

Pb(Sc_{1/2}Nb_{1/2})O₃–Pb(Mg_{1/3}Nb_{2/3})O₃–PbTiO₃ 织构陶瓷的电学性能和热稳定性

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摘 要: 采用 BaTiO₃ (BT) 片状微晶制备了 [001] 取向的 0.29PSN–0.39PMN–0.32PT(0.29Pb(Sc_{1/2}Nb_{1/2})O₃–0.39Pb(Mg_{1/3}Nb_{2/3})O₃–0.32PbTiO₃) 织构陶瓷。制备了不同模板含量的织构陶瓷, 并在织构度、微观结构以及压电、铁电和介电性能方面进行了比较。在 PSN–PMN–PT 织构陶瓷中, 在 3% (体积分数) BaTiO₃ 模板下实现了高达 98.6% 的 [001] 织构度, 并且获得了高的压电性能 ($d_{33} = 816$ pC/N, $d_{33}^* = 1080$ pm/V) 和高 Curie 温度 ($T_c = 185$ °C), 这远远优于随机取向的陶瓷。此外, 与具有相近压电常数的铅基陶瓷相比, 织构化 PSN–PMN–PT 陶瓷表现出显著改善的热稳定性。高质量的织构化三元陶瓷在新一代高性能、宽温度范围的机电器件中具有广阔的应用前景。

关键词: 织构陶瓷; 铌钪酸铅–铌镁酸铅–钛酸铅; 压电性能; 热稳定性

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以 Pb(Mg_{1/3}Nb_{2/3})O₃–PbTiO₃ (PMN–PT) 和 Pb(Zn_{1/3}Nb_{2/3})O₃–PbTiO₃ (PZN–PT) 为代表的弛豫铁电材料具有良好的电学性能被广泛应用于传感器、换能器和能量收集器等领域中^[1–3]。长期以来, 对高性能、高灵敏度以及高度集成化的电子器件的需求日益增长。为了满足这些不断攀升的需求, 研究者们一直致力于提升压电材料的电性能。钙钛矿铁电体的电性能与其晶体取向密切相关^[4–5]。因此, 制备沿优势性能方向的单晶是提高钙钛矿型铁电体电性能的有效途径。然而, 单晶的实际应用受到其相对较高的成本、低的三方–四方相变温度以及由于融化不一致的问题而导致的组成成分不均匀等因素的限制^[6–9]。

为了解决上述单晶的应用问题, 模板晶粒生长 (TGG) 法制备织构陶瓷被认为是最有前途的方法之一。TGG 法是将定向的模板籽晶引入陶瓷中, 引导陶瓷晶粒沿某一取向择优生长, 在优势方向上获得类似单晶的电学性能^[10–12]。以 PMN–PT 为例, 高质

量的 [001] 取向织构陶瓷的压电性能为 1 200 pC/N, 可达到同类单晶体的 50%~70%, 是普通无取向陶瓷的性能的 1.6 倍^[13]。但由于二元 PMN–PT 弛豫铁电固溶体的低的相变温度 ($T_{r-t} < 60\sim 90$ °C) 以及较小的矫顽场 ($E_c < 3$ kV/cm), 无法满足高温高功率超声换能器的应用。研究者在二元弛豫铁电固溶体中引入具有高居里温度的第三组元, 形成新的三元弛豫铁电固溶体来提升陶瓷的相变温度^[14–17]。Wu 等成功制备出织构度高达 93% 的 PIN–PMN–PT 织构陶瓷, 获得了 $d_{33} = 824$ pC/N, $T_c = 203$ °C 的优异性能^[18]。Liu 等^[19]也制备出织构度为 98% 的 PMN–PZ–PT 织构陶瓷, 表现出优于 PMN–PT 等二元体系的压电性能和温度稳定性: $d_{33} = 860$ pC/N, $d_{33}^* = 1050$ pm/V, $T_c = 222$ °C。由上述结果可以看出, 在 PMN–PT 二元体系中引入具有高相变温度的组元, 通过 TGG 法实现陶瓷织构化, 可以同时提升陶瓷的压电性能和温度稳定性。

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在本工作中,以 PSN-PMN-PT 三元弛豫固溶体压电材料为研究对象,采用模板晶粒生长法进行陶瓷的[001]取向生长研究,讨论籽晶模板含量对陶瓷相结构、微观形貌、电学性能及热稳定性的影响规律。

1 实验

1.1 材料制备

通过拓扑化学转化法^[20]在熔盐中合成 BaTiO₃ 模板。首先,将 Bi₂O₃ 和 TiO₂ 与等质量的 NaCl 和 KCl 混合,将该混合物加热至 1 100 °C 反应 1 h 以获得 Bi₄Ti₃O₁₂; 第二步是将 Bi₄Ti₃O₁₂、BaCO₃ 和 TiO₂ 与 BaCl₂ 和 KCl 混合后在 1 080 °C 反应 2 h 获得 BaBi₄Ti₄O₁₅; 最后,将 BaBi₄Ti₄O₁₅、BaCO₃ 和 NaCl 和 KCl 混合后在 850~900 °C 反应 3 h 获得 BaTiO₃ 模板。

采用两步法制备 0.29Pb(Sc_{1/2}Nb_{1/2})O₃-0.39Pb(Mg_{1/3}Nb_{2/3})O₃-0.32PbTiO₃ 基体粉末。首先,以 MgO (99.99%, 质量分数)、Sc₂O₃ (99.9%) 和 Nb₂O₅ (99.9%) 为原料,分别在 1 000 °C 和 1 150 °C 合成 MgNb₂O₆ (MN) 和 ScNbO₄ (SN) 前驱体。将 PbO (99.9%)、MN、SN 和 TiO₂ (99%) 球磨 24 h, 并将混合物在 750 °C 预烧结 2 h, 形成纯钙钛矿结构。通过拓扑化学微晶转化方法合成具有[001]取向的片状 BaTiO₃ (BT) 微晶作为模板, 加入到由 0.29PSN-0.39PMN-0.32PT 基体粉末、溶剂、分散剂、增塑剂和黏合剂球磨后的均匀浆料中。分别将 $x = 0$ 、1%、2%、3%、4% (体积分数) BT 模板添加到浆料中并在较低的转速下球磨 30 min。接下来,通过控制刮刀的高度和基带的速率来流延浆料。将干燥后的胚带切割、叠放、排胶。最后将样品在 1 270 °C 保温 9 h 进行烧结。

1.2 样品表征

用 Bruker D2 型 X 射线衍射分析普通陶瓷和织构陶瓷的相结构, 测试条件: Cu 靶, 扫描步长 0.02°, 间隔时间 0.2 s。BT 模板的定向过程: 将分散在乙醇中的 BT 模板滴在载玻片上, 烘干后对样品进行 XRD 测试。通过 XRD 数据计算 Lotgering 因子 F_{001} 。 F_{001} 计算公式如式(1)所示^[21]:

$$F_{001} = \frac{P - P_0}{1 - P_0}, P = \frac{\sum I_{(00l)}}{\sum I_{(hkl)}}, P_0 = \frac{\sum I_{0(00l)}}{\sum I_{0(hkl)}} \quad (1)$$

其中: I 和 I_0 分别为织构陶瓷和普通陶瓷的衍射峰强度。用场发射扫描电子显微镜(VEGA II XMU)与能量色散光谱仪(EDS)结合来表征陶瓷的微观结构

和成分特征。为了进行电性能测量,将样品涂上银浆并在 550 °C 烧结 40 min 以形成电极,然后在 90 °C 的硅油中在 25 kV/cm 下极化 15 min。用 ZJ-6A 型准静态 d_{33} 测试仪测量压电系数 d_{33} 。用 E4980A 型 LCR 计测量介电系数 ϵ_r 和介电损耗 $\tan \delta$ 的温度依赖性。用 TF Analyzer 2000E 型铁电测试系统测量极化电场($P-E$)曲线和应变电场($S-E$)曲线。大信号 d_{33}^* 由单极 $S-E$ 曲线的斜率确定。对于温度稳定性测试,样品在高温下退火 30 min, 然后在室温下用 ZJ-6A 型准静态 d_{33} 测试仪测量 d_{33} 。

2 结果与讨论

2.1 XRD 分析

图 1 为 BaTiO₃ 模板的 XRD 谱、SEM 照片和不同 BT 模板含量的 0.29PSN-0.39PMN-0.32PT- $x\%$ (体积分数) 织构陶瓷 XRD 谱。由图 1 可见,通过拓扑化学转化法制备了获得了纯钙钛矿相的模板, BaTiO₃ 以(001)、(100)和(002)、(200)衍射峰为主, 非(001)衍射峰强度基本可以忽略, 这些特征表明了片状 BaTiO₃ 模板[001]取向特征优异。通过测试了 BaTiO₃ 模板微观结构发现, 其长度为 10~15 μm , 厚度约为 0.5~1.0 μm , 径厚比 > 10 , 且分散性良好, 符合 TGG 法制备高质量织构陶瓷的模板要求, 所有样品都具有纯钙钛矿结构, 没有检测到任何的第二相存在。与随机取向的陶瓷相比, 织构化的陶瓷显示出(110)衍射峰的强度降低, 而(001)和(002)衍射峰的强度显著增加。这表明织构化样品具有沿[001]方向的择优取向生长。随着模板含量的增加, 织构化陶瓷的 Lotgering 因子 F_{001} 呈现先增大后几乎不变的趋势。这是因为当 BaTiO₃ 模板含量为 1% 时, 织构陶瓷中的模板不足以完全诱导基体晶粒沿[001]方向生长, 其织构度为 97.6%。当模板含量为 3% 时, 基体晶粒几乎沿[001]方向生长, 陶瓷的织构度 F_{001} 也达到了最大值 98.6%。织构化过程中高度择优取向的晶粒严重依赖于 BT 模板的含量, 当模板含量较低时织构驱动力较低, 不能引导所有的晶粒实现取向生长, 此时陶瓷基体中仍有大量的随机晶粒因此表现出较低的织构度。而随着模板含量的增加织构驱动力增加, 足以引导大部分晶粒实现取向生长过程, 在基本消耗完随机晶粒后取向晶粒之间相互挤压碰撞不再生长, 此时织构化过程基本完成, 实现了高度的[001]择优取向生长。所有的织构陶瓷的 F_{001} 在 97%~99%, 表明 BaTiO₃ 作为模板可以有效地来提高 PSN-PMN-PT 陶瓷的[001]取向度。此外,

由图 1d 可以看出,随着 BT 模板含量的增加,(200) 衍射峰相较普通陶瓷变得更加展宽,并且不再对称。(200) 峰的宽化主要归因于织构化 PSN-PMN-PT 陶瓷中 BT 模板和 PSN-PMN-PT 基体之间的晶格失配导致的应力引起的晶格畸变^[22]。

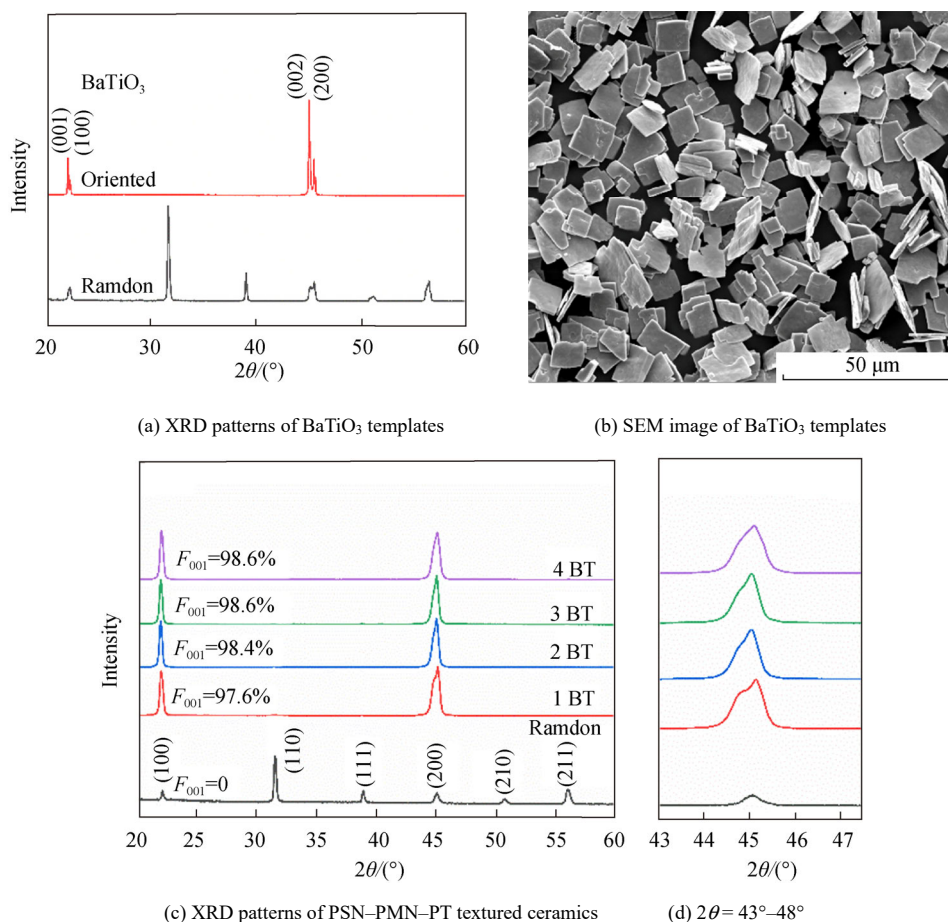
2.2 微观性能分析

图 2 为随机取向的和不同模板含量的织构化 0.29PSN-0.39PMN-0.32PT 陶瓷的 SEM 照片。由图 2 可见,在随机取向陶瓷中,晶粒没有特定的生长方向,表现为杂乱无一的状态。而织构陶瓷的晶粒则展现出明显的择优取向,但仍然存在少量的随机晶粒。陶瓷织构的形成直接来自于模板向基质陶瓷的外延成核和生长。由于模板在基质陶瓷中的不均匀分布,造成了少量随机晶粒的产生。模板在基质陶瓷中稳定存在且排列良好,这是织构陶瓷具有较高织构度的根本原因。织构陶瓷中在 BT 模板周围存在少量闭口气孔,这导致陶瓷介电性能的降低^[23]。

2.3 介电与压电性能分析

图 3 为随机取向和不同模板含量的织构

0.29PSN-0.39PMN-0.32PT 织构陶瓷的压电和介电性能。由图 3 可见,随着 BT 含量和[001]织构含量的增加,压电常数和介电常数均表现出相同的趋势。1% BT 织构 PSN-PMN-PT 陶瓷的压电常数 d_{33} 为 698 pC/N, 3% BT 织构 PSN-PMN-PT 陶瓷的压电常数(d_{33} 为 816 pC/N)明显高于随机取向的 PSN-PMN-PT 陶瓷(d_{33} 为 402 pC/N)。当模板含量为 4%, 织构陶瓷的压电常数和介电常数均降低。一方面,织构化陶瓷中所有晶粒具有与单晶相似的[001]取向,从而形成与 PMN-PT 单晶类似的各向异性以及电场极化后形成的“4R 工程畴”结构可以提高压电性能和介电性能^[24];另一方面,由于 BaTiO_3 具有较低的介电常数,其作为模板复合在织构陶瓷中导致介电常数下降。因此,沿[001]方向的取向晶粒对压电、介电性能的促进作用和由 BaTiO_3 模板复合而引起的对压电、介电性能的负面影响之间存在竞争,当 BT 含量超过一定量时,这会降低总的压电效应^[25]。当模板含量较低时,织构度随 BT 含量的增加而增加,晶粒取向效应占主导地位,从而



$x\%$ is volume fraction of BaTiO_3 templates. 0.29PSN-0.39PMN-0.32PT is 0.29 $\text{Pb}(\text{Sc}_{1/2}\text{Nb}_{1/2})\text{O}_3\text{-0.39Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-0.32PbTiO}_3$.

图 1 BaTiO_3 模板的 XRD 谱、SEM 照片和 0.29PSN-0.39PMN-0.32PT 陶瓷 XRD 谱

Fig. 1 XRD patterns, SEM image of BaTiO_3 template and XRD patterns of 0.29PSN-0.39PMN-0.32PT ceramics

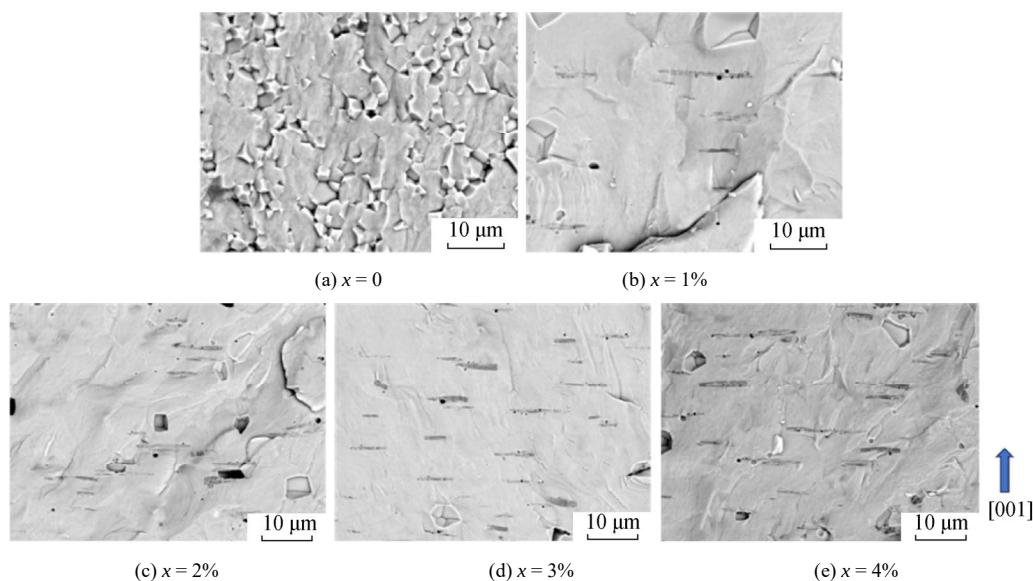
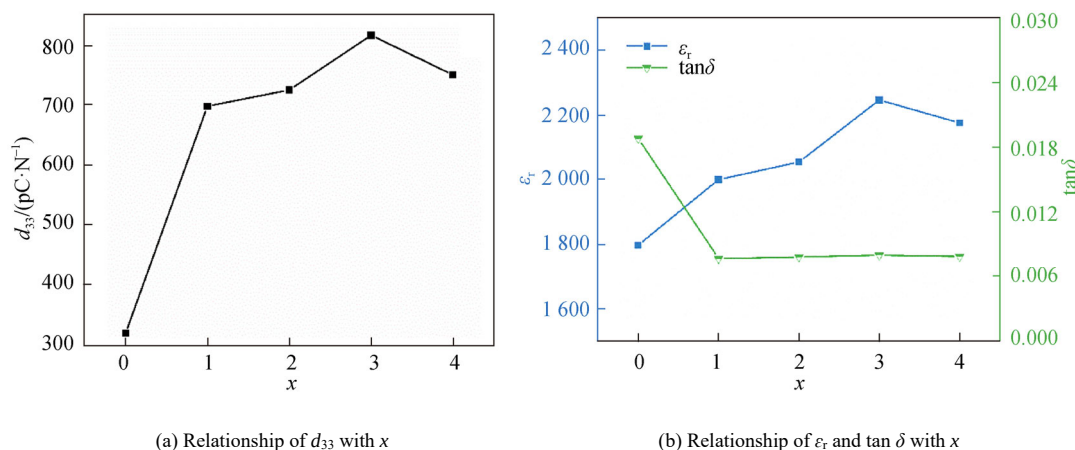


图2 0.29PSN–0.39PMN–0.32PT 陶瓷的 SEM 照片

Fig. 2 SEM images of 0.29PSN–0.39PMN–0.32PT ceramics

(a) Relationship of d_{33} with x (b) Relationship of ϵ_r and $\tan \delta$ with x

d_{33} is piezoelectric constant. ϵ_r is dielectric constant. $\tan \delta$ is dielectric loss.

图3 0.29PSN–0.39PMN–0.32PT 陶瓷的压电和介电性能

Fig. 3 Piezoelectric and dielectric properties of 0.29PSN–0.39PMN–0.32PT ceramics

使压电性能随模板含量的增加而提高。当织构度达到饱和时, 由于 BT 组分的压电和介电常数较低, 引入更多的 BT 模板也会导致总的有效压电、介电常数降低。根据我们的实验结果, 3%BT 是在织构化 PSN–PMN–PT 陶瓷中实现最高 d_{33} 的最佳模板含量。在 3%BT–PSN–PMN–PT 织构陶瓷中, 压电常数和介电常数分别为随机取向陶瓷的 2.03 倍和 1.24 倍。相比于压电常数, 介电常数较低的增长率是因为织构陶瓷中 BT 模板和闭口孔隙的共同作用导致的。通过测试室温下的介电损耗发现, 织构陶瓷的介电损耗降低为 0.008, 比随机取向陶瓷低得多, 并且低于大多数软压电陶瓷(大于 0.020)。介电损耗的

降低主要是由于织构陶瓷晶粒尺寸的增大, 织构陶瓷由于模板的存在, 其晶粒尺寸在 15~20 μm 左右^[26], 比普通陶瓷有较大幅度提升。织构陶瓷的晶粒尺寸增加, 晶界减少, 而晶界是电荷和能量损失的主要来源。

图 4 为随机取向和不同模板含量的织构化 0.29PSN–0.39PMN–0.32PT ($x\%$) 陶瓷介电常数 ϵ_r 和损耗 $\tan \delta$ 对于温度依赖性。由图 4 可见, 在随机取向的陶瓷中观察到对应于三方到四方(T_{r-t})和四方相到顺电立方相(T_c)的 2 个相变。在织构陶瓷中三方到四方相变不太明显, 只观察到明显的四方到立方相的转变, 这表明了织构陶瓷具有较好的温度稳定性,

这将在后面的测试中体现。而三方-四方相变的不明显可能与织构陶瓷四方相含量的增加有关,这是因为 BaTiO_3 在室温到 120°C 处于四方相,因此, BT 模板的存在会使周围基体中形成四方相,而导致陶瓷的三方-四方相变不明显^[19]。通过对比随机取向和织构陶瓷的介电温谱,发现 BT 模板的引入使陶瓷的 T_C 略微降低并且随 BT 模板含量增加而减小,

这与具有低 $T_C \sim 120^\circ\text{C}$ 的 BT 模板有关^[19, 26]以及高温环境下 PSN-PMN-PT 基质中潜在的模板扩散^[19]。尽管模板含量在一定程度上降低了织构陶瓷的 Curie 温度,但与以前报道的二元 PMN-PT 织构陶瓷相比^[27], PSN-PMN-PT 织构陶瓷的 Curie 温度仍具有较高水平。因此,由于其具有更宽的温度使用范围,为实际应用提供了巨大的潜力。

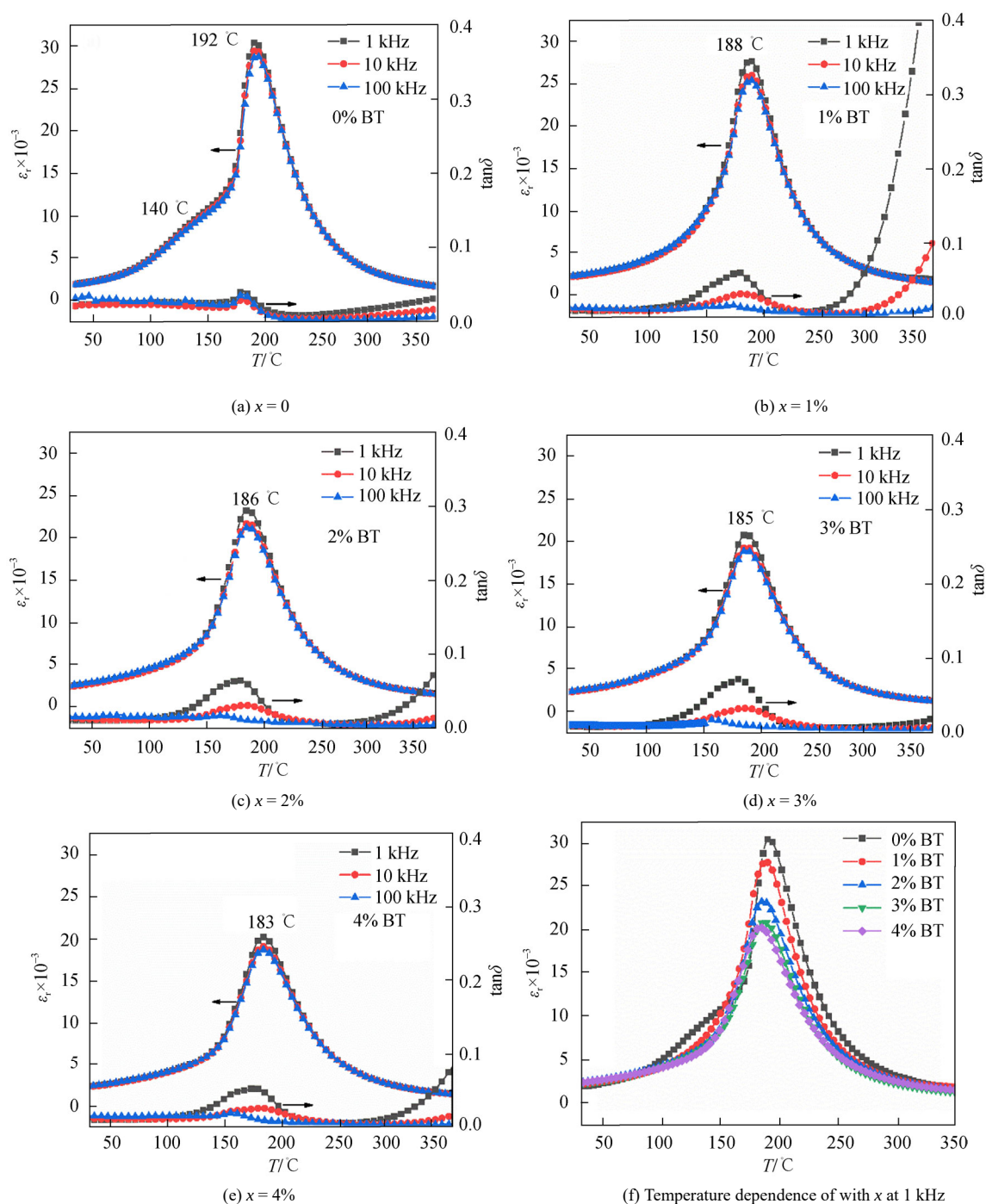


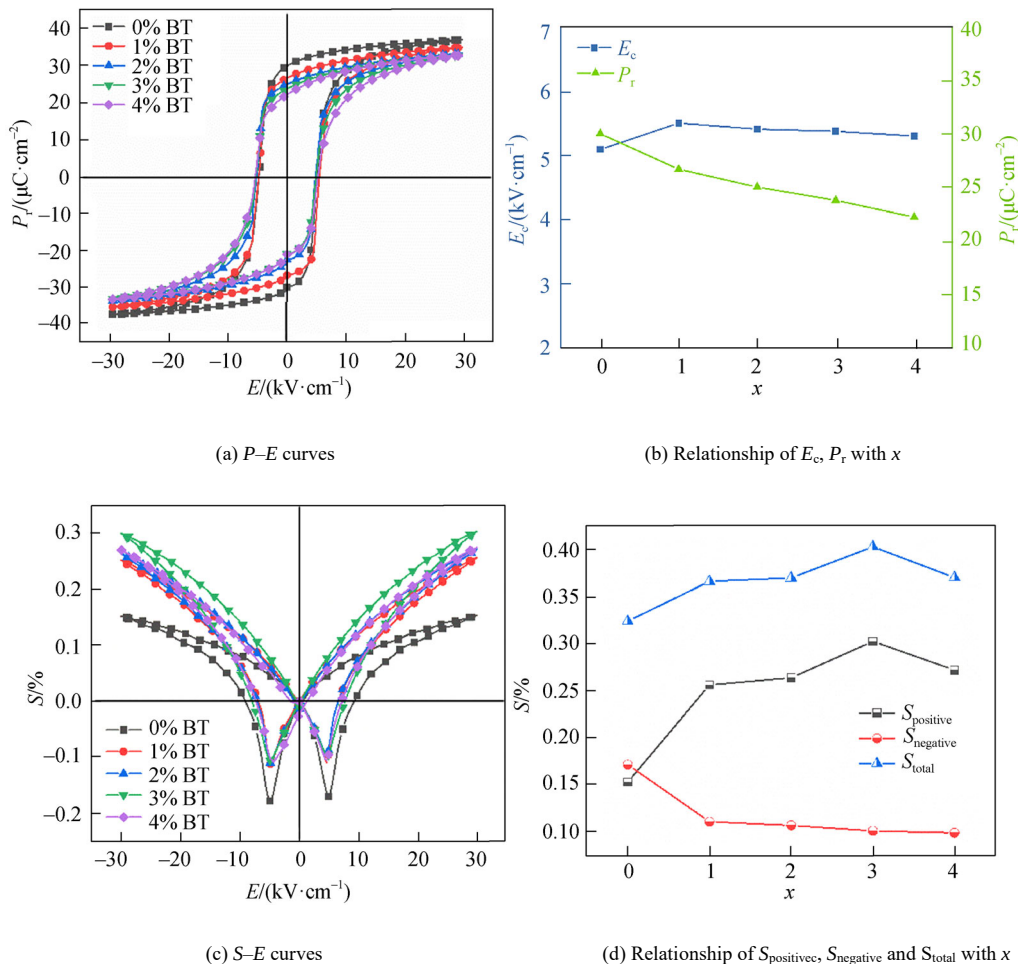
图 4 0.29PSN-0.39PMN-0.32PT 陶瓷的介电温谱

Fig. 4 Temperature dependence of dielectric constant of 0.29PSN-0.39PMN-0.32PT ceramics

2.4 铁电性能分析

图 5 为随机取向和不同模板含量的 0.29PSN–0.39PMN–0.32PT 织构陶瓷的电滞回线和电滞应变。由图 5 可见, 在 30 kV/cm 下的所有陶瓷都具有饱和的电滞回线。随着 BT 模板的增加, 织构陶瓷的 P_r 逐渐减小, E_c 几乎不变。随着 BT 的变化, P_r 从 30.0554 $\mu\text{C}/\text{cm}^2$ ($x = 0$) 减小到 22.2607 $\mu\text{C}/\text{cm}^2$ ($x = 4$)。这种现象归因于以下原因: 1) 对于三方相铁电材料, 其自发极化沿[111]方向, 最低的剩余极化沿[001]方向^[28], 考虑到三维空间中的极化平均, [001]取向的织构陶瓷的剩余极化小于随机取向的陶瓷; 2) BT 模板的剩余极化(12 $\mu\text{C}/\text{cm}^2$) 远远小于 PSN–PMN–PT 陶瓷, 因此, 随着模板含量的增加, 织构陶瓷的剩余极化逐渐减小。随机取向和织构陶瓷的矫顽场 E_c 在 5.1~5.5 kV/cm 之间波动。类似于压电性能和剩余极化, 织构陶瓷的双极 S – E 曲线也

与其沿[001]方向高度取向有关。图 5c 显示了随机取向和不同模板含量的 0.29PSN–0.39PMN–0.32PT 织构陶瓷的双极电滞应变, 对应的正应变 S_{positive} 、负应变 S_{negative} 和总应变 S_{total} 在图 5d 中。织构陶瓷的正应变 S_{positive} 和总应变 S_{total} 相比随机取向的陶瓷明显增大, 这是由于[001]极化的三方相晶体处于“4R 工程畴”, 而[111]极化的三方相晶体处于“1R 单畴”态。畴工程晶体的压电系数比单畴状态的压电系数高得多^[29], 这是由于“极化旋转过程”, 有助于高的线性应变。与随机取向的 0.29PSN–0.39PMN–0.32PT 相比, 织构陶瓷的负应变 S_{negative} 变小。这是因为[001]取向的三方相晶体的负应变是由线性压电引起的, [111]极化的三方相晶体是由非 180°铁弹畴引起的而导致高水平的负应变^[28]。因此, [001]取向的织构陶瓷的负应变 S_{negative} 相比于随机取向的陶瓷减小。



P_r is remanent polarization. E_c is coercive field. S_{positive} is positive strain. S_{negative} is negative strain. S_{total} is total strain.

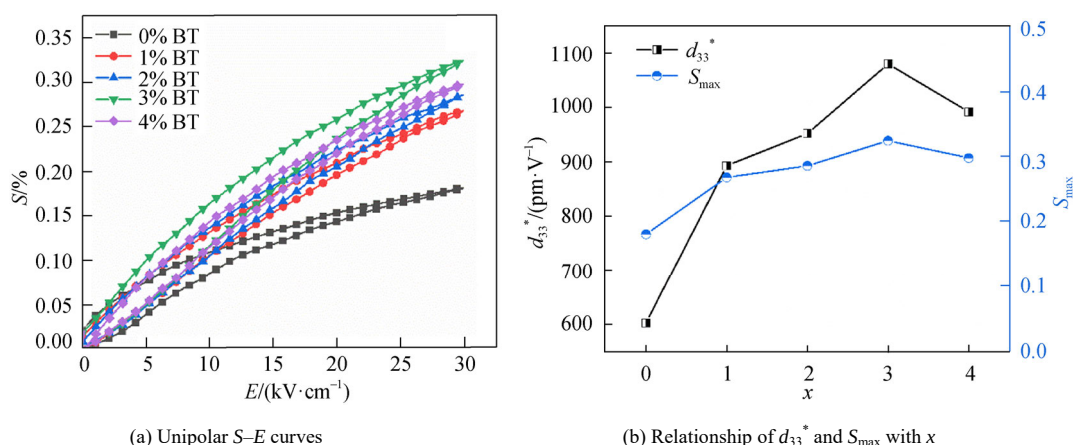
图 5 0.29PSN–0.39PMN–0.32PT 陶瓷的电滞回线和电滞应变
Fig. 5 P – E and S – E curves of 0.29PSN–0.39PMN–0.32PT ceramics

图 6 为随机取向和织构化的 0.29PSN-0.39PMN-0.32PT 陶瓷在 30 kV/cm 电场下的单极应变曲线。由图 6 可见, 在相同的电场作用下, 织构陶瓷的应变明显大于随机陶瓷, 0.29PSN-0.39PMN-0.32PT-3BT 陶瓷表现出最大的应变 0.324%, 这是由于织构化使晶粒沿[001]方向取向所致。所有样品的大信号压电常数 d_{33}^* 通过公式 $d_{33}^* = S_{\max}/E_{\max}$ ($S_{\max}$ 为最大单极应变, $E_{\max}$ 是所施加的电场) 计算以及 d_{33}^* 与组分之间的关系如图 6b 所示。对于随机取向的陶瓷样品, d_{33}^* 在 30 kV/cm 时为 602 pm/V, 而对于织构陶瓷样品, d_{33}^* 取决于 BT 含量。0.29PSN-0.39PMN-0.32PT-3BT 陶瓷在 30 kV/cm 下具有 1 080 pm/V 的 d_{33}^* , 超过了大多数 Pb 基铁电陶瓷^[30-31], 以及一些织构陶瓷, 包括具有含 5% PMN-PT 模板的 PMN-32.5PT 织构陶瓷($d_{33}^* \approx 1\ 000$ pm/V)^[32]。

2.5 热稳定性分析

在压电材料的实际应用中, 热稳定性是衡量材

料性能的重要指标之一。为了更清楚地了解织构化对陶瓷热去极化行为的影响, 图 7 将随机取向和织构陶瓷样品在 30~200 °C 退火 30 min, 然后冷却到室温测量其压电系数 d_{33} 。由图 7a 可见, 随着温度的升高, 随机取向陶瓷的 d_{33} 分别在 140 °C 和 190 °C 表现出明显的下降, 从温谱上可以看出, 这与陶瓷的三方-四方相和四方-立方相相转变相关。织构陶瓷都表现出相似的下降趋势, 在超过 160 °C 时 d_{33} 都出现骤降的趋势, 这是因为温度接近陶瓷的四方-立方相变。在 30~160 °C 之间 0.29PSN-0.39PMN-0.32PT-3BT 陶瓷的 d_{33} 变化只有 12%, 表现出良好的温度稳定性, 这归因于 2 个因素: 平滑的三方-四方相变和 BT 模板在织构陶瓷中产生应力而导致的“夹持效应”。由图 7b 可以看出, 0.29PSN-0.39PMN-0.32PT-3BT 织构陶瓷具有较好的温度稳定性, 极大的促进了压电材料宽温度范围的应用。



d_{33}^* is large-signal piezoelectric constant. $S_{\max}$ is maximum value of field-induced strain.

图 6 0.29PSN-0.39PMN-0.32PT 陶瓷的单极电滞应变曲线
Fig. 6 Unipolar S-E curves of 0.29PSN-0.39PMN-0.32PT ceramics

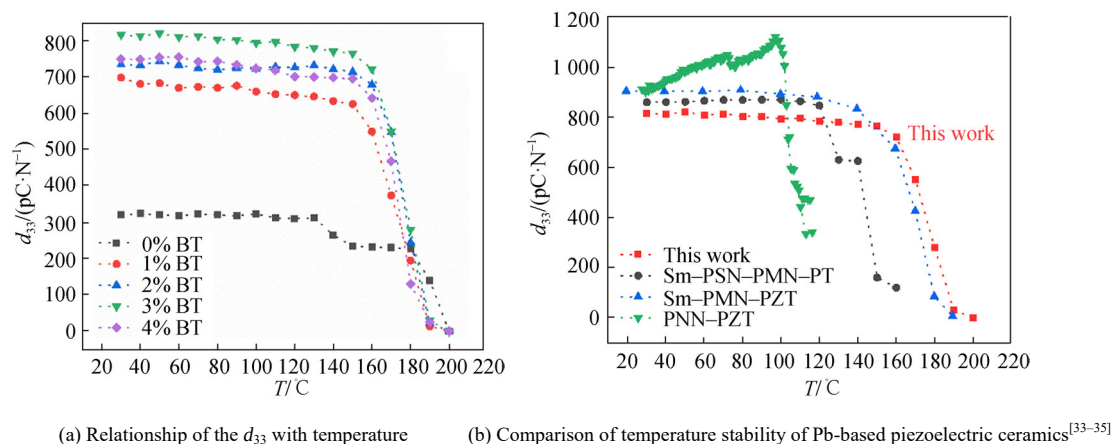


图 7 0.29PSN-0.39PMN-0.32PT-xBT 陶瓷压电系数 d_{33} 的温度稳定性
Fig. 7 Temperature stability of piezoelectric coefficient d_{33} for 0.29PSN-0.39PMN-0.32PT-xBT ceramics

3 结论

通过 TGG 法系统研究了不同 BT 模板含量对 0.29PSN–0.39PMN–0.32PT 织构压电性能、介电性能和铁电性能以及温度稳定性的影响, 探究出 0.29PSN–0.39PMN–0.32PT 基体的最佳 BaTiO₃ 模板含量 3%。合成了织构度 F_{001} 为 98.6% 的 [001] 取向 PSN–PMN–PT (3%) 织构陶瓷, 同时获得了高的压电性能 ($d_{33} = 816 \text{ pC/N}$, $d_{33}^* = 1\,080 \text{ pm/V}$) 和高 Curie 温度 ($T_C = 185 \text{ }^\circ\text{C}$), 这远远优于随机取向的陶瓷, 显著增强的压电响应归因于晶粒取向特征。此外, 与以前报道的其他具有相近压电常数的铅基陶瓷相比, 织构化 PSN–PMN–PT–3BT 陶瓷表现出显著改善的热稳定性。这一工作为开发既具有单晶特性又具有高温稳定性的新型功能陶瓷提供了重要指导, 也极大地拓展了织构陶瓷在新一代高性能、宽温度范围的机电器件中具有广阔的应用前景。

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Electrical Properties and Thermal Stability of $\text{Pb}(\text{Sc}_{1/2}\text{Nb}_{1/2})\text{O}_3\text{--Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{--PbTiO}_3$ Textured Ceramics

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Extended Abstract

Introduction As a core element of the sensor, the piezoelectric constant and dielectric constant of piezoelectric materials determine the sensitivity of the sensor. Piezoelectric materials with a high performance and a high stability are urgently needed. The electrical properties of perovskite ferroelectrics are closely related to the crystal orientation. Growing single crystals is thus an effective way to improve the electrical properties of perovskite ferroelectrics. However, the practical application of single crystals is limited due to their relatively high cost, low trigonal–rhombohedral phase transition temperature, and insufficient composition uniformity in inconsistent melting. To solve the above-mentioned single crystal application problems, the template grain growth (TGG) method for preparing textured ceramics is considered as one of the most promising methods.

In this paper, a PSN–PMN–PT ternary solid solution piezoelectric material was prepared, and the [001] oriented growth of ceramics by TGG method was analyzed. In addition, the influence of template content on the phase structure, micromorphology, electrical properties and temperature stability of textured ceramics was also discussed.

Methods Oxide raw materials were mixed in stoichiometric ratios and pre-sintered at 750 °C for 2 h to form a pure perovskite structure. The [001] oriented BaTiO_3 (BT) microcrystals were added as templates into a uniform slurry with 0.29 $\text{Pb}(\text{Sc}_{1/2}\text{Nb}_{1/2})\text{O}_3\text{--}0.39\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{--}0.32\text{PbTiO}_3$ matrix powder, solvent, dispersant, plasticizer, and binder, and then ground in a ball mill. The BT templates with x of 0, 1%, 2%, 3%, and 4% were added into the slurry. Afterwards, the slurry was cast *via* controlling the height of the scraper and the speed of the substrate. The dried raw embryo tapes were cut, stacked, and kept at 550 °C for 2 h to remove organics. Finally, the samples were sintered at 1270 °C for 9 h.

The phase structure of the random and textured ceramics was determined by X-ray diffraction. The microstructure of the ceramics was characterized by scanning electron microscopy. To measure the electrical properties, the samples were coated with a silver paste and sintered at 550 °C for 40 min to form electrodes and then polarized in silicone oil at 25 kV/cm for 15 min. The piezoelectric coefficient d_{33} was measured using a model ZJ–6A quasi-static d_{33} tester. The temperature dependence of the dielectric constant ϵ_r and dielectric loss $\tan \delta$ was determined by a model E4980A LCR meter. The polarization electric field ($P\text{--}E$) curve and the strain electric field ($S\text{--}E$) curve were analyzed by a model TF Analyzer 2000E ferroelectric test system. For the determination of temperature stability, the samples were annealed at high temperatures for 30 min and then tested for d_{33} at room temperature.

Results and discussion From the XRD analysis, BaTiO_3 as a template can effectively improve the [001] orientation of PSN–PMN–PT ceramics and achieve a high degree of [001] preferred orientation growth. The F_{001} of all the textured ceramics is 97%–99%, among which the F_{001} of 3%(volume fraction) BT–PSN–PMN–PT textured ceramics is 98.6%. From the scanning of the textured ceramics, the template exists stably and is well arranged in the matrix ceramics, which is a fundamental reason why the

textured ceramics have a high texture degree. The piezoelectric constant of 3% BT textured PSN–PMN–PT ceramics (816 pC/N) is greater than that of random PSN–PMN–PT ceramics (402 pC/N). Based on the analysis of the dielectric loss at room temperature, the dielectric loss of the textured ceramics reduces to 0.008, which is lower than that of randomly oriented ceramics and most soft piezoelectric ceramics (*i.e.*, >0.020). The high d_{33} , S_{positive} , S_{total} , and d_{33}^* of textured ceramics are all related to the "4R" domain engineering state. Only a transition from tetragonal to cubic phase occurs in the textured ceramics, indicating that the textured ceramics have a good temperature stability. The results of annealed piezoelectric constant test show that the d_{33} change of 3% BT–PSN–PMN–PT ceramics at 30–160 °C is only 12%, further proving a good temperature stability of textured ceramics.

Conclusions The [001] oriented texture PSN–PMN–PT ceramic with a texture degree F_{001} of 98.6% was synthesized by the TGG method, obtaining the high piezoelectric properties (*i.e.*, $d_{33} = 816 \text{ pC/N}$, $d_{33}^* = 1\,080 \text{ pm/V}$) and high Curie temperature (*i.e.*, $T_C = 185 \text{ °C}$). This enhanced piezoelectric response was attributed to the grain orientation characteristics. Furthermore, the 3% BT–PSN–PMN–PT texture ceramic exhibited an improved temperature stability, compared with previously reported lead–based ceramics with the similar piezoelectric constants. These results showed that the textured PSN–PMN–PT ceramics had high piezoelectric properties and good temperature stability, having the application prospects of textured ceramics in electromechanical devices.

Keywords texture ceramics; $\text{Pb}(\text{Sc}_{1/2}\text{Nb}_{1/2})\text{O}_3$ – $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ – PbTiO_3 –Ferroelectric; piezoelectric properties; thermal stability