Multifunctional energy storage and piezoelectric properties of 0.65Pb(Mg\textsubscript{1/3}Nb\textsubscript{2/3})O\textsubscript{3}–0.35PbTiO\textsubscript{3} thick films on stainless-steel substrates

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Keywords: multifunctional, piezoelectric, energy storage, aerosol deposition, stainless steel, PMN-PT

Supplementary material for this article is available online

Abstract

The miniaturization of electronic devices and power systems requires the fabrication of functional components in the form of micrometer-sized thick films. A major challenge is the integration of functional ceramics with metals, which are considered incompatible with high-temperature ceramic processing. To overcome the integration barrier, an aerosol deposition (AD) spray-coating method based on room temperature deposition can be used. By employing the AD method, we were able to deposit relaxor-ferroelectric 0.65Pb(Mg\textsubscript{1/3}Nb\textsubscript{2/3})O\textsubscript{3}–0.35PbTiO\textsubscript{3} ceramic thick films on low-cost stainless-steel substrates. The as-deposited films were dense, with ∼97% of the theoretical density. Moreover, the post-deposition annealing at 500 °C did not result in any microstructural changes. Compared to the as-deposited films, the annealed films exhibit improved energy storage and electromechanical properties. The annealed thick films achieve a recoverable energy density of 15.1 J·cm\textsuperscript{-3} at an electric field of 1350 kV·cm\textsuperscript{-1} and an electric-field cycling stability of 5 million cycles. A piezoelectric response was detected through the entire film thickness by piezoelectric force microscopy. Macroscopic displacement measurements revealed a maximum relative strain of 0.38% at 1000 kV·cm\textsuperscript{-1}, corresponding to inverse effective piezoelectric coefficient of ∼40 pm·V\textsuperscript{-1}. In this study, we overcame the integration challenges and demonstrated the multifunctionalization of future ceramic-metal structures, as the deposited thick films on stainless steel exhibit energy storage capability and piezoelectric properties.

1. Introduction

Ceramic perovskite materials are featuring many promising characteristics for use in electronic applications [1]. One of the most promising materials are relaxor-ferroelectrics with a (100–x)Pb(Mg\textsubscript{1/3}Nb\textsubscript{2/3})O\textsubscript{3}–xPbTiO\textsubscript{3} (PMN–100xPT) solid solution, as they exhibit versatile dielectric, piezoelectric and relaxor-ferroelectric properties [2]. Of particular interest is the 0.65Pb(Mg\textsubscript{1/3}Nb\textsubscript{2/3})O\textsubscript{3}–0.35PbTiO\textsubscript{3} (PMN–35PT) material with a composition close to the morphotropic phase boundary due to its high piezoelectric performance in bulk form [3–5]. PMN–35PT possesses all the attributes that are important for electrical energy storage devices in pulsed-power and power-conditioning electronic applications and for electromechanical energy conversion in energy harvesting applications. Both energy storage and energy harvesting are crucial for the efficient use and management of electrical energy, which can ensure the sustainability of integrated electronic systems in the increasingly connected internet of things [6–8].
To miniaturize energy systems, functional components need to be fabricated in the form of thick films. Ceramic thick films with a thickness of a few micrometers enable device miniaturization and are a good compromise between the high volume-specific energy conversion performance offered by ceramic thin films and the large absolute energy conversion offered by conventional bulk ceramics [9]. However, integrating ceramic thick films into everyday devices is challenging. The fabrication of dense and high quality ceramic thick films by conventional thick-film deposition methods, such as screen-printing, tape-casting, or electrophoretic deposition consists of high temperature processes that often exceed 1000 °C [10]. The required high temperature treatment is a limiting factor as it prevents the integration of ceramics with materials that cannot withstand high temperatures, such as polymers and metals.

To overcome the integration barrier, a so-called aerosol deposition (AD) can be used. AD is a room temperature spray-coating method for the deposition of dense, several-µm-thick films on any type of substrate. The deposition mechanism is based on the collision of high-speed particles with the substrate’s surface and the densification occurs as a result of high kinetic energies involved [11–13].

Metals and their alloys are frequently considered as substrate materials in the electronics industry because they offer both heat sink and electrode functionality. As such, various metal substrates can expand the range of applications (actuators, sensors, transducers, energy harvesters and energy storage systems) [14, 15]. Metals as substrates offer good electrical conductivity and mechanical stability, which makes them a promising choice also for the field of flexible electronics [16]. Stainless steel (SS) is a versatile metal, durable, and resistant to corrosion and wear. As a low-cost material, it is in everyday use. However, little research has been done on the integration of ferroelectric ceramics on SS. Currently, there are few reports on the deposition of PMN–35PT thick films on SS. In our previous work, energy storage-efficient PMN–10PT thick films were integrated on SS substrates [26], while this study focuses on a ferroelectric PMN–35PT with a composition close to the morphotropic phase boundary responsible for high piezoelectric response in bulk ceramics [3–5].

In this work, we have deposited ceramic PMN–35PT thick films on SS substrate by employing the AD method, which allows densification of ceramics at room-temperature. We have shown that the integration of ceramic thick films on metals is possible and that such integration enables the multifunctionalization of future ceramic-metal structures, since the deposited PMN–35PT thick films exhibit energy storage capabilities and piezoelectric properties.

2. Materials and methods

PbO (99.9%, Aldrich), MgO (99.95%, Alfa Aesar), TiO₂ (99.8%, Alfa Aesar) and Nb₂O₅ (99.9%, Aldrich) were used for the synthesis of PMN–35PT ceramic powder. Two sets of oxide mixtures corresponding to the stoichiometry of Pb(1/3−x)Nb₂/3O₃ and PbTiO₃ were separately homogenized for 2 h in a planetary ball mill (PM 400, Retsch, Germany) at 200 min⁻¹, in isopropanol, using milling balls made of yttria-stabilized zirconia (YSZ) with diameter of 3 mm. After that, the powder mixture corresponding to the stoichiometry of PbTiO₃ was calcined at 750 °C for 2 h as previously suggested by Dragomir et al [27] and additionally milled for 2 h in a planetary ball mill (PM 400, Retsch, Germany) under the same conditions. Eventually, both powder mixtures were mixed together and homogenized for 2 h with a stoichiometry of 0.65Pb(Mg₁/₃Nb₂/₃)O₃–0.35PbTiO₃. The homogenization was carried out in a planetary ball mill (PM 400, Retsch, Germany) under the same conditions as before. This mixture was reacted for 24 h by mechanochemical-activation-assisted synthesis, using the same planetary ball mill, but at 300 min⁻¹ in a tungsten-carbide milling vial with a volume of 250 cm³, filled with 10 tungsten-carbide milling balls with a diameter of 20 mm. After synthesis, the powder was milled for 2 h, heated in a furnace at 900 °C for 1 h (with heating and cooling rates of 5 K·min⁻¹), and finally milled for 0.5 h. The milling processes were carried out in a planetary ball mill (PM 400, Retsch, Germany) at 200 min⁻¹, in isopropanol, using milling balls made of yttria-stabilized zirconia (YSZ) with diameter of 3 mm.

Prior to AD of the PMN–35PT powder on the substrate, the powder was sieved through an 80 µm mesh and vacuum dried for 12 h at 100 °C and 10 mbar. The substrate used was a commercial SS No. 304 (American Iron and Steel Institute) with a bright, polished surface A480: No. 8 (American Society for Testing and Materials) with dimensions 15 mm × 15 mm × 0.8 mm. The AD equipment was provided by InVerTec, Germany and the scheme of the set-up is represented elsewhere [28]. The process parameters during AD are listed in (supplementary material S1 available online at stacks.iop.org/JPEnergy/6/024004/mmedia). After AD, the samples were cleaned with ethanol and annealed at 500 °C for 1 h with heating and cooling rates of 2 K·min⁻¹. From now on, the samples will be referred to as ‘as-deposited’ and ‘annealed’.

The x-ray diffraction (XRD) analysis was performed with a high-resolution diffractometer (X’Pert PRO, PANalytical, Netherlands) using Cu–Kα₁ radiation. Diffraction patterns were recorded in the Bragg–Brentano geometry using a 100-channel X’Celerator detector in a 2θ range of 10°–120° with a step of
0.017° and an integration time of 100 s per step. For the Rietveld refinement, the Topas R package (version 2.1, Bruker AXS GmbH, Germany) was employed as described elsewhere [26]. According to the literature, a tetragonal (P4mm) and a monoclinic (Pm) crystal structure co-exist in the PMN–35PT solid solution [2, 3]. However, to calculate the crystallite size and microstrain of the film only the P4mm structure was selected. To fit the SS reflections originating from the substrate a cubic (Fm-3 m) crystal structure was used. The fundamental parameters approach (FPA) was used to fit the line-profile of the samples [29]. The FPA uses the geometrical properties of the diffraction experiment to build up the instrumental linewidth from first principles. Therefore, the FPA allows determination of the sample-dependent line-broadening contributions to the peak profile, which are dominated by microstrain and crystallite size broadening [30]. The effects of the microstructure were modeled using the Double-Voigt approach [31]. The volume-weighted mean crystallite size was calculated based on the integral breath method integrated in the Topas software. The calculated peak profiles and the agreement factors of the Rietveld refinement are listed in supplementary material S2.

Scanning electron microscopy (SEM) analyses of thick films’ surfaces and cross-sections were performed with a field-emission scanning electron microscope (FE-SEM, JSM-7600F, JEOL, Japan). For the cross-sectional analyses, the samples were cut, mounted in epoxy resin, ground and fine-polished using a colloidal silica suspension. Prior to the SEM analyses, the samples were coated with a 3 nm thick carbon layer using a Precision Etching and Coating System (PECS 682, Gatan, USA). The thickness and the porosity of thick films were evaluated from SEM cross-sectional images. For the estimation of porosity, the Image Tool software (UTHSCSA Image Tool Version 3.00, 2002 [32]) was used.

Piezoelectric force microscopy (PFM) was performed on two types of thick film samples; first, on the polished surface and second, on the polished cross-section of the thick films. The samples were polished as described above. An atomic force microscope (AFM; MFP-3D, Asylum Research, USA) and Ti/Ir coated silicon tips (Asyelec_01_R2, Oxford Instruments, Germany) were used for the analysis. To examine the polished surface, the electric voltage of 25 V and a frequency of ~350 kHz were applied between a conductive AFM tip and the bottom electrode of the films. To examine the samples in cross-section, electric voltage of 30 V and a frequency of ~350 kHz were applied to the thick film/epoxy resin composite in the virtual ground regime as previously described in [33, 34]. After PFM scans, the PFM phase hysteresis loops were measured on the polished surface samples in the switching spectroscopy off-electric-field mode with the pulsed DC step signal and the superimposed AC drive signal, as described in [35]. The waveform parameters were as follows: The sequence of rising steps of the DC electric field was driven at 20 Hz and a maximum amplitude of 100 V; the frequency of the triangular envelope was 0.99 Hz; a superimposed sinusoidal AC signal with an amplitude of 25 V and a frequency of ~350 kHz was used. Three cycles were measured in an off-electric-field mode.

For the electrical and electro-mechanical measurements, the top Au electrodes with a diameter of 1.5 mm were deposited by RF-magnetron sputtering. Measurements of dielectric permittivity and dielectric losses versus temperature are given in supplementary material S3. The unipolar polarization–electric field (P–E) hysteresis loops and bipolar strain–electric field (S–E) hysteresis loops of the films were measured at room temperature using an aixACCT TF analyzer 2000 (aixACCT Systems GmbH, Germany) equipped with a single beam laser interferometer. The applied electric field had a single sinusoidal waveform with a frequency of 100 Hz. In the electric-field fatigue cycling experiment the P–E loops were measured at an electric field of 500 kV·cm⁻¹. The direct piezoelectric coefficient (d33, eff) measurements of annealed thick films were performed using a Berlincourt piezometer (Take Control PM10, UK). Prior to the measurements, the samples were polarized by applying a DC electric field of 200 kV·cm⁻¹ at 160 °C for 15 min and then field-cooled to 30 °C.

3. Results

The structural and microstructural properties of as-deposited and annealed PMN–35PT thick films were investigated by XRD and SEM. The XRD patterns of the thick films and the PMN–35PT starting powder are shown in figures 1(a) and (b). None of the secondary phases were detected. The thick film samples exhibit additional minor SS reflections originating from the surrounding substrate. The perovskite reflections showed no significant peak shift after film deposition or after further annealing of the film. However, compared to the PMN–35PT powder, the as-deposited thick films exhibit a remarkable peak broadening. This broadening is due to reduced crystallite size and/or microstrain, and is typically observed in aerosol-deposited thick films, as the AD process involves cracking and fragmentation of colliding powder particles. Importantly, the broadening decreases slightly after annealing of thick films, indicating stress relaxation, which can occur even at moderate temperatures such as 300 °C [23, 36, 37].

The XRD patterns were examined in detail using Rietveld refinement, which quantitatively evaluates the crystallite size and microstrain of the samples (figures 1(c) and (d)). The calculated crystallite size of the PMN–35PT powder is 146 nm ± 3 nm and the microstrain is 8.4 × 10⁻⁴ ± 0.3 × 10⁻⁴. After the AD process
Figure 1. (a) XRD patterns in the 2θ range of 20°–60° for PMN–35PT powder, as-deposited and annealed thick films; (b) enlarged view of (200) reflection. Diffraction peaks corresponding to perovskite phase are indexed according to the cubic notation (JCPDS 81–0861). The corresponding (c) crystallite size and (d) microstrain evaluated using Rietveld refinement. SEM micrographs of as-deposited (e), (g), (i) and annealed (f), (h), (j) films in cross-section (e)–(h) and surface (i), (j) views. Green and yellow arrows mark pores several tens of nm in size and smaller pores in the nm-range, respectively.

the crystallite size of as-deposited thick films decreases to 17.2 nm ± 0.3 nm and the microstrain increases to 22.7 × 10^{-4} ± 2.1 × 10^{-4}. These results indicate a remarkable (8-fold) decrease in crystallite size that occurs during the high-energy particle collisions in the AD process. According to Exner et al [38] significant crystallite size reduction is a prerequisite for successful film consolidation. A crystallite size reduction is indicating the extent of converted kinetic energy into fracture energy, forming new surfaces that allow the particles to rebond and consolidate into a film [38]. Further annealing of the thick films has a minor effect on the crystallite size (21.9 nm ± 0.3 nm), but much more on the microstrain, which decreases significantly by 41% (down to 13.4 × 10^{-4} ± 1.2 × 10^{-4}).

The as-deposited and annealed thick films were analyzed by SEM in cross-sectional (figures 1(e)–(h)) and surface view (figures 1(i) and (j)). Both samples exhibit similar microstructure, despite the difference in the annealing process. Figures 1(e) and (f) reveals around 4 μm thick films firmly attached to SS substrates. The film-substrate interface does not show any damaging effects or any signs of chemical reaction between the film and the substrate. The polished cross-sections (figures 1(g) and (h)) show a crack free microstructure with a density of 97% ± 1%. The remaining porosity of 3% ± 1% is similar in as-deposited and annealed films and consists of isolated pores of several tens of nm in size (green arrows) and smaller pores in the nm-range (yellow arrows). The SEM micrographs of films’ surfaces confirm (figures 1(i) and (j)) a high film density.

Figure 2(a) shows the unipolar P–E hysteresis loops of the as-deposited and annealed films. The as-deposited films exhibit linear dielectric-like behavior at low electric field (300 kV·cm⁻¹), while a more lossy, round-like behavior is observed at high electric fields (1350 kV·cm⁻¹). The annealing of the films leads
Figure 2. (a) Unipolar P–E hysteresis loops of as-deposited (grey to black lines) and annealed (orange to red lines) thick films. P–E loops measured up to 300, 700 and 1350 kV cm\(^{-1}\) are shown in light to dark shades. \(U_{\text{rec}}\) and \(U_{\text{loss}}\) of the annealed film are represented by the red and grey patterned areas, respectively. (b) The electric-field dependence of \(U_{\text{rec}}\) and \(\eta\) for the as-deposited and annealed thick films. (c) The electric-field cycling dependence of a normalized \(U_{\text{rec}}\) and \(\eta\) for the as-deposited and annealed thick films at 500 kV cm\(^{-1}\). The lines between the values are only a guide for the eye.

...to a relaxor-ferroelectric character of the P–E loop. Moreover, the shape of the P–E loop remains similar with decreasing frequency (supplementary material S4). As we have seen in the SEM analysis (figures 1(e)–(j)), the microstructure is preserved after annealing the films. Therefore, one of the possible reasons for the changes in P–E behavior after annealing is related to stress relaxation, as previously shown for PMN–10PT thick films on SS [26]. Dielectric breakdown strength (DBS) was evaluated using Weibull analysis (supplementary material S5). The determined DBS is high, i.e. 1334 and 1351 kV cm\(^{-1}\) for as-deposited and annealed thick films, respectively. Compared to the as-deposited films, the annealed films exhibit lower hysteresis losses and higher polarization. At 1350 kV cm\(^{-1}\) the polarization of as-deposited and annealed films reaches 43 and 59 \(\mu\)C cm\(^{-2}\), respectively. The high electric polarization and DBS indicate possibility of achieving excellent energy storage properties.

The energy storage performance of the PMN–35PT thick films was evaluated by calculating the recoverable energy density \((U_{\text{rec}})\) and energy storage efficiency \((\eta)\). The \(U_{\text{rec}}\) represents the energy that is available in the discharge cycle and is therefore calculated by integrating the area between the discharge curve and the polarization axis (red patterned area in figure 2(a)). The charge–discharge cycle generates also hysteresis or polarization losses \((U_{\text{loss}})\), which are calculated by integrating the area between the charge and discharge curves (grey patterned area in figure 2(a)). The \(\eta\) is determined from the ratio between the recovered and stored energy: \(\eta = U_{\text{rec}}/(U_{\text{rec}} + U_{\text{loss}})\) and therefore, highly efficient energy storage capacitors, should have low \(U_{\text{loss}}\) [9].

In as-deposited thick films, the \(U_{\text{rec}}\) increases linearly with the electric field and reaches 10.7 J cm\(^{-3}\) at 1350 kV cm\(^{-1}\), which is already high for AD thick films prepared at room temperature (figure 2(b)). These values are almost an order of magnitude higher than \(U_{\text{rec}}\) of various bulk ceramics, reaching between 1 and 5 J cm\(^{-3}\) [39–41]. However, the \(\eta\)-value decreases with the electric field. It decreases from 78% at 100 kV cm\(^{-1}\) to 32% at 1350 kV cm\(^{-1}\) (\(\Delta\eta = 46\%\)). The annealed thick films have improved energy storage
Figure 3. PFM out-of-plane amplitude images of (a), (d) surface and (b), (e) cross-section views for (a), (b) as-deposited and (d), (e) annealed PMN–35PT thick films on SS. Switching spectroscopy experiment: the local hysteresis loops of the PFM out-of-plane (c), (f) phase measurements obtained at the point marked by blue crosses in panels (a), (d).

properties. The $U_{rec}$ value reaches 15.1 J cm$^{-3}$ at 1350 kV cm$^{-1}$, which corresponds to an increase of 41% compared to the as-deposited films (figure 2(b)). Overall, the annealed PMN–35PT films on low-cost SS substrate exhibit $U_{rec}$ values comparable to other AD thick films on conventional, more expensive platinized silicon substrates (Pt/Ti/SiO$_2$/Si) [42–46]. The $\eta$ of the annealed films is more stable with increasing electric field compared to the as-deposited films. It reaches 70% at 100 kV cm$^{-1}$ and 55% at 1350 kV cm$^{-1}$ ($\Delta \eta = 15\%$).

The properties during fatigue cycling under electric field are shown in figure 2(c). Both the deposited and annealed films withstood a large number of electric field cycles, i.e. $5 \times 10^6$. During the fatigue test, the $\eta$-value of both samples is stable while the $U_{rec}$ value of the deposited and annealed films decreases to $\sim$0.6 and $\sim$0.8 of the initial $U_{rec}$ value after 5 million cycles, respectively.

The local electromechanical response of the as-deposited (figures 3(a)–(c)) and annealed films (figures 3(d)–(f)) was investigated by PFM. The PFM out-of-plane amplitude images of the polished film surfaces (figures 3(a) and (d)) show a piezoelectric response associated with the fine-grained microstructure of as-deposited and annealed films (figure 1). It is evident that the annealed thick film exhibits more areas with enhanced piezoelectric response (bright regions) in comparison to the as-deposited film. The cross-sectional PFM out-of-plane amplitude images (figures 3(b) and (e)) confirm that both films are piezoelectrically active throughout their thickness, while the SS substrate or the encapsulating epoxy resin above the film does not show any piezoelectric response (dark areas). Furthermore, PFM switching spectroscopy was performed on the polished film surfaces. The analysis shows a local hysteresis loop of the PFM phase (figures 3(c) and (f)), which is evidence that the ferroelectric domains in the as-deposited and annealed films can be effectively switched.

Macroscopic displacement measurements were performed on the as-deposited and annealed films. The measured relative longitudinal strain with the applied electric field ($S$–$E$) is shown in figure 4(a). The $S$–$E$ loops of both samples are symmetrical i.e. without any imprint and their shape is typical of relaxor-ferroelectric materials. The strain response is much larger for the annealed films. The as-deposited and annealed films reach a maximum strain of 0.25% and 0.38%, respectively, at 1000 kV cm$^{-1}$. Figure 4(b) shows the $S$/$E$ coefficient (or inverse effective piezoelectric coefficient, $d_{33}^{*\text{eff}}$) evaluated from the slope of the $S$–$E$ loops by linear regression. In as-deposited films, $d_{33}^{*\text{eff}}$ gradually increases with the electric field, reaching 25 pm V$^{-1}$ at 1000 kV cm$^{-1}$, while in annealed films, $d_{33}^{*\text{eff}}$ reaches a constant value of 41 pm V$^{-1}$ from 650 to 1000 kV cm$^{-1}$. A small difference between the $d_{33}^{\text{eff}}$ and $d_{33}^{\text{eff}}$ coefficients (determined from the right and left sides of the ($S$–$E$) hysteresis loops, respectively) confirms a symmetrical electromechanical behavior. The displacement measurements were also performed with a double beam laser interferometer (DBLI, supplementary material S6) and the determined $d_{33}^{\text{eff}}$ is similar, indicating that there is no displacement due to substrate bending during the measurements. Additional measurements of the direct effective piezoelectric coefficient ($d_{33}^{\text{eff}}$) were performed after the DC poling at elevated temperatures. The $d_{33}^{\text{eff}}$ of the annealed films reaches $\sim$25 pC N$^{-1}$, which is close to the measured $d_{33}^{*\text{eff}}$ at low electric field. However, the
Figure 4. (a) Strain vs. electric field ($S$–$E$) hysteresis loops of as-deposited (grey-black) and annealed (orange-red) thick films. (b) The $d^*{_{33\text{eff}}}$ vs. $E$ of as-deposited (grey and black) and annealed (orange and red) thick films. The $d^*{_{33\text{eff}}}$ and $d^*{_{33\text{eff}}}'$ coefficients are evaluated with a linear regression of the positive and negative $E$ (i.e. the right and left sides of the $S$–$E$ hysteresis loops), respectively.

piezoelectric coefficient of PMN–35PT AD thick films is lower than that of PMN–35PT ceramics (typically above 525 pC·N$^{-1}$) [4, 47–49] and screen-printed thick films on platinized Al$_2$O$_3$ substrates (typically between 130 and 170 pC·N$^{-1}$) [50–52]. This can be attributed to several factors: first, the film-substrate clamping effect in thick films [51, 53] and second, the much smaller grain size in AD thick films [47] in comparison to screen-printed films or bulk ceramics, due to the low processing temperature.

4. Summary and conclusions

By employing the AD method, which enables the fabrication of dense ceramic layers at room temperature, we deposited ceramic PMN–35PT thick films on low-cost stainless-steel substrates. The deposited 4 µm thick films exhibit high density, namely 97% of the theoretical density, with pores in the range of a few to tens of nm. Post-deposition annealing at moderate temperatures of 500 °C does not lead to any microstructural changes, but reduces the microstrain by 41%.

The annealed thick films exhibit high polarization (59 µC·cm$^{-2}$) and high DBS ($\sim$1350 kV·cm$^{-1}$), leading to promising energy storage performance. The $U_{rec}$ value of the annealed PMN–35PT films on the low-cost SS reaches 15.1 J·cm$^{-3}$ at 1350 kV·cm$^{-1}$, which is similar to other AD thick films on conventional, more expensive silicon substrates (Pt/Ti/SiO$_2$/Si). Moreover, both the as-deposited and annealed films withstand a large number of electric field cycles, namely $5 \times 10^6$. The electromechanical response of the thick films was investigated in cross-section using the PFM technique. The as-deposited and annealed films are piezoelectric/ferroelectric over the entire film thickness. The macroscopic electromechanical measurements of the as-deposited and annealed films revealed a longitudinal strain of 0.25% and 0.38% at 1000 kV·cm$^{-1}$, corresponding to $d^*_{33\text{eff}}$ of 25 and 41 pm·V$^{-1}$, respectively.

In summary, we have shown that the integration of PMN–35PT thick films on SS is possible and that such integration enables the multifunctionalization of future ceramic-metal structures, since the deposited PMN–35PT thick films exhibit energy storage capability and piezoelectric properties. The multifunctional properties are already observed in films deposited at room temperature. However, they are further enhanced by post-deposition annealing at moderate temperatures of 500 °C.

Data availability statement

The data generated and/or analyzed during the current study are not publicly available for legal/ethical reasons but are available from the corresponding author on reasonable request.

Acknowledgments

H U and M S acknowledge the Slovenian Research Agency (research project J2-3058, young researcher project PR-08977, bilateral projects BI-FR/21-22-PROTEUS-004 and BI-DE/20-21-012 and research core
funding P2-0105) and JSI Director’s fund 2017-ULTRACOOL. They thank R Elshuraf (Erasmus+ program), V Fišinger and J Gilenšek for help in the laboratory.

**Conflict of interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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