

Specific heat capacity and thermal conductivity of the electrocaloric $(1-x)\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-x\text{PbTiO}_3$ ceramics between room temperature and 300 °C

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Abstract: We report the specific heat capacity and thermal conductivity of electrocaloric $(1-x)\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-x\text{PbTiO}_3$ ($x = 0, 0.1, 0.3$ and 0.35) ceramics between room temperature and 300 °C. The specific heat capacity for all ceramic samples is between 0.323 and 0.326 J/gK at 35 °C. For the samples with a high PbTiO_3 content ($x = 0.3$ and 0.35), a pronounced anomaly is observed in the specific heat capacity versus temperature at 130 °C and 153 °C, indicating the phase transition from the polar to a non-polar phase. The thermal conductivity in this system significantly depends on the PbTiO_3 content. The lowest thermal conductivity is obtained for $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$, and it increases with increasing PbTiO_3 content in the whole temperature range. For example at 23 °C the values of thermal conductivity of $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ and $0.65\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-0.35\text{PbTiO}_3$ are 1.25 W/mK and 1.43 W/mK, respectively.

Keywords: PMN-PT; thermal conductivity; specific heat capacity; relaxor-ferroelectric; electrocaloric

Specifična toplotna kapaciteta in toplotna prevodnost elektrokalične keramike $(1-x)\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-x\text{PbTiO}_3$ v temperaturnem območju od sobne temperature do 300 °C

Izvleček: V članku poročamo o specifični toplotni kapaciteti in toplotni prevodnosti elektrokalične keramike $(1-x)\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-x\text{PbTiO}_3$ ($x = 0, 0.1, 0.3$ in 0.35) v temperaturnem območju od sobne temperature do 300 °C. Specifična toplotna kapaciteta vseh merjenih vzorcev se pri 35 °C giblje v intervalu med 0,323 in 0,326 J/gK. Ko vzorce segrevamo, pri vzorcih z večjim deležem PbTiO_3 ($x = 0.3$ in 0.35) v meritvah toplotne kapacitete opazimo anomalijo, ki je značilna za premeno iz polarne v nepolarno fazo. Toplotna prevodnost trdne raztopine je izrazito odvisna od deleža PbTiO_3 . $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ izkazuje manjšo toplotno prevodnost kot $0.65\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-0.35\text{PbTiO}_3$ v celotnem temperaturnem območju. Na primer, pri temperaturi 23 °C je toplotna prevodnost $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ enaka 1,25 W/mK, medtem, ko je toplotna prevodnost $0.65\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-0.35\text{PbTiO}_3$ za 13 % višja.

Ključne besede: PMN-PT; toplotna prevodnost; specifična toplotna kapaciteta; relaksor-feroelektrik; elektrokaličnik

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1 Introduction

The relaxor-ferroelectric $(1-x)\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-x\text{PbTiO}_3$ (PMN-100xPT) ceramics exhibit a high dielectric permittivity, polarization, electromechanical [1-4] and electrocaloric (EC) effects [5, 6] and can be used for different applications, such as piezoelectric sensors, actuators, transducers [7-10] and cooling devices of a

new generation [11, 12]. In our previous work we have shown that PMN-30PT [13] and PMN-10PT [14] bulk ceramics possess the EC temperature changes (ΔT_{EC}) as high as 2.7 °C and 3.5 °C, respectively. These values are the highest reported for the lead-based ceramics [5, 6]. By using such ceramic PMN-PT elements in a cooling device with the efficient heat regeneration system, the

temperature span between the hot and the cold sides of the regenerator can exceed the temperature change of a single PMN–PT ceramic plate by several times [11]. However, when designing an EC device not only the EC properties, but also the thermal properties of the EC materials may have a great impact on its efficiency. Heat diffuses out of an EC layer of the thickness d in the time $t_{diff} \sim d^2 C_p / \lambda$, where C_p is the specific heat capacity and λ is the thermal conductivity [15, 16]. Therefore for an efficient cooling device, the EC material should possess a low C_p to λ ratio.

In the present study we report the specific heat capacity and thermal conductivity of the PMN–100xPT ceramics of different compositions, i.e., $x = 0, 0.1, 0.3$ and 0.35 . These material compositions were intentionally selected due to their specific properties. The PMN-rich compositions are of interest because of their large room temperature EC effects [13, 17, 18]. Furthermore, it has been recently shown that for a highly-efficient device the PMN and PMN-10PT ceramic elements are preferable than the elements from the PT-rich compositions due to their slim polarization versus electric field hysteresis loops and consequent lower losses [19]. On the other hand the PMN-35PT composition is the morphotropic phase boundary (MPB) composition and possesses high piezoelectric [1-3] as well as EC properties [14, 19], which could be an added value in the development of multifunctional devices.

The C_p was mainly studied in PMN and PMN-PT single crystals [20-25]. Only the C_p of PMN ceramics can be found in the literature [26, 27]. The λ of PMN and PMN-PT materials were previously studied mainly in the single-crystal form [20, 28-31] in the range between -271 °C and 117 °C. A few studies report the λ of PMN ceramics, but only at very low temperatures (i.e., from -271 °C to -173 °C) [26, 32]. The temperature dependence of ΔT_{EC} for PMN-PT exhibits the maximum value at the relaxor-to-ferroelectric phase-transition temperature [13, 14], which typically takes place in the middle of the low-temperature slope of the dielectric permittivity peak, i.e., below room temperature for PMN, and at ~ 160 °C for PMN-35PT. For designing cooling devices based on EC materials, the knowledge of the thermal properties of the PMN-PT ceramics is needed, especially at elevated temperatures.

2 Materials and methods

For the synthesis of the stoichiometric PMN-100xPT ($x = 0, 0.1, 0.3$ and 0.35) ceramic powder, PbO (99.9 %, Aldrich), MgO (99.9 % Alfa Aesar), Nb₂O₅ (99.9 %, Aldrich) and TiO₂ (99.8 %, Alfa Aesar) were used. The homog-

enized powder mixture was high-energy milled in a Retsch PM 400 planetary mill at 300 rpm for up to 140 h, and additionally milled in a Netzsch PE 075 / PR 01 attritor mill at 800 rpm for 4 h in isopropanol. The powder compacts were pressed uniaxially at 50 MPa and then isostatically at 300 MPa. The compacts were sintered in double alumina crucibles in the presence of the packing powder of the same composition at 1200 °C for 2 h with the heating and cooling rates 2 °C/min. Regarding the synthesis procedure of PMN-100xPT please see also [2, 13, 14, 17].

The densities of the sintered pellets were measured with a gas-displacement density analyser (Micromeritics, AccuPyc III 1340 Pycnometer). The relative densities (RD) of the samples were 97.08 %, 95.52 %, 96.31 % and 97.59 % for the compositions with increasing PT content. For these calculations the theoretical density of PMN was used, i.e., 8.13 g/cm³ (JCPDS 81-0861). For the microstructural analysis with a field-emission scanning electron microscope FE-SEM (JEOL FEG-SEM JSM-7600) the samples were ground, polished and thermally etched. The FE-SEM images of the etched surfaces reveal homogeneous and uniform microstructures (Figure 1). For the stereological analyses more than 250 grains per sample were measured. The grain size (GS) is expressed as the Feret's diameter (Figure 1).

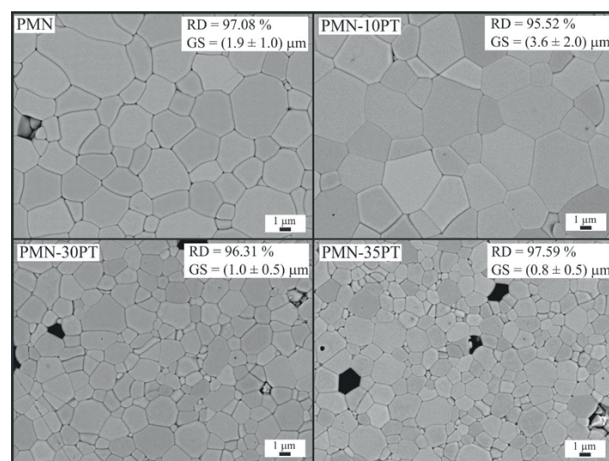


Figure 1: FE-SEM micrographs of PMN-100xPT ($x = 0, 0.1, 0.3$ and 0.35) ceramics.

The specific heat capacity versus temperature $C_p(T)$ of the ceramic samples was determined from the differential scanning calorimetry curves measured using a differential scanning calorimeter DSC (Netzsch, DSC 204 F1). The samples of ~ 5 mm in diameter and the thickness of ~ 1 mm were put in Pt crucibles with lids, and heated in a calorimeter with a heating rate of 2 °C/min from 35 °C to 300 °C. Each measurement was repeated two times. To determine the C_p of the samples,

sapphire (Netzsch, diameter of 5.2 mm, thickness of 1 mm) was used as the standard material.

For the measurements of the thermal transport properties versus temperature, the 8 mm thick pellets with 18 mm in diameter were prepared. The measurements were performed by the transient plane source technique [33] using the HotDisk TPS 2500S equipment (Hot Disk AB, Gothenburg, Sweden). The HotDisk sensor (Mica, 3.2 mm diameter, C5465) was placed between two ceramic pellets. The basic principle of the system is to supply the constant power to the sample via a Hot Disk sensor. The sensor is used as the heat source and the temperature monitor. The material was heated at 100 mW for 10 s. The length of the heating pulse was chosen short enough so that the sensor could be considered in contact with an infinite solid throughout the recording. In this way the thermal properties of the studied material could be determined by measuring the temperature increase of the sensor in a short period of time [34, 35]. The measurements at elevated temperatures (from 50 °C to 300 °C) were performed in a tube furnace (Entech) in nitrogen atmosphere to prevent the oxidation of the sensor. The step and the stabilization time of the measurements were 10 °C and 15 min, respectively. The step of 2 °C was used for the measurements performed in the range close to the phase transition temperature from polar to non-polar phases for PMN-30PT and PMN-35PT samples. At each temperature 5 measurements were performed. The waiting time between each measurement was long enough that the sample temperature reached the equilibrium with the furnace temperature (5 min). The room temperature measurements (i.e. 23 °C) were performed in air. In order to compare the thermal conductivity of ceramic samples with different chemical composition, i.e., PMN-100xPT ($x = 0, 0.1, 0.3$ and 0.35), the thermal conductivities were normalized to the sample density using the equation:

$$\lambda = \frac{\lambda_m - \lambda_{air} \cdot (1 - \rho)}{\rho} \quad (1)$$

where λ_m is the measured thermal conductivity of the sample, λ_{air} is the thermal conductivity of the air, which is equal to 0.026 W/mK [36] and ρ is the RD of the sample.

3 Results and discussion

The specific heat capacity, C_p , versus temperature of PMN-100xPT ($x = 0, 0.1, 0.3$ and 0.35) ceramic samples is shown in Figure 2, and the values of C_p at selected temperatures are collected in Table 1. At 35 °C the C_p

is between 0.323 and 0.326 J/gK for all samples. These values are in a good agreement with previously published data for PMN-29PT single crystals [24]. The C_p of PMN and PMN-10PT increases continuously with increasing temperature. In PMN such slow increase of C_p is observed up to ~220 °C, and at higher temperatures it remains almost constant. In PMN-10PT, the plateau is reached at a lower temperature, at ~120 °C (Table 1).

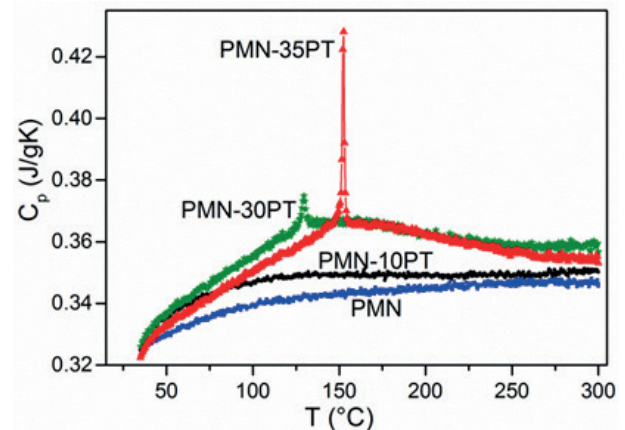


Figure 2: The $C_p(T)$ of PMN-100xPT ($x = 0, 0.1, 0.3$ and 0.35) ceramics.

Table 1: Selected C_p values of PMN-100xPT samples (from Figure 2).

T	C_p (J/gK)			
	PMN	PMN-10PT	PMN-30PT	PMN-35PT
35 °C	0.326	0.325	0.326	0.323
50 °C	0.330	0.335	0.336	0.332
100 °C	0.340	0.347	0.355	0.350
200 °C	0.345	0.349	0.363	0.362
300 °C	0.346	0.350	0.357	0.353

In PT-rich compositions the $C_p(T)$ behaviour is different; clear anomalies in C_p are observed at 130 °C and 153 °C related to the phase transitions from monoclinic and tetragonal phases to the high temperature cubic phase for PMN-30PT and PMN-35PT, respectively. These temperatures are in a good agreement with the published temperatures of dielectric permittivity anomalies; for PMN-30PT at ~130 °C (at 0.4 kHz) [14] and for PMN-35PT at ~160 °C (at 1 kHz) [1]. The phase transition anomaly observed in PMN-35PT is much more pronounced ($\Delta C_p = 0.062$ J/gK) in comparison to the one observed in PMN-30PT ($\Delta C_p = 0.008$ J/gK). No anomalies have been detected in PMN and PMN-10PT samples within our temperature measurement range. Namely, for PMN and PMN-10PT ceramics the dielectric anomalies were reported at ~15 °C [37] and ~40 °C [13, 37] (at 1 kHz), respectively, which is below or close to the lower limit of our measurement range.

The temperature dependence of thermal conductivity, λ , of PMN-100xPT ceramic samples is shown in Figure 3 and the λ values at selected temperatures are given in Table 2. At 23 °C the λ values of $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ and $0.65\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-0.35\text{PbTiO}_3$ are 1.25 W/mK and 1.43 W/mK, respectively. The λ increases with increasing temperature for all samples until approximately 250 °C, while above this temperature it is approximately constant. Such non-linear behaviour of the $\lambda(T)$ has been also observed in PMN-PT single crystals [20]. An interesting observation deduced from Figure 3 is that λ significantly increases with increasing PT content in the whole temperature range which is in agreement with [31], where PMN single crystals possess lower λ in comparison to PT single crystals in the whole measurement range, i.e., from -271 °C to 117 °C. Thermal conductivity measurements of BaTiO_3 and KNbO_3 single crystals revealed anomalies in $\lambda(T)$ at the phase transition temperatures [31]. In our study, no such anomalies were observed for PMN-30PT and PMN-35PT at our measurement conditions, i.e., with the step of 2 °C in the temperature intervals close to the respective phase transition temperature from ferroelectric to high-temperature cubic phase, see the insets in Figure 3.

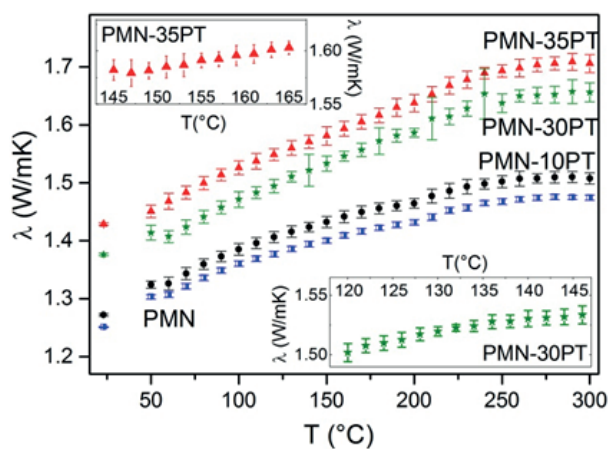


Figure 3: The $\lambda(T)$ of PMN-100xPT ($x = 0, 0.1, 0.3$ and 0.35) ceramics. Insets: The $\lambda(T)$ of PMN-30PT (below) and PMN-35PT (above) measured in the range close to the phase transition temperature from ferroelectric to the high-temperature cubic phase.

Table 2: Selected values of λ from Figure 3.

λ (W/mK)				
T	PMN	PMN-10PT	PMN-30PT	PMN-35PT
23 °C	1.25	1.27	1.38	1.43
50 °C	1.30	1.32	1.41	1.45
100 °C	1.36	1.39	1.47	1.53
200 °C	1.43	1.46	1.59	1.64
300 °C	1.47	1.51	1.66	1.71

As previously mentioned for the efficient cooling device, the C_p to λ ratio of EC material should be as low as possible. The values of the C_p/λ ratio of PMN-100xPT ceramic samples ($x = 0, 0.1, 0.3$ and 0.35) are shown in Figure 4. In the whole measurement range the lowest C_p/λ ratio is obtained for PMN-35PT ceramics and the highest ones are obtained for PMN and PMN-10PT ceramics. For example, at 50 °C the C_p/λ ratio of PMN-35PT is for 10 % lower than the one of PMN-10PT and PMN samples. Note also that the anomaly in the $C_p/\lambda(T)$ of PMN-30PT and PMN-35PT appears at 130 °C and 153 °C, respectively, and it is the result of the pronounced anomaly obtained in the $C_p(T)$ measurement (see Figure 2).

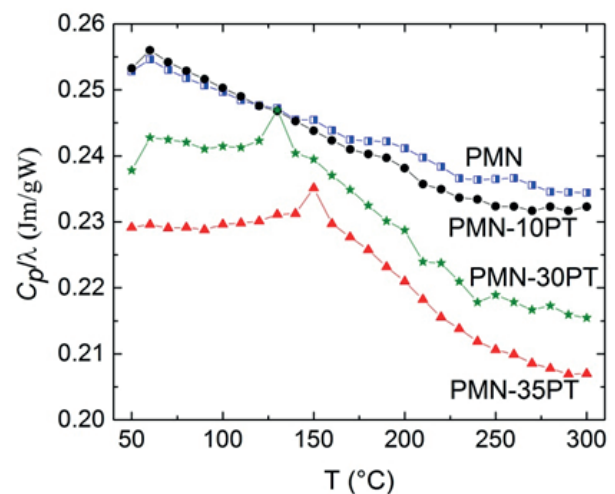


Figure 4: The $C_p/\lambda(T)$ of PMN-100xPT ($x = 0, 0.1, 0.3$ and 0.35) ceramics.

4 Summary and conclusions

In the present work, we examined the thermal properties of PMN-PT, which is one of the most promising EC materials compositions. We studied four different compositions; from relaxor PMN to the MPB composition PMN-35PT. The measurements of specific heat capacity versus temperature $C_p(T)$ vary for different PMN-PT compositions. At 35 °C the C_p is between 0.323 and 0.326 J/gK. With increasing temperature, the C_p increases continuously until it reaches the plateau value in PMN and PMN-10PT. No anomalies are observed in the whole measured temperature range. In PT-rich compositions the $C_p(T)$ dependence is different; pronounced anomalies are observed at 130 °C and 153 °C for PMN-30PT and PMN-35PT, respectively. The temperatures at which the anomalies occur correspond to the previously published phase transition temperatures from polar to high-temperature non-polar phases obtained from dielectric spectroscopy data. The heat capacity anom-

ally observed in PMN-35PT is much more pronounced ($\Delta C_p = 0.062$ J/gK) in comparison to the one observed in PMN-30PT ($\Delta C_p = 0.008$ J/gK).

The thermal conductivity λ of PMN at 23 °C is 1.25 W/mK, while the one of PMN-35PT is about 13 % larger. Such increase of λ with increasing PT content in the PMN-100xPT materials persists within the whole measurement temperature range.

To conclude, for an efficient cooling element the EC material should possess a low C_p/λ ratio. PMN-35PT ceramics possess the lowest C_p/λ ratio in the whole measurement range in spite of the anomaly at ~153 °C. On the other hand, the PMN and PMN-10PT materials possess a higher C_p/λ ratio, but no pronounced anomaly in C_p/λ is detected in this measurement range. The answer to the question: "Which composition of PMN-PT material is more appropriate for use in efficient EC cooling device of new generation?" is not trivial. From the thermal point of view, the more appropriate compositions are the ones with higher amounts of PT (i.e., PMN-30PT and PMN-35PT), but on the other hand these compositions possess high piezoelectric coefficients and well defined ferroelectric hysteresis loops, which could be a drawback for some specific electrocaloric applications.

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6 References

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