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**COMPARISON OF CONVENTIONAL AND SPARK PLASMA SINTERING ROUTES
FOR THE FABRICATION OF ROOM TEMPERATURE MAGNETOCALORIC MN-FE-P-SI**

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ABSTRACT — We report on the structural and magnetic characterization of Mn-Fe-P-Si room temperature magnetocaloric compounds. The samples are prepared by different routes: powder annealing, conventional and spark plasma sintering. The effect of preparation route on the magnetic and structural properties is investigated.

1. INTRODUCTION

Hexagonal Mn-Fe-P-Si compounds are known to represent an extremely promising rare-earth-free road to room temperature magnetic refrigeration [1]. The giant magnetocaloric effect (MCE) associated with a magneto-elastic first-order phase transition has been studied in this system with particular emphasis on the tuning of critical temperature and magnitude of the entropy change as a function of the composition [2]. The goal is to obtain a material with optimal properties for applications and to reduce the thermal hysteresis. Many studies have been devoted to the tuning of hysteresis either by varying the composition [2, 3] or by using different preparation routes [4, 5]. Here we focus on the latter approach by comparing the influence of sintering technique on the Mn-Fe-P-Si powders produced by high-energy ball milling.

2. EXPERIMENTAL

Three different $\text{Mn}_{1.25}\text{Fe}_{0.7}\text{P}_{0.5}\text{Si}_{0.5}$ samples have been prepared. First, nanocrystalline precursors were produced from pure Fe_2P powder and P, Mn and Si chips by high-energy ball milling under Ar atmosphere. The rotation speed was 200 rpm during 30 min followed by 600 rpm for subsequent 5 hours. Next, a part of the obtained powder was pressed into a pellet of 6 mm diameter (“Bulk” sample). The pellet was sealed in a quartz ampoule in vacuum, sintered at 1373 K for 2 hours and annealed at 1123 K for 20 hours (with a heating/cooling rate of 1.2 K/min). The same heat treatment was applied directly to the fine powder (“Powder” sample). Finally, a sample of 8 mm diameter was prepared from the not annealed powder by spark plasma sintering (SPS) in Ar atmosphere at 1173 K for 30 min (with a heating/cooling rate of 100 K/min) and an applied pressure of 70 MPa (“SPS” sample). Low-field magnetic measurements were carried out using a LakeShore 7400 vibrating sample magnetometer (VSM). X-ray diffraction (XRD) patterns were collected by a Bruker D8 Advance diffractometer with Cu K α radiation.

3. RESULTS AND CONCLUSION

The temperature dependence of magnetization measured in a field of 1 T on cooling and heating is presented in Figure 1. The room temperature XRD patterns (Figure 2) show the presence of a single Fe_2P -type hexagonal phase (P-62m space group). The quantitative results are summarized in Table 1. The lattice parameters and grain size are estimated from the XRD measurements using Rietveld refinement. The inflection points *up*↑ and *down*↓ are determined from the magnetization curves. The transition temperature presented in Table 1 is taken as the average of the two inflection points, whereas the thermal hysteresis is calculated as the difference between them.

According to the measured lattice parameters and literature data [1], all the three samples are expected to be in the paramagnetic state at room temperature. The transition temperature, thermal hysteresis and low temperature saturation magnetization of the conventionally sintered “Bulk” sample here correspond to the data [1]. Sharper transition and lower thermal hysteresis are demonstrated by the annealed “Powder” sample.

The presence of uniaxial pressure and high heating/cooling rate during spark plasma sintering substantially accelerates processes and changes conditions of the material synthesis. To avoid melting of any component, we decreased the maximum temperature used in SPS compared to the conventional sintering. The “SPS” sample has a higher density and bigger grain size as compared to the “Bulk” sample. The lattice parameters *a* and *c* are also slightly different after SPS, which is confirmed by splitting of peaks (211) and (002) around 53.5° visible in Figure 2. The different heat treatment can change the final composition of the sample (in particular, fast heating can cause a loss of P in the milled powder). This can explain the shift of phase transition to lower temperature and larger thermal hysteresis [5]. The saturation magnetization of the “SPS” sample is found to be smaller with respect to the others at 150 K. The effect of additional annealing on the properties of the samples prepared by SPS will be presented.

Table 1. Structural and magnetic parameters of the samples.

Sample	Density	Grain size	Lattice parameters		Inflection point ↓	Inflection point ↑	Transition temperature	Thermal hysteresis
			<i>a</i>	<i>c</i>				
Powder	-	270 nm	609.5 pm	344.8 pm	282 K	290 K	8 K	286 K
Bulk	5.76 g/cm ³	160 nm	609.7 pm	345.4 pm	263 K	278 K	15 K	270.5 K
SPS	6.03 g/cm ³	230 nm	610.9 pm	342.9 pm	245 K	279 K	34 K	262 K

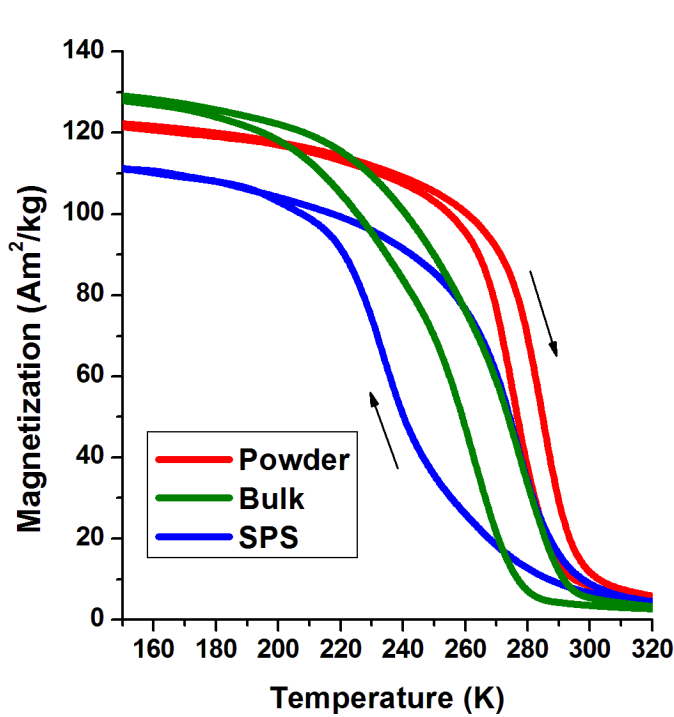


Fig. 1. Temperature dependence of the magnetization measured in 1 T field on cooling and heating.

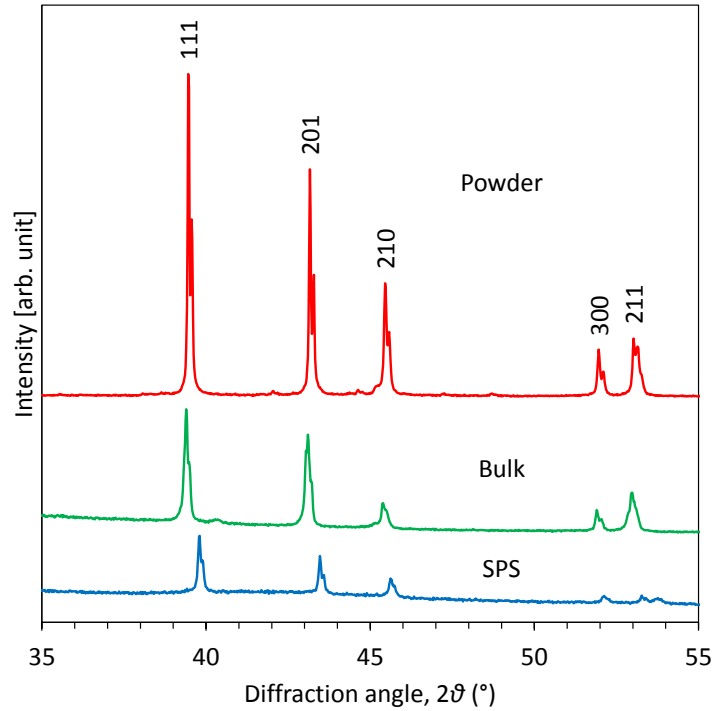


Fig. 2. X-ray diffraction patterns of the studied samples at room temperature.

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