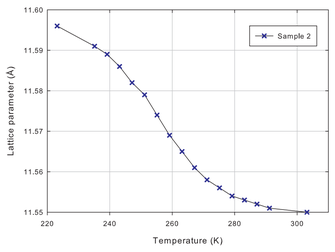
Adiabatic temperature change, ΔT_{ad}



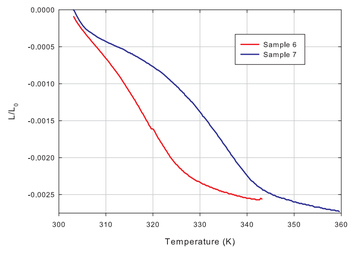
For an applied field of 1.1T a plate of $LaFe_{11.06}Co_{0.86}Si_{1.08}$ (not one of the seven samples) is seen to have a ΔT_{ad} that is less than that of the magnetocaloric material Gd.

The adiabatic temperature change was measured directly by a thermocouple attached to the magnetocaloric material.

Lattice expansion



X-ray diffraction at a range of temperatures and dilatometry show a lattice expansion on the order of 0.4 %.



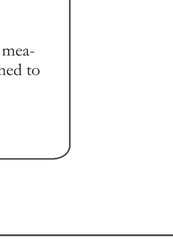
$La(Fe,Co,Si)_{13}$ - as a magnetic refrigerant

Good	Bad
Magnetic entropy change, ΔS_M	Adiabatic temperature change, Δt_{ad}
No significant lattice expansion	Brittleness
Lack of hysteresis	Corrosivity
Ease of production	
Can be shaped into plates	

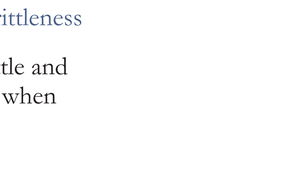
Brittleness

The material is very brittle and can be broken by hand, when made into plates.

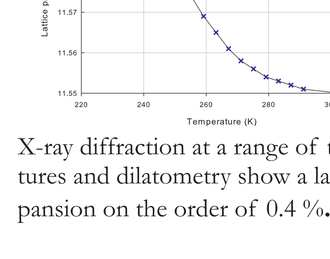
Corrosiveness

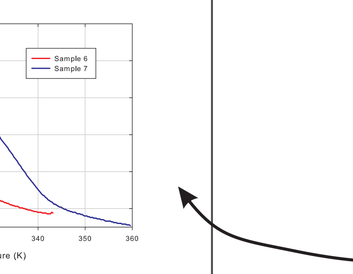


After 20 hours in demineralised water, the sample has oxidized. However, if the fluid is a mixture of anti-freeze and water, corrosion is greatly reduced.



Hysteresis

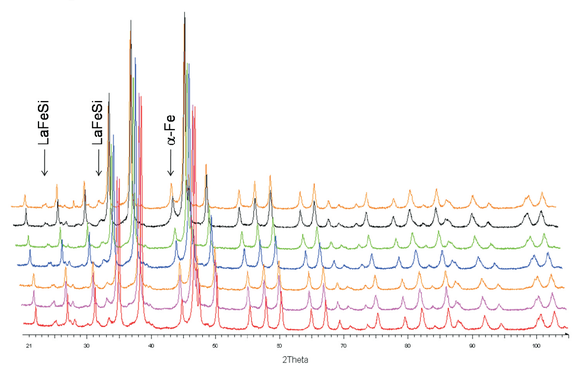




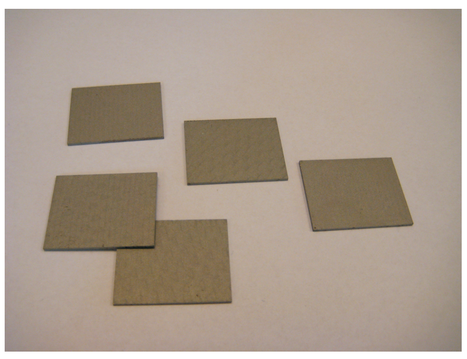
No magnetic or thermal hysteresis is observed in magnetization or calorimetry data. Presented are magnetization as a function of temperature for sample 1, hysteresis curves for sample 1 and heat capacity with and without an applied magnetic field for sample 2 and 4.

X-ray diffraction at room temperature

X-ray diffractometry confirms that the samples, which were made using powders that are pressed and then sintered (Katter *et al.*) for four to eight hours, have $La(Fe,Co,Si)_{13}$ as the main phase with small amounts of LaFeSi and α-Fe.



Plates



The plates were cut from blocks using wire EDM (Electrical Discharge Machining).

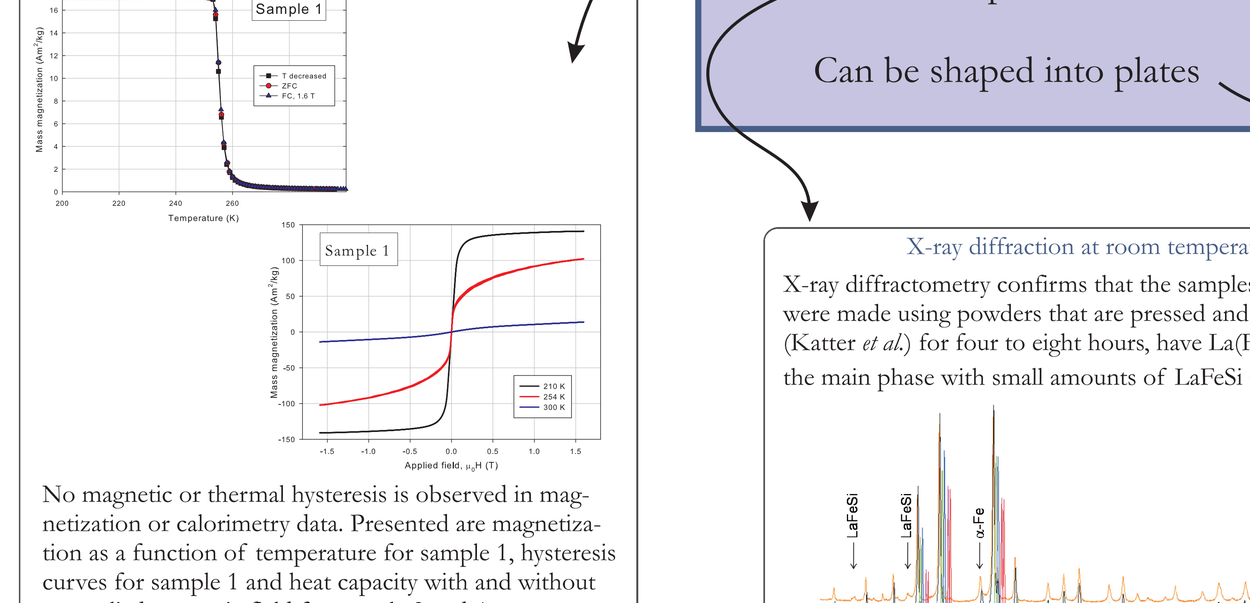
How to cool using magnets

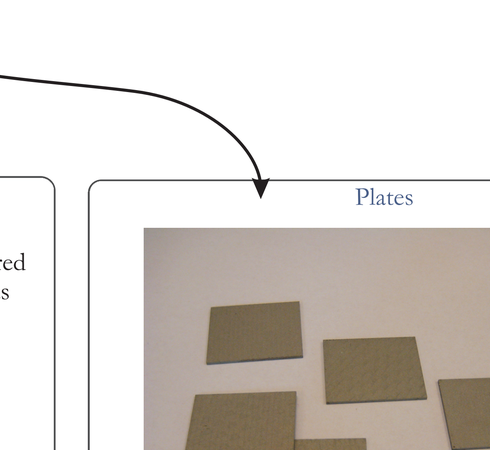
A magnetic field is applied adiabatically heating the plates and thus the water.

The cooling fluid is moved between the plates to move heat to the 'hot end' and expel it.

The magnetic field is removed adiabatically cooling the plates and thus the water.

The cooling fluid is moved over the plates and a 'cold end' is established.





Measurement of the temperature at the hot and cold end when using plates of the material $LaFe_{10.96}Co_{0.98}Si_{1.06}$, which has a transition temperature around 15°C. Without a heat load a temperature difference of 8°C is achieved.

What's happening with the magnetocaloric material:

- $T=T_0$
- The spins align with the field and the magnetic entropy decrease. The total entropy is preserved and therefore the lattice entropy increases. $T = T_0 + \Delta t_{ad}$
- Heat is removed, while the spins are still aligned with the field.
- The spins become disordered and the magnetic entropy increases, causing the lattice entropy to decrease. $T = T_0 - \Delta T_{ad}$

Conclusion

The materials $LaFe_{11.9-x}Co_xSi_{1.1}$ for $0.65<x<1.45$ show a 2nd order ferromagnetic-to-paramagnetic transition at temperatures between ~254 K and ~336 K.

The materials were studied as possible magnetic refrigerants for magnetic cooling at room temperature. For this purpose they are promising. The corrosiveness and brittleness should, however, be resolved.

What's next?

Further studies on the $La(Fe,Co,Si)_{13}$ series: Thermal conductivity measurements and a study of the microstructure.

References:
Katter *et al.*, IEEE Trans. Magn., 22 (2008) 3044