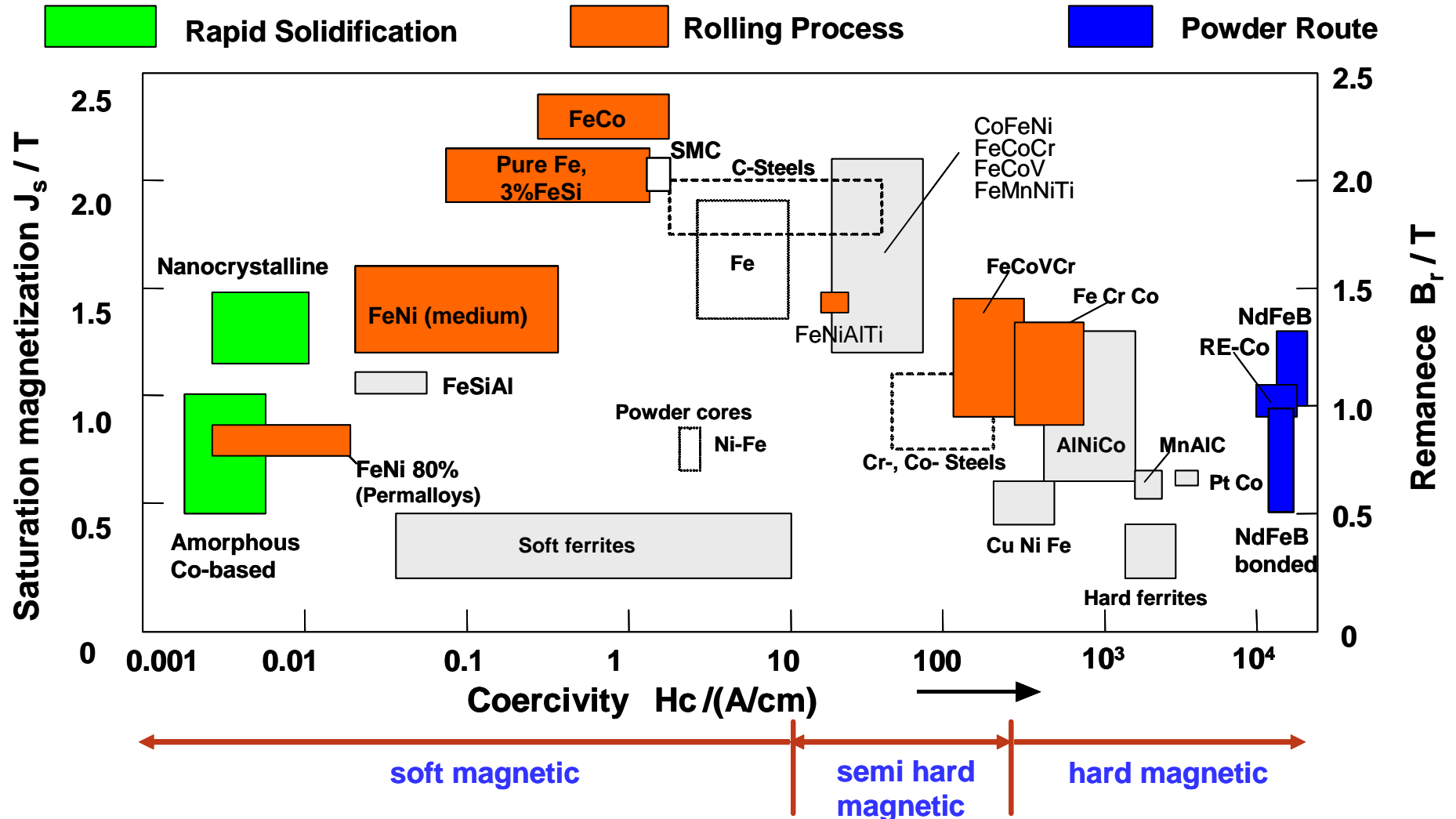


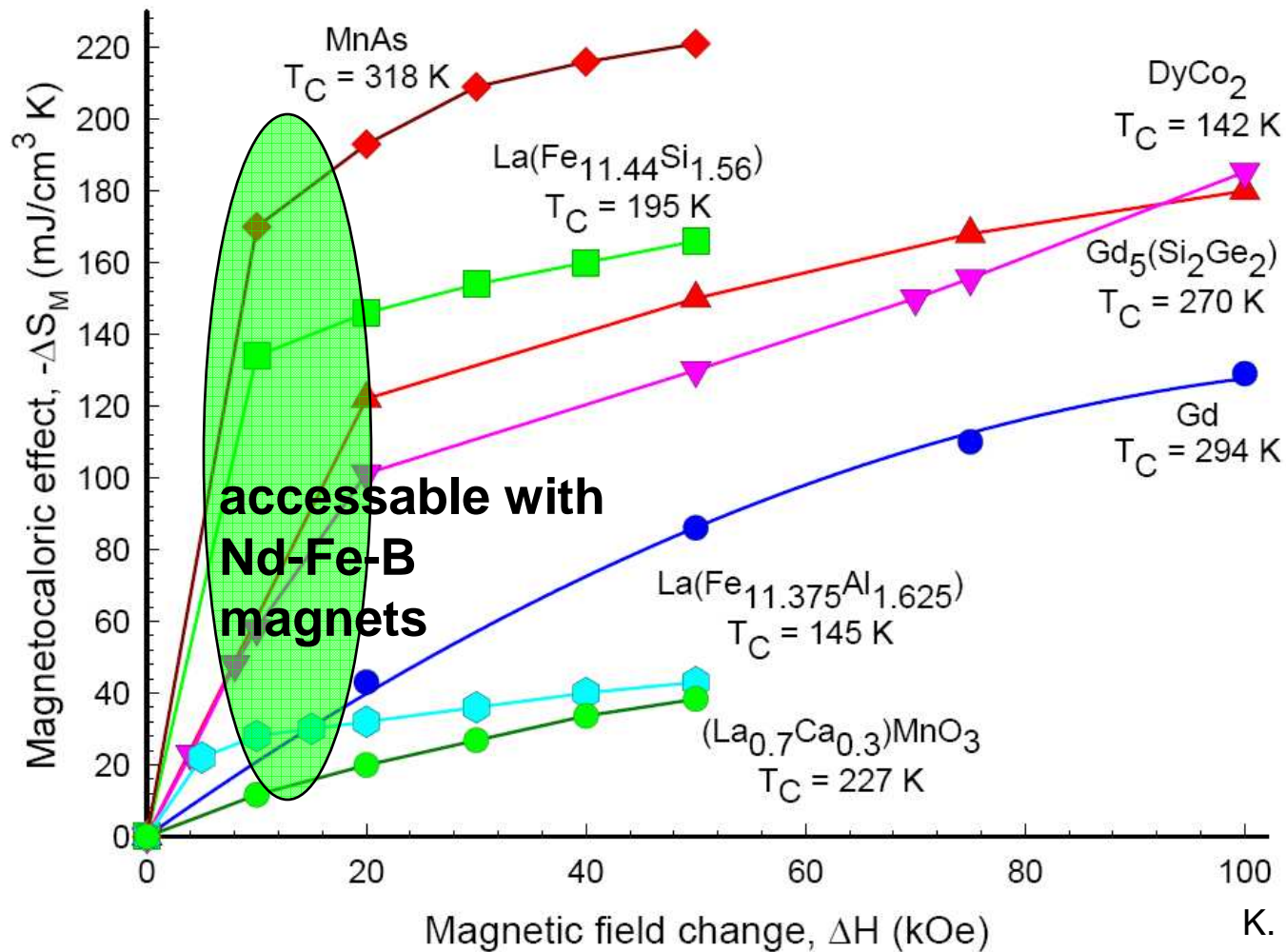
Magnetocaloric properties of reactively sintered $\text{La}(\text{Fe},\text{Co},\text{Si})_{13}$

M. Katter, V. Zellmann, G.W. Reppel, K. Uestuener
Vacuumschmelze GmbH & Co. KG
Delft Days on Magnetocalorics

overview magnetic materials

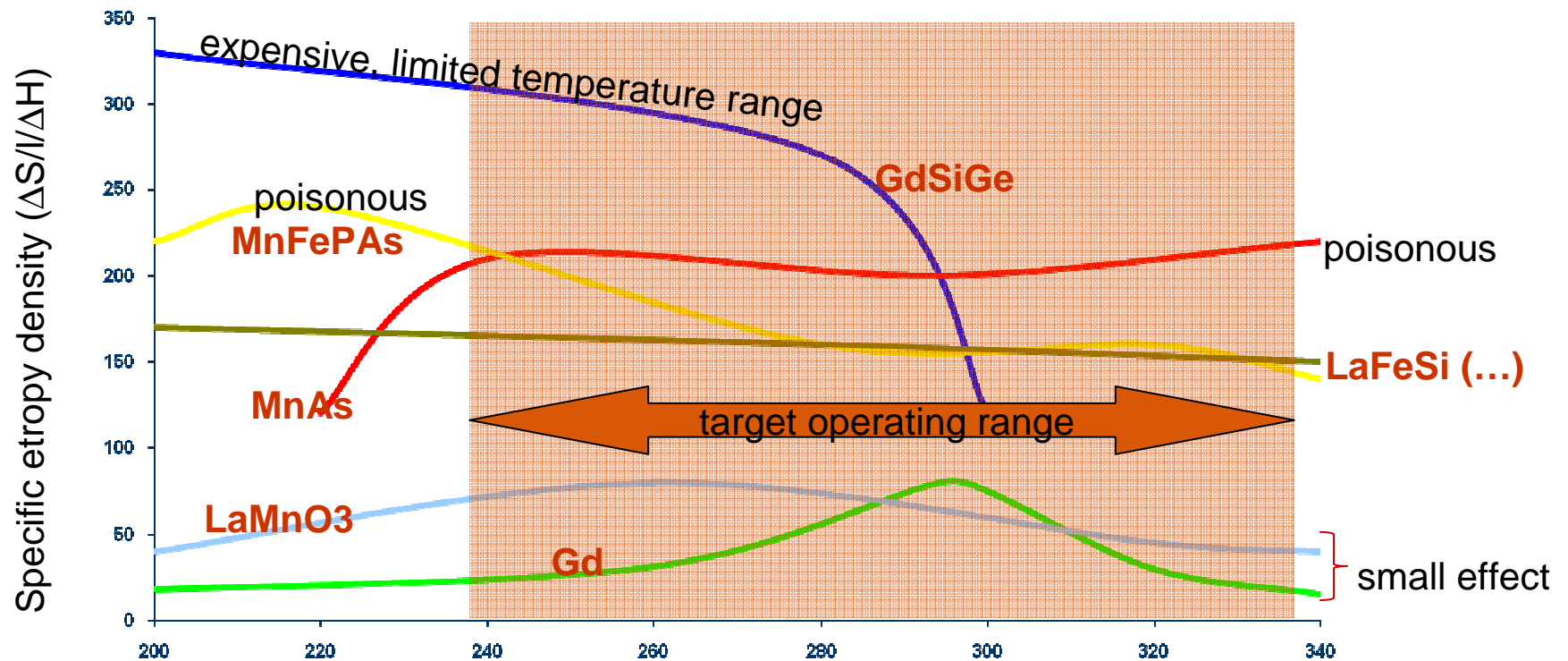


new magnetocaloric materials



K. Gsneidner (2007)

magnetocaloric materials



most promising at the moment: La-Fe-Si

Cooltech Applications (2008)

conventional preparation of $\text{La}(\text{Fe},\text{Si})_{13}$

- casting
- anneal, e.g. 1080°C/140 h
- optionally load with hydrogen

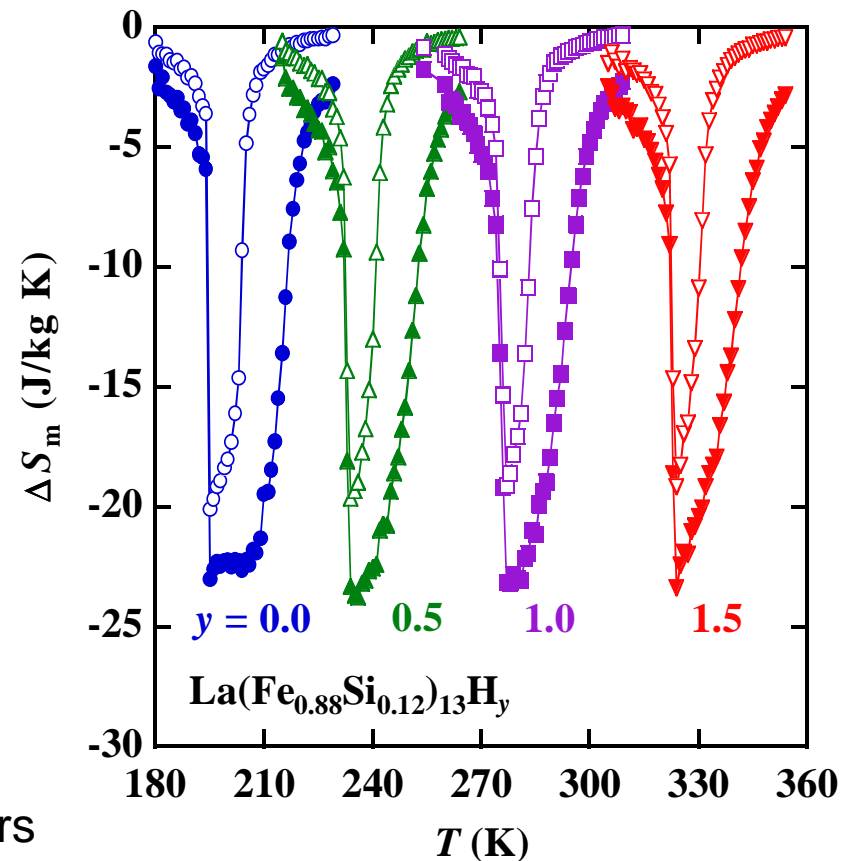
⇒ long annealing times,
 ⇒ with hydrogen only powder

S. Fujieda et al. 2005
 F.X. Hu et al. 2005 (with Co)

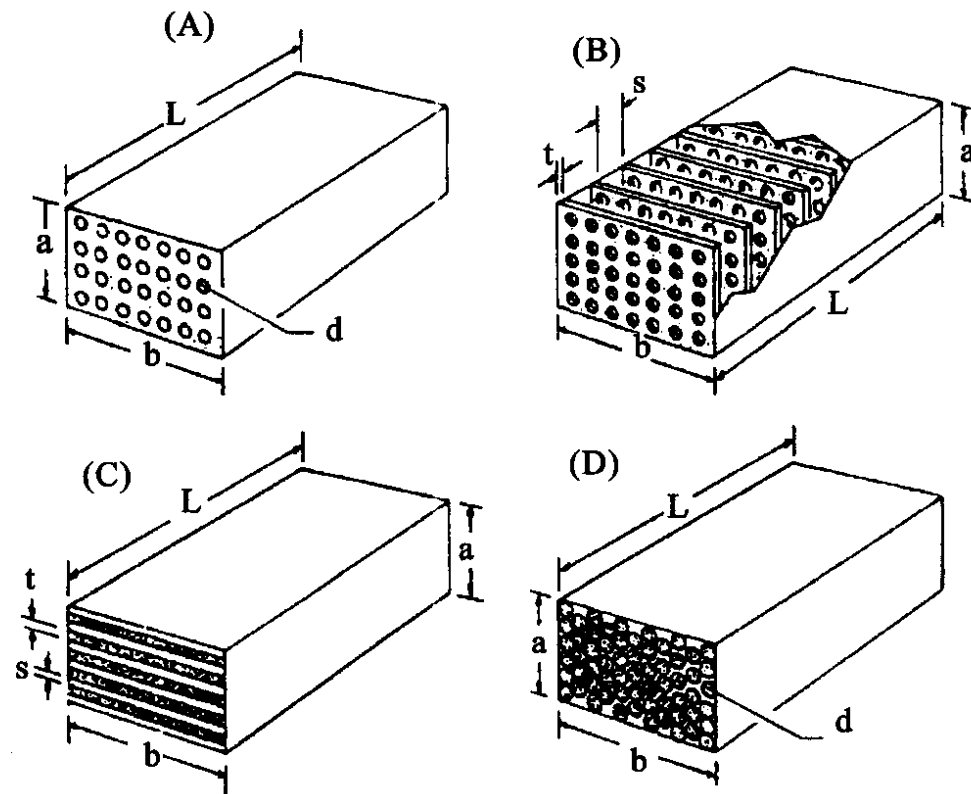
- melt spinning
- short anneal, e.g. 1000°C/2 h

⇒ only flakes or powder,
 no shaped parts for heat exchangers

O. Gutfleisch et al. 2005
 S. Hirosawa et al. 2006



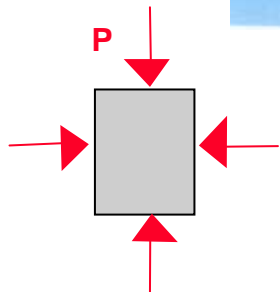
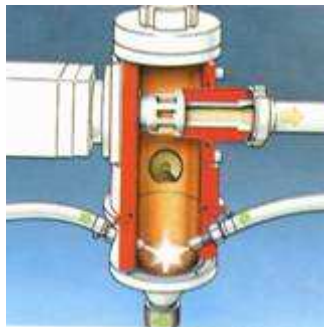
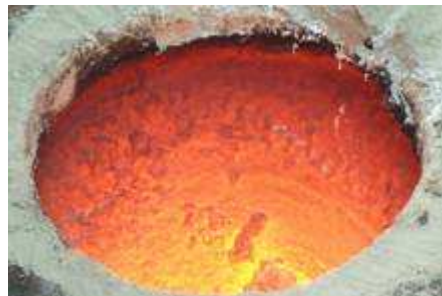
Designs for active magnetic regenerators



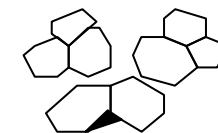
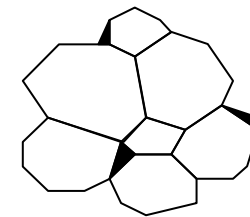
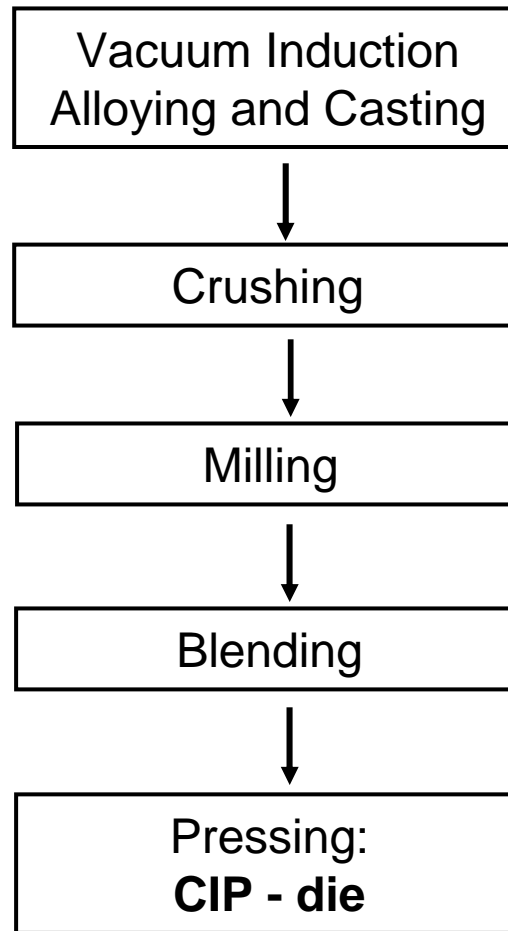
large pressure drops
in powder beds?

J.A. Barclay and S. Sarangi 1984 in A.M. Tishin and Y.I. Spichkin 2003

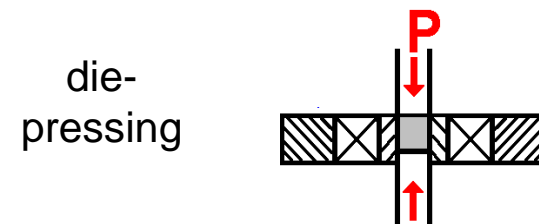
Powder Metallurgical (PM) Processing of La-Fe-Si



isostatic
pressing

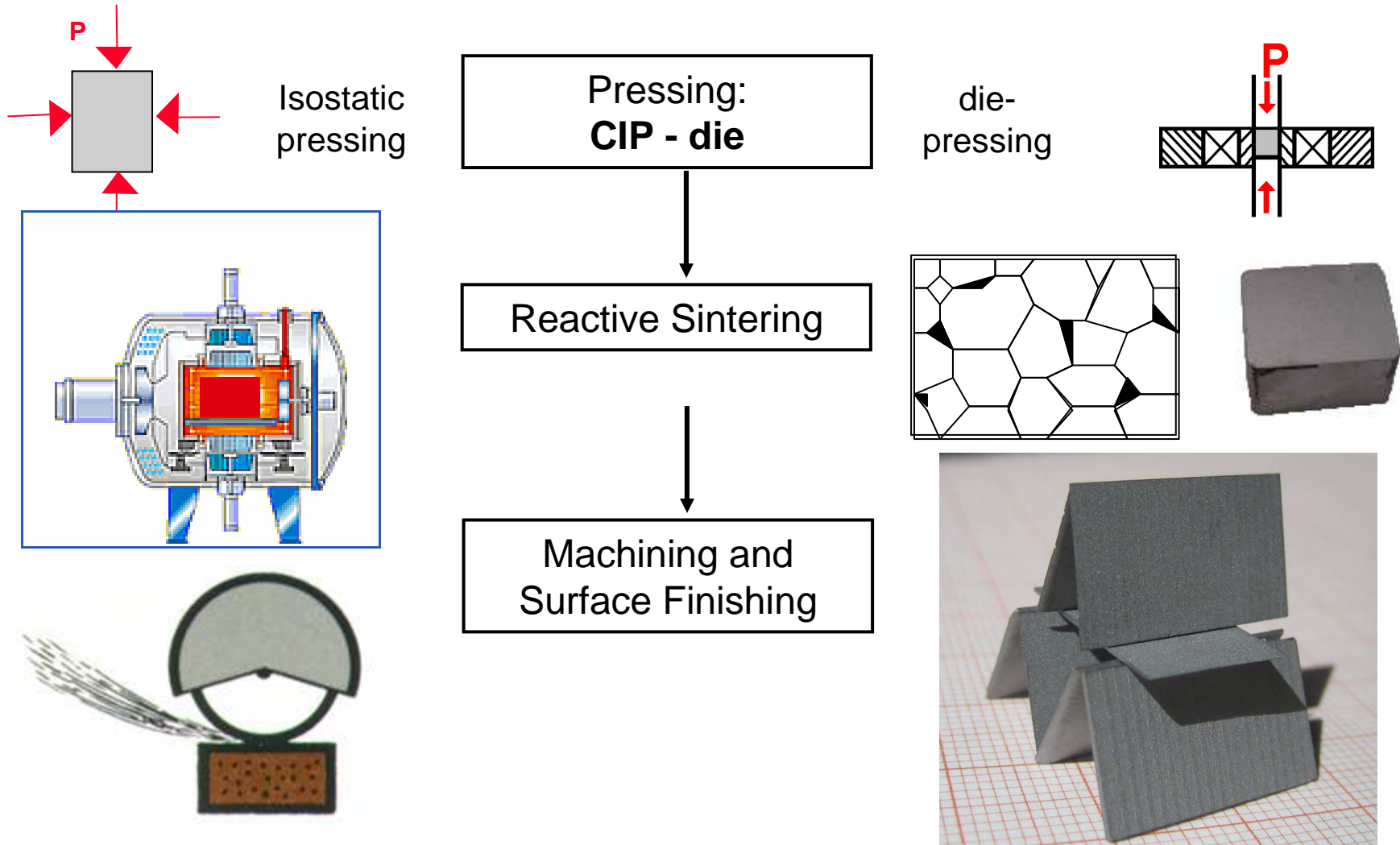


elemental powders



die-
pressing

Powder Metallurgical (PM) Processing of La-Fe-Si





powder blend of
Fe, Si and LaH_x
(no melting at all)

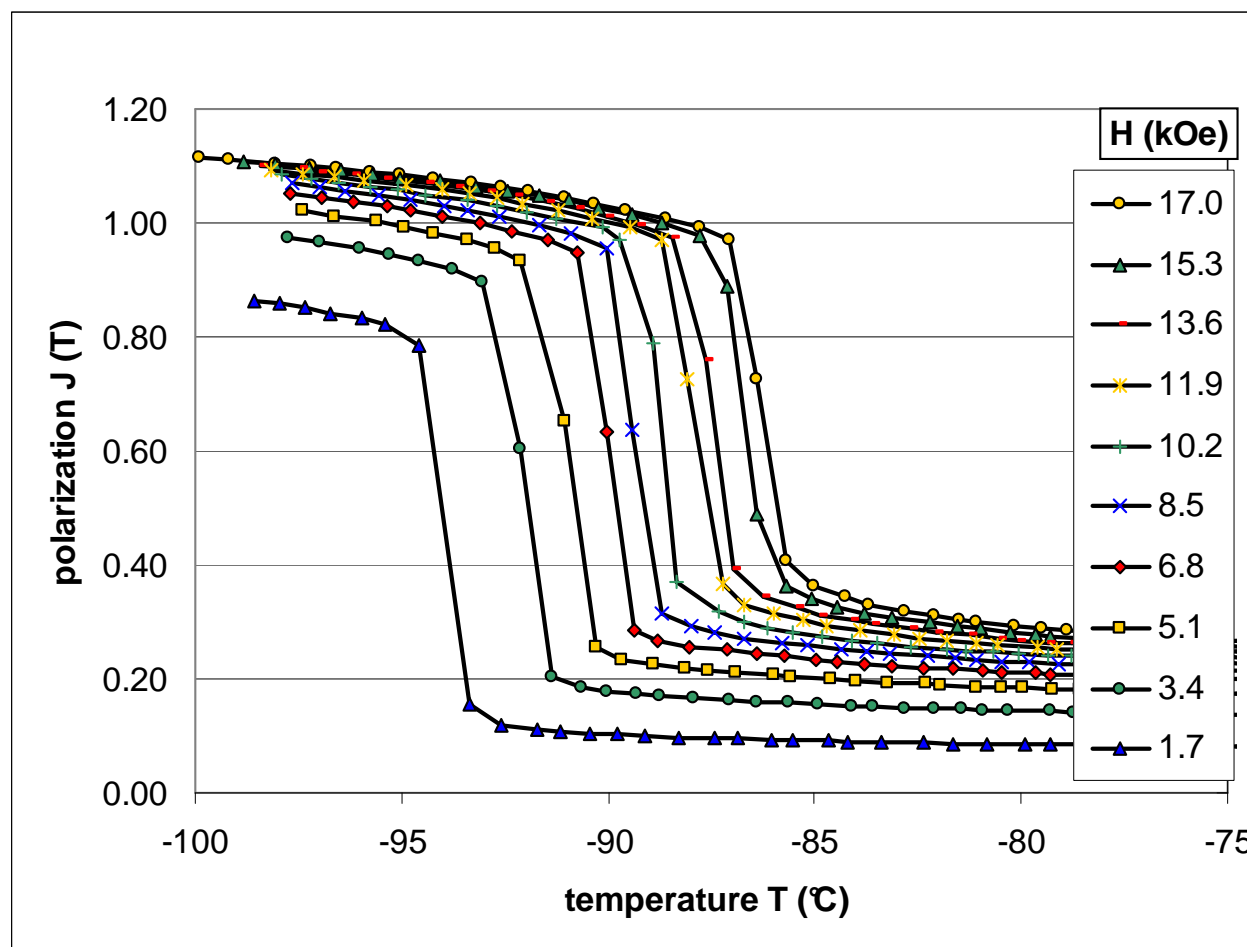
milled to
F.S.S.S. < 5 μm

isostatically
pressed

sintered
at 1120°C/4h

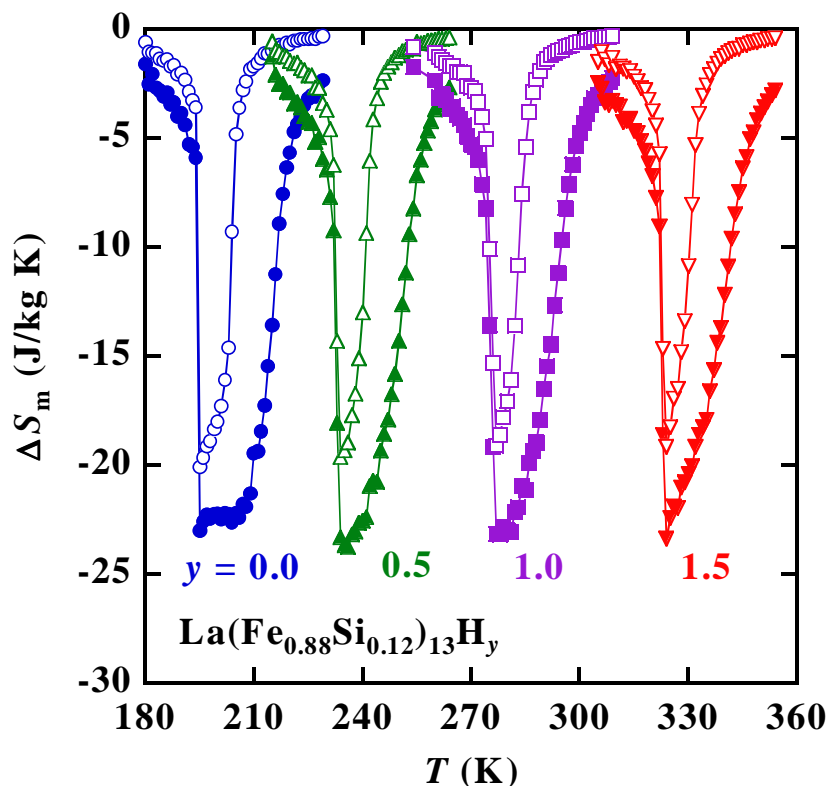
density = 6.6 g/cm³

sample: 4,2 x 2 x 1,2 mm

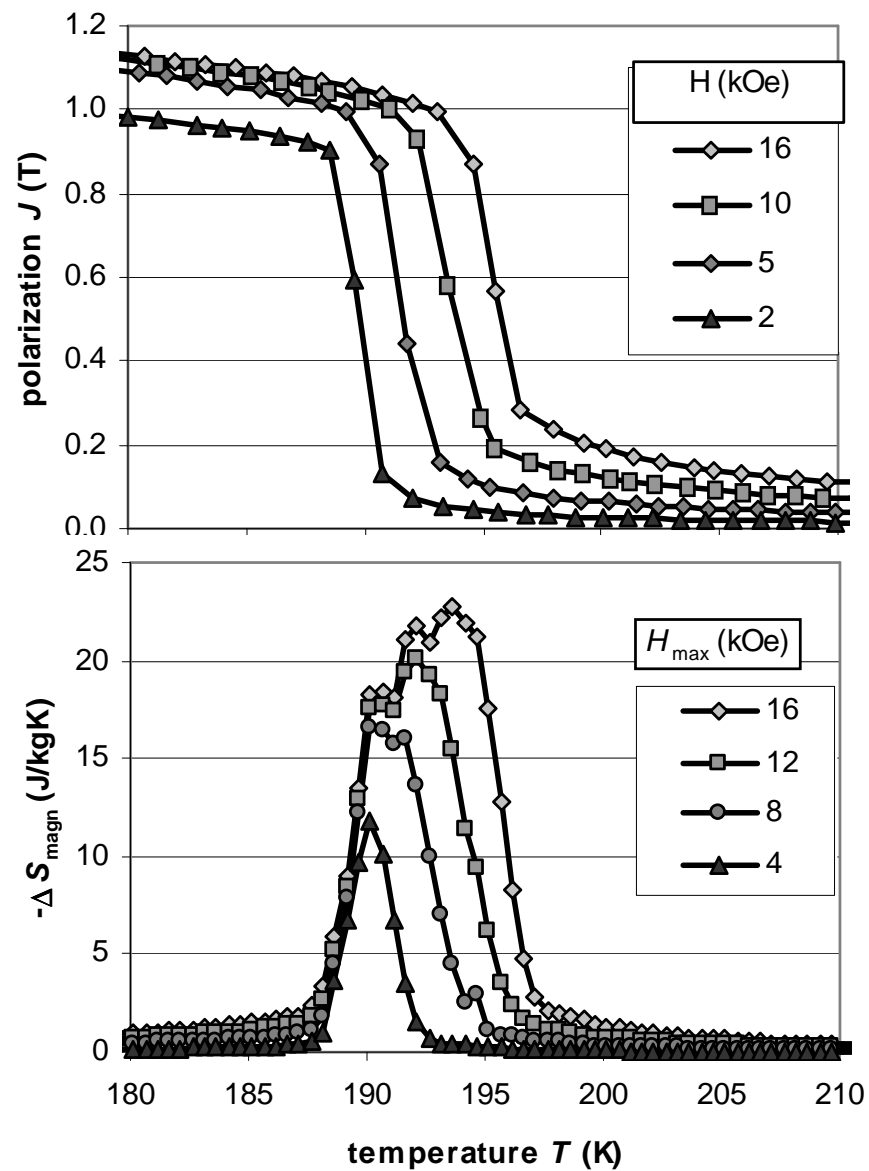


La(Fe_{0.893}Si_{0.107})₁₃, sintered

$$\Delta S_T = \int_0^H \left(\frac{\partial M(T, H)}{\partial T} \right)_{P, H} dH$$

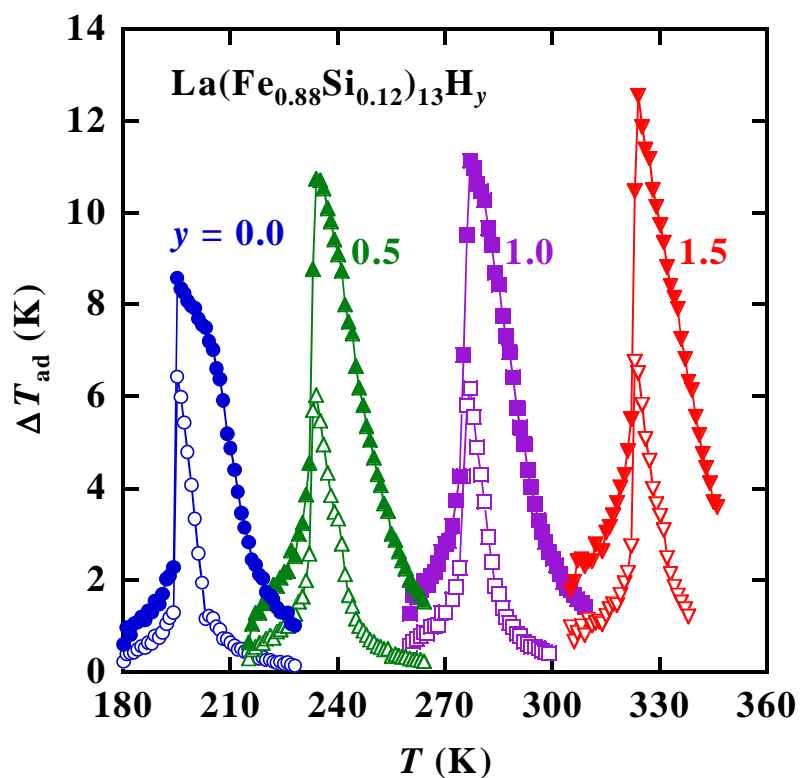


S. Fujieda et al. 2005, 0 - 2 bzw. 0 - 5 T

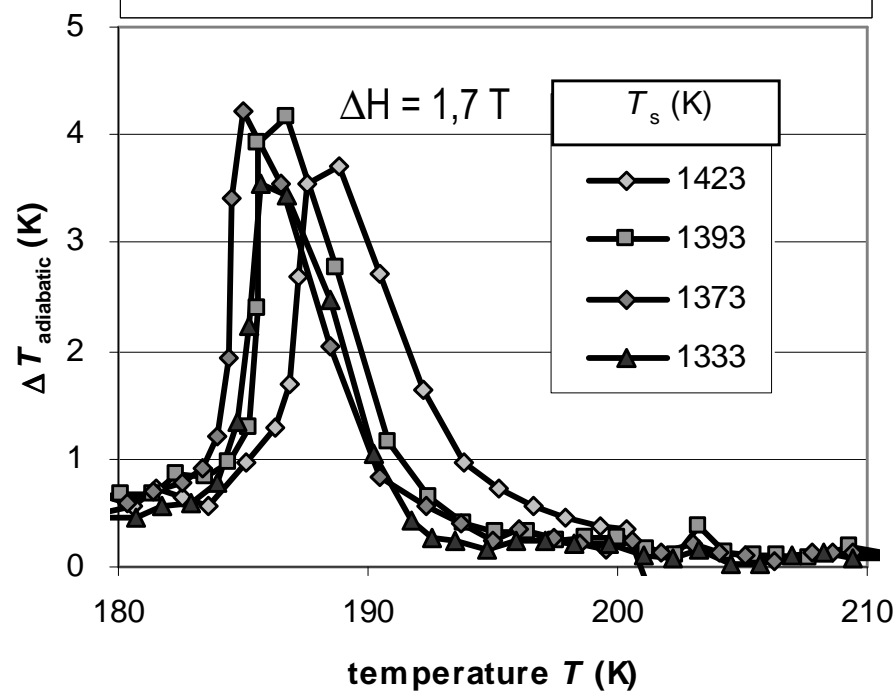
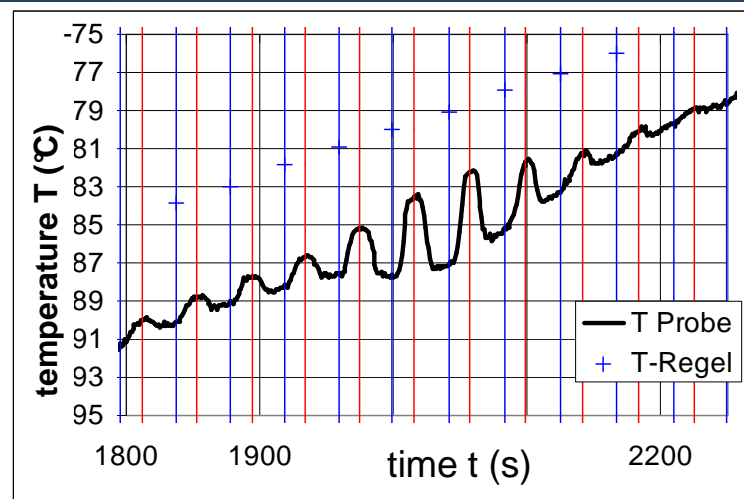


La(Fe_{0.915}Si_{0.085})₁₃, sintered

adiabatic temperature change

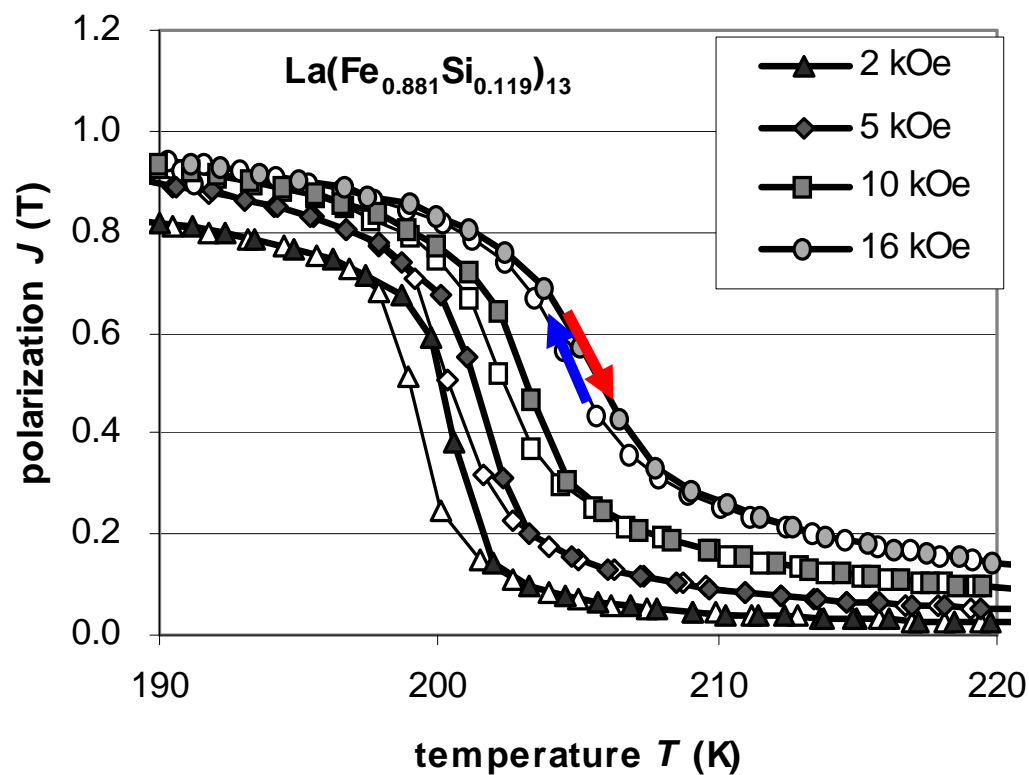
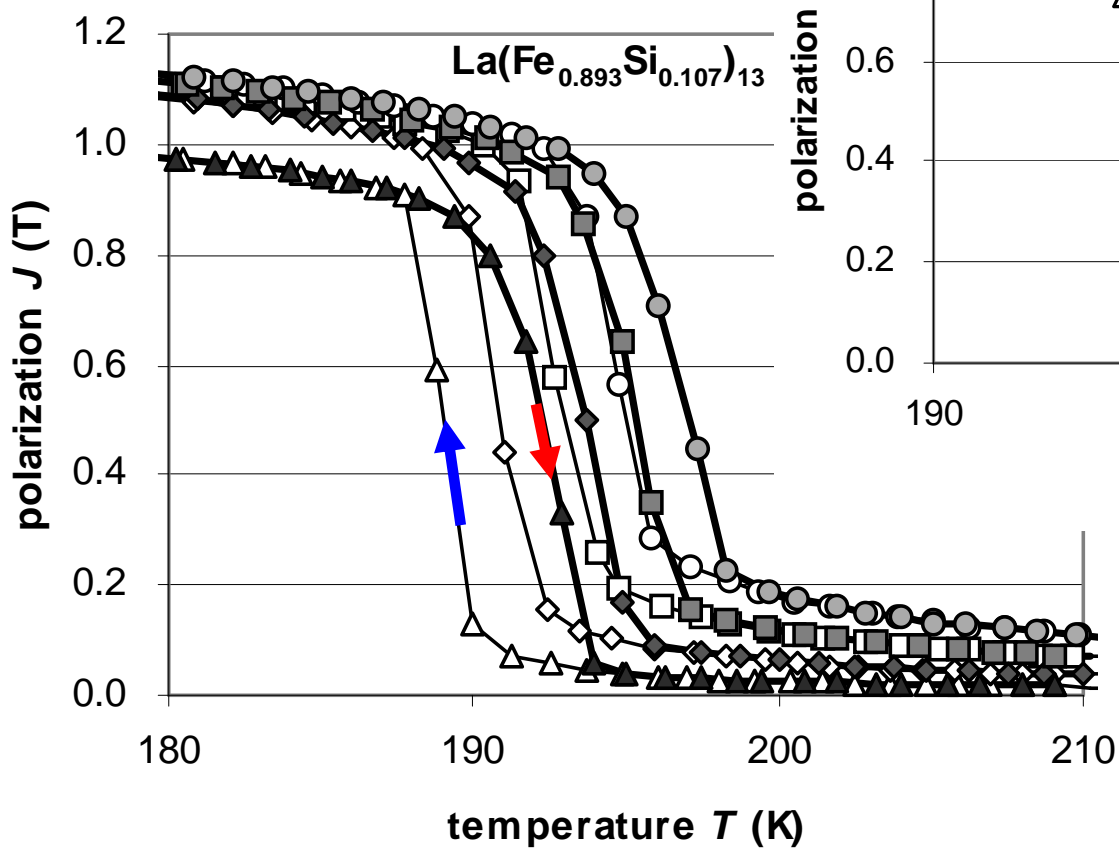


S. Fujieda et al. 2005, 0 - 2 bzw. 0 - 5 T



thermal hysteresis

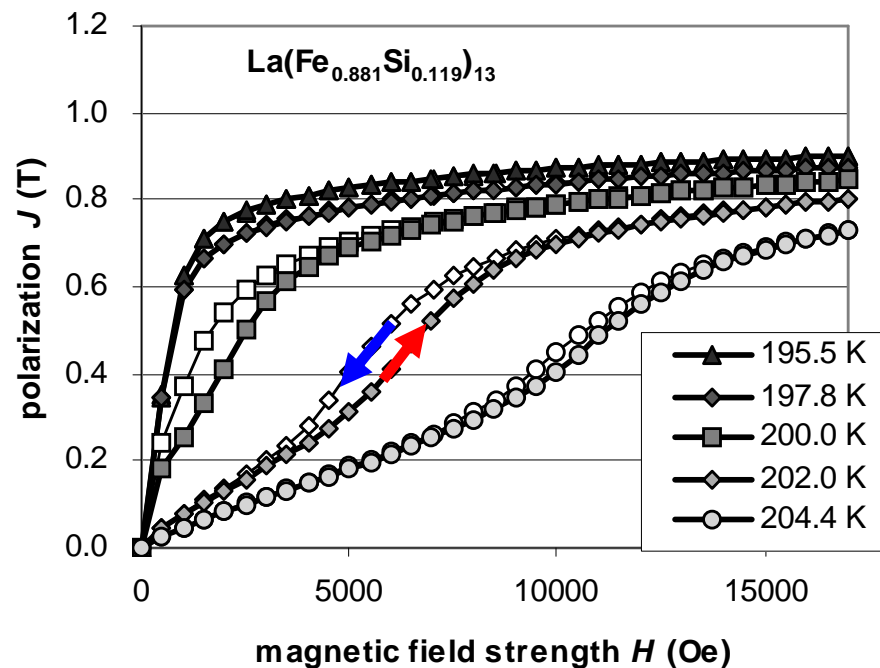
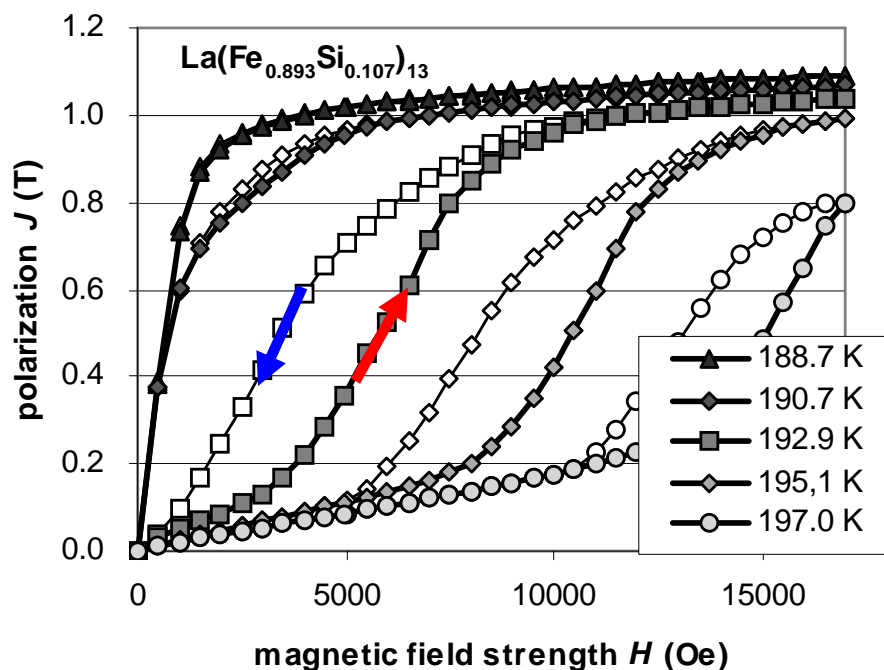
temperature
change rate: 8 K/min



hysteresis decreases
with increasing
Si content
and field strength

field hysteresis

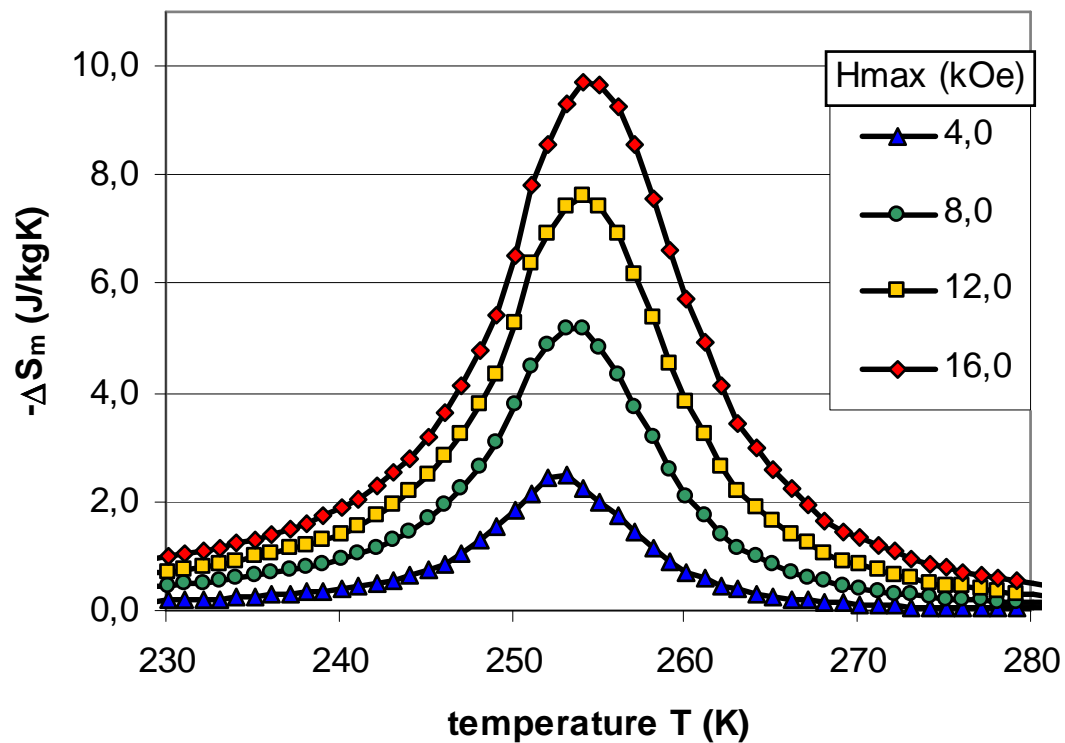
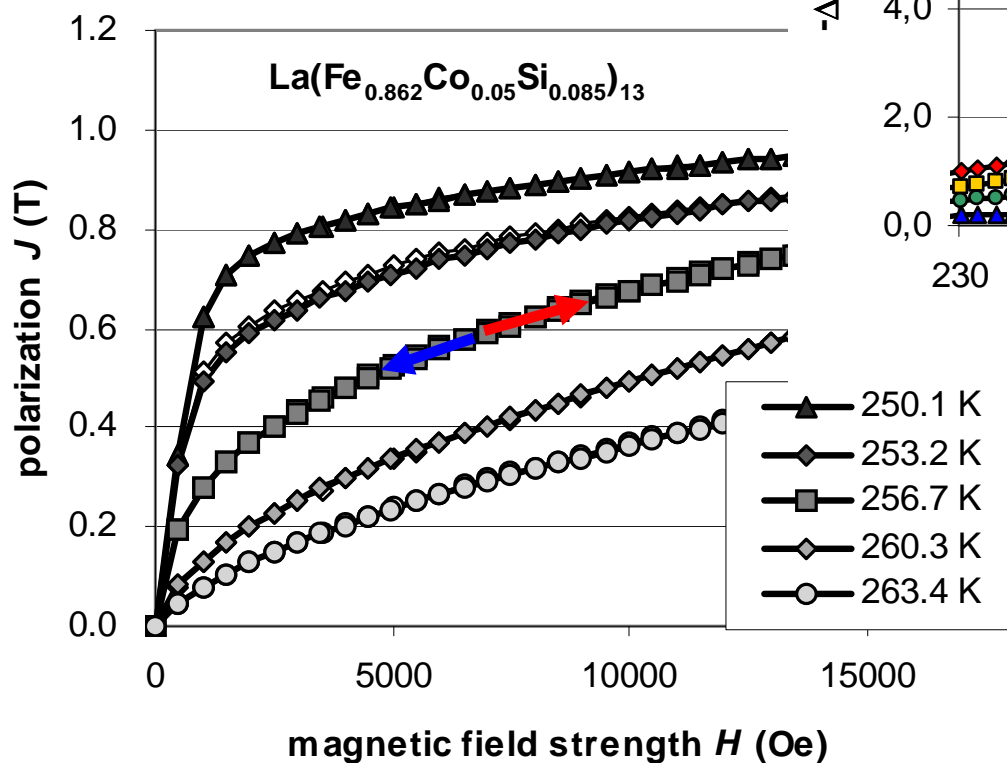
field sweep rate: 4.3 kOe/min



composition	temperature decreasing at -8 K/min			temperature increasing at +8 K/min			temperature hysteresis	field hysteresis
	$-\Delta S_{\text{magn}}$ (J/kgK)	T_{peak} (K)	ΔT_{WHH} (K)	$-\Delta S_{\text{magn}}$ (J/kgK)	T_{peak} (K)	ΔT_{WHH} (K)	ΔT_{peak} (K)	ΔH (kOe)
La(Fe _{0.893} Si _{0.107}) ₁₃	21.9	192.7	6.6	22.0	195.2	6.6	2.5	2.2
La(Fe _{0.887} Si _{0.113}) ₁₃	18.7	196.5	7.0	18.9	198.9	7.0	2.4	1.6
La(Fe _{0.881} Si _{0.119}) ₁₃	16.9	202.2	6.7	16.2	202.9	7.2	0.7	0.9

La(Fe_{0.862}Co_{0.05}Si_{0.085})₁₃ sintered

almost no
hysteresis at all



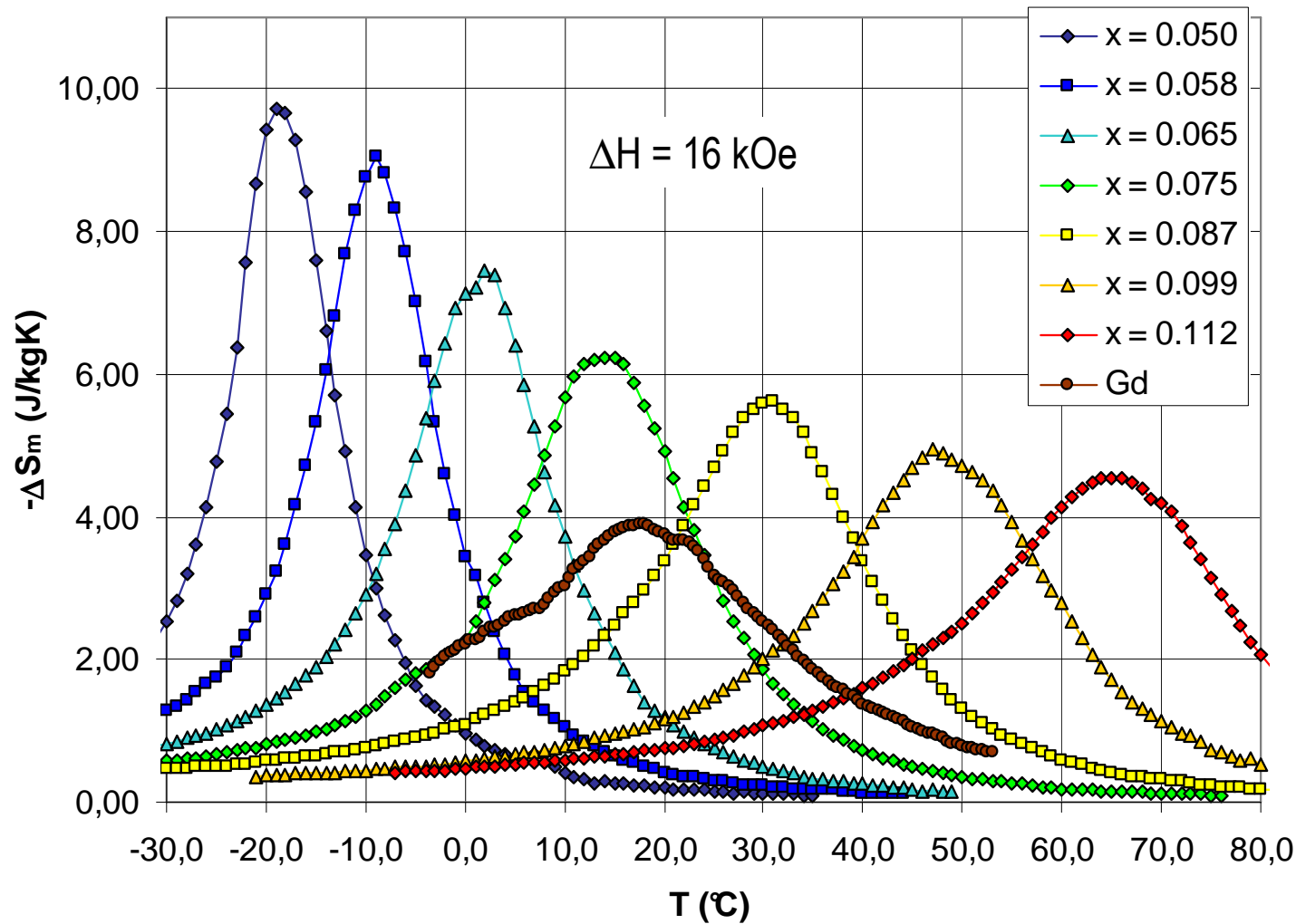
$$\rho = 7.24 \text{ g/cm}^3$$

$$-\Delta S_m @ 16 \text{ kOe} = 67 \text{ kJ/m}^3\text{K}$$

$$T_{\text{peak}} = -19 \text{ }^\circ\text{C}$$

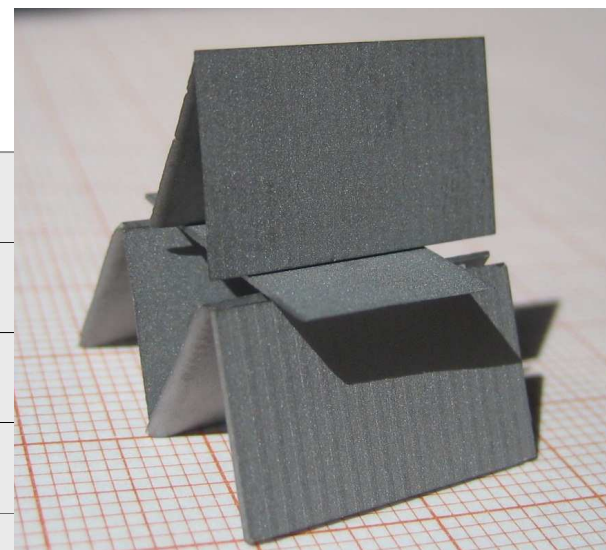
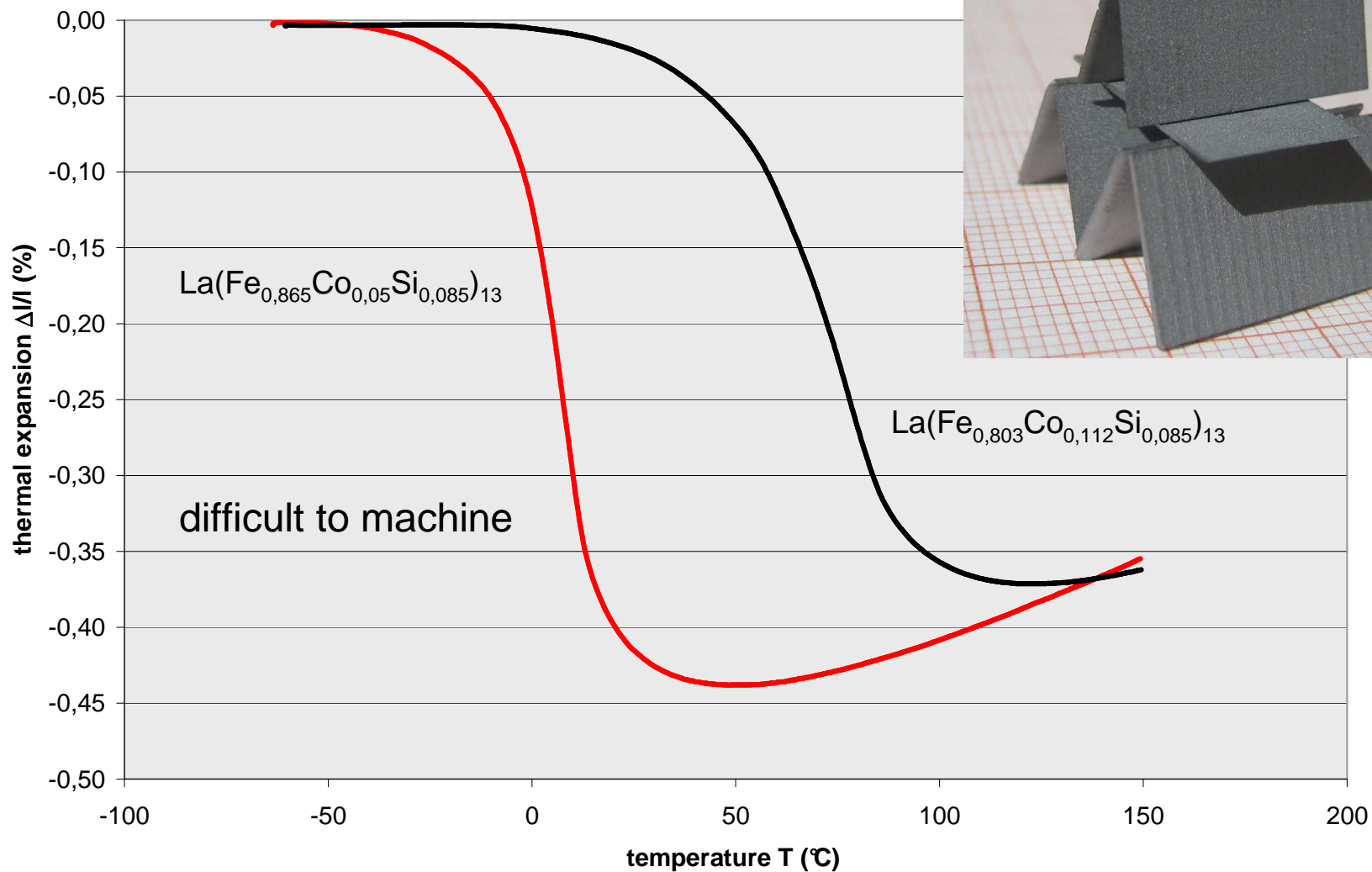
$$\Delta T_{\text{WHH}} = 14 \text{ K}$$

sintered $\text{La}(\text{Fe}_{0.915}\text{Co}_x\text{Si}_{0.085})_{13}$

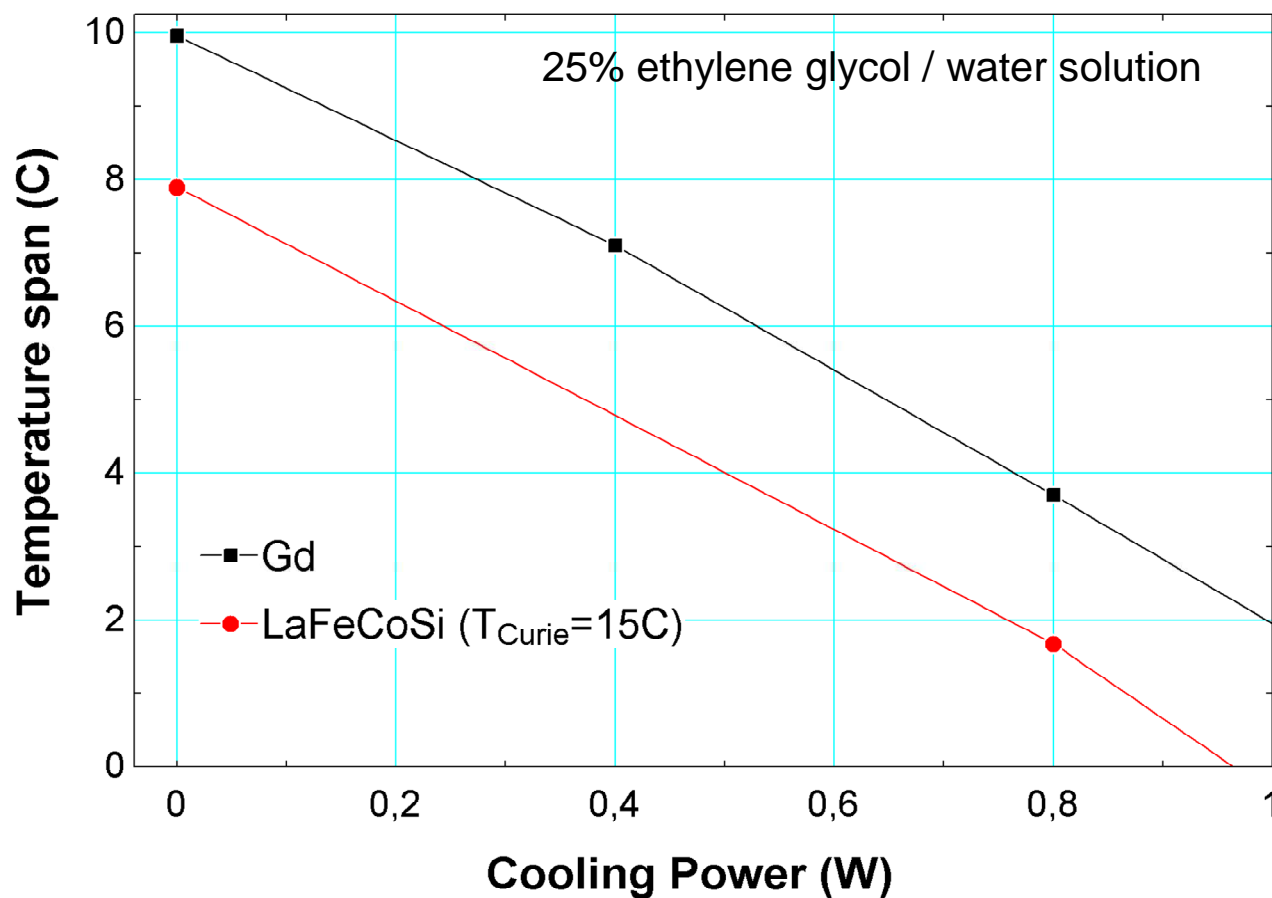


several kg
blocks
23x19x12 mm
produced on
lab scale for
prototyping

thermal expansion of sintered La-Fe-Co-Si



performance in a magnetic refrigeration prototype at Risø National Laboratory for Sustainable Energy



K.K. Nielsen
 J.B. Jensen
 K. Engelbrecht
 Ch. Bahl

2008

Adiabatic temperature change measured at Risø

$\Delta H = 1.1 \text{ T}$

Cast bulk samples

$\text{La}(\text{Fe}_{0.855}\text{Co}_{0.06}\text{Si}_{0.085})_{13}$

F.X. Hu 2005

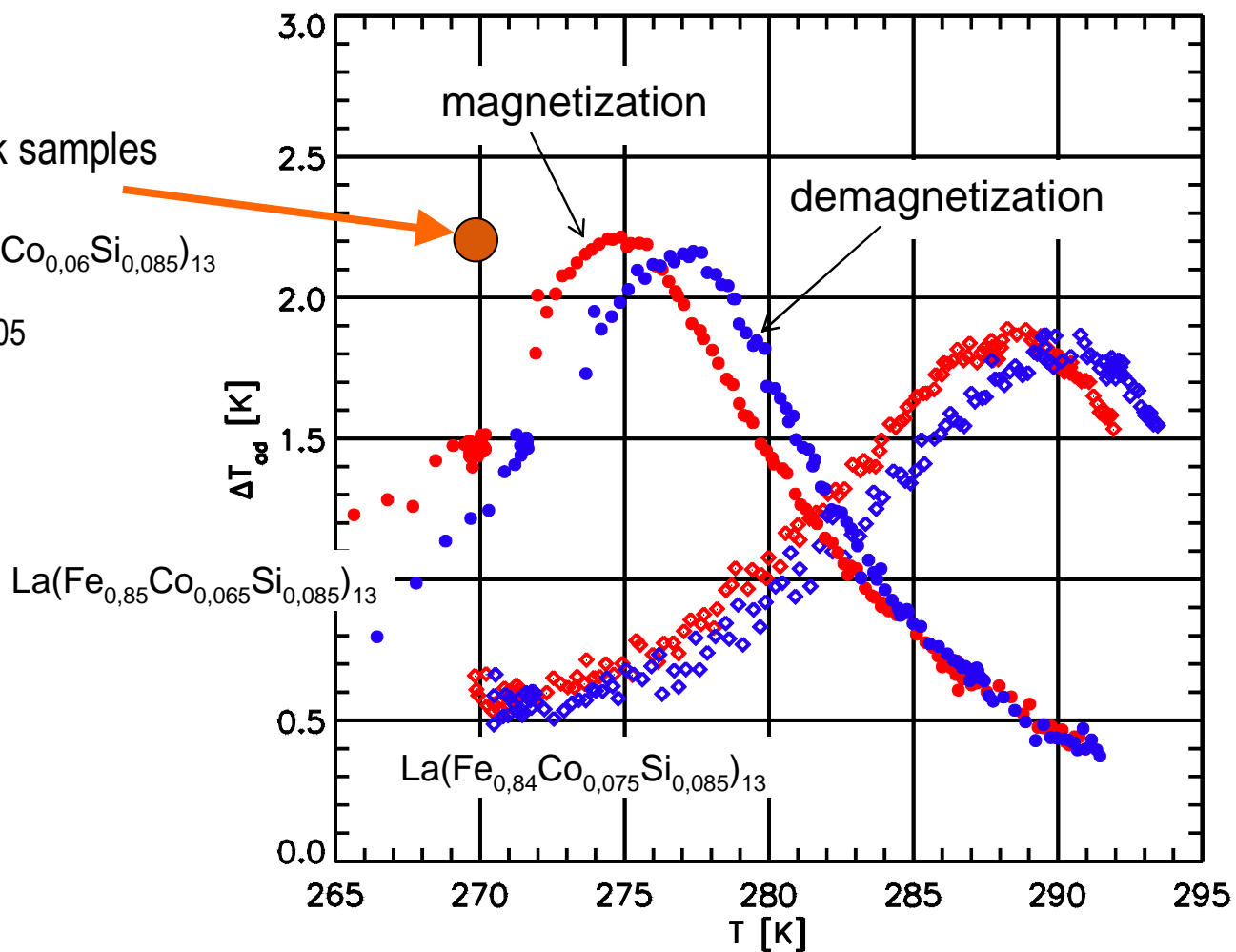
K.K. Nielsen

J.B. Jensen

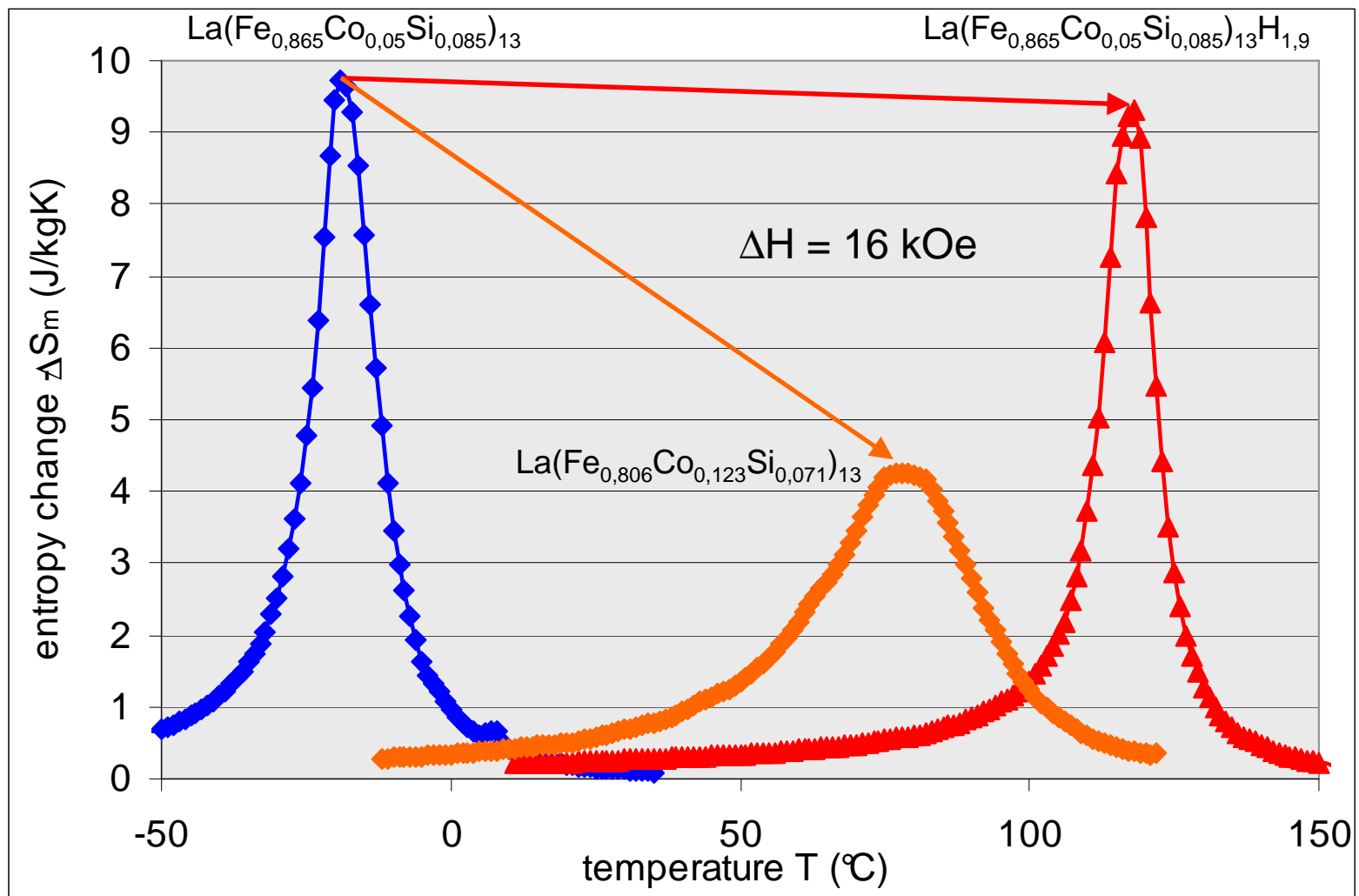
K. Engelbrecht

Ch. Bahl

2008



Hydrogenation of sintered La-Fe-Co-Si



Conclusion

- $\text{La}(\text{Fe},\text{Si})_{13}$ successfully prepared by powder metallurgy
- bulk material available after reactive sintering for a few hours
- hysteresis decreases with increasing Si and Co content
- Curie temperature controllable by Co or H substitution
- entropy change similar or better compared to Gd
- several kg with various Curie temperatures produced
- process upscalable on an industrial basis
- temperature span comparable to Gd

