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PZT-based Antiferroelectric Material Based on Building Roof Film Rolling Process and Preparation Method Thereof

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Abstract: Antiferroelectric material is a kind of ferroelectric material, which has been widely used in electronic equipment and construction industry. It has always been a research hotspot in the field of material science. In practical applications, the two main factors affecting its energy storage are polarization intensity and breakdown electric field. Due to the large thickness, poor internal consistency and low compression capacity, the energy storage function has not been improved. Therefore, this paper focuses on PZT based antiferroelectric materials based on building roof rolling process. At the same time, the relevant theories and various preparation methods of PZT antiferroelectric materials are introduced in detail. Finally, the solid-state reaction method is used to prepare PZT based antiferroelectric materials. The electrical properties of PZT based antiferroelectric materials were also studied. The experimental results show that the phase transition temperature at 100Hz is close to the theoretical value of 550K, and the test indexes are close to the theoretical value of PZT based antiferroelectric materials.

1. Introduction

1.1. Background

The idea of ferroelectricity was first put forward by Schrodinger, but he believes that there is no ferroelectricity in the world. At that time, some scholars discovered potassium sodium tartrate with ferroelectricity, which opened the door to the understanding of ferroelectric materials. In addition to ferroelectric properties, ferroelectric materials also have many excellent physical properties, such as dielectric properties, piezoelectric properties, acoustic visual effects, thermoelectric properties, photoelectric effects, indirect optical effects and Photorefractive Effects, and are widely used in piezoelectric sensors, optical memories and ferroelectric memories. So far, the annual output value of ferroelectric electronic components in the world has reached tens of billions of dollars. Therefore, due to the rapid development of science and technology, ferroelectric materials have attracted the attention of researchers all over the world.

1.2. Significance

Hysteresis loop is the manifestation of spontaneous polarization in ferroelectric materials, which will change under the action of electric field. In addition, ferroelectric materials also have high dielectric properties, high pyroelectric effect and strong optoelectric effect. With the development of microelectronics and integrated circuits, the requirement of machine shrinkage is higher and higher, and the research of ferroelectric thin films has attracted more and more attention. Compared with bulk materials, ferroelectric films have the characteristics of low density and low dimension, which is a significant trend in the development of ferroelectric materials.

1.3. Related Work

The thickness of oil film directly affects the voltage distribution between oil film and rotating interface. There are some unavoidable factors that will affect the thickness of the imported oil film. Kuo F U introduces a non-stop rotating film model to explore the direction of oil film surface. The small changes of inlet angle, perfect reduction rate, reduction rate, inlet flow thickness and radius rotation are used as input variables, and the conversion of inlet film thickness is used as output variables, which is not the boundary between input and output. The results show that there is a 180 ° phase difference between the inlet oil film thickness and the inlet variables (such as rotation deceleration rate, conversion change and glue free thickness), but there is no phase difference between them. Unstable cylinder end, output phase difference. Rotation angle, vertical rotation radius and surface direction have significant effects on the change rate of nonlinear oil film, but they have not been applied in practice [1]. For ABO_3 perovskite found in PZT based non-ferrous metal applications, cations with different radii A or B will affect the surface level of the material. Liu Y used (Pb13x / 200Lax / 100) $(Zr_{0.75}Sn_{0.16}Ti_{0.09})$ O_3 ceramics (x = 0,2,4,6) instead of Pb^{2+}, and discussed the phase process, dielectric properties and factors affecting the phase transition in PZT based ceramics. All samples were prepared by conventional phase transfer method. The results show that in the rhombic ferroelectric plane with x = 0, the sample changes into a square antiferroelectric element with the increase of La content. With the increase of La content, the front / back field will increase and the lag will decrease. The results also show that an appropriate amount of La can improve the antihistamine stability of PZT based anti sacral ceramics. In addition, with the increase of La content, the released energy density and energy storage capacity reach the maximum value of 3.45 J / cm^3, but the research on stability is not deep enough [2].Ferroelectric thin films are widely used in electronic and low power ferroelectric converters. Shkuratov S I obtained the experimental results of $PbZr_{0.95}Ti_{0.05}O_3$ layer doped with 2% Nb (PZT 95 / 5) polarized Fe film under uniaxial adiabatic compression perpendicular to and opposite to the polarization direction. At a density of 2.4 GPA, thirty-two μm thick film is then transformed in a non-polar anti-corrosion manner,, and a dipole is completely formed under the condition of high voltage double charge. The experimental results show that the current behavior of long press film is more complex than that under the influence of change. This complex behavior may be caused by the stress distribution in a short distance through the film, which is equivalent to the thickness of the front of the voltage wave. It is mainly the result of specific charge released by PZT95 / 5 film at different temperatures, but the experiment is not representative [3]. It is very important to extend the application of metal film to high strength systems. The research on the development of lead-free piezoelectric ceramics by Shrout T r shows that its properties are equivalent to those of iron perovskite based substrates represented by Pb (Zr, Ti) O_3 or PZT. In this work, the scientific and technological effects of these materials are compared with different series of piezoelectric ions "soft" and "hard". Technically, the inherent characteristics of dielectric and piezoelectric properties are related to Curie (TC) temperature and limit phase limit (MPB) life. Similar to PZT, the MPB components in the three part system (Na, Bi) TiO_3-$BaTiO_3$ and (K, Bi) TiO_3 have improved efficiency, but the efficiency provided is significantly reduced. The effect of effective ferrous to ferrous conversion below TC further limits their utility. Although equivalent to TC, the high-level piezoelectric property reported in (K, Na) NbO_3 is the result of polarization improvement related to the partial motion of rectangular square polycrystalline part, and its practicability is not strong at present [4]. Perovskite $PbTiO_3$ (PT) - $PbZrO_3$ (PZ) - Pb ($Fe_{0.5}Nb_{0.5}$) O_3 (PFN) in ternary system has attracted much attention because of its ability to show ferroelectricity, magnetism and ferroelasticity at the same time. The most sensitive element in the external field response may be located near the crystal boundary region (MPB) of Pb ($Zr_{0.53}Ti_{0.47}$) O_3 (PZT) - PFN binary chain. Schiemer J A studied the strength and dynamics of the deformation behavior of pzt-pfn ceramic samples by resonance ultrasonic spectroscopy. It is found that the elasticity before moving the rectangular square axis is not suitable for models based on elastic position distribution or relaxation characteristics, but is determined by the correlation between the acoustic position and the midline of relevant relaxation and / or microstructure exercises. After modification, the elasticity of fatigue factor is about 50%, which corresponds to the expected position of linear / square pressure / parameter command, but the actual operation is more complex [5]. It is an important work to explore the stability of PZT materials. Hu Y was prepared by sol-gel method to prepare rice noodles Pb ($Zr0_{.95}Ti_{0.05}$) O_3 (PZT95/5), and the effect of temperature roasting on the stability of PZT (95/5) perovskite system was studied. According to the results of TGA-DSC, the average working temperature of xerogels was determined to be 550℃~750℃. XRD results show that with the increase of temperature T1, the peak intensity of the first crystal stage gradually increases, while the peak intensity of the unknown phase gradually decreases and finally disappears. Perovskite levels were created for PZT (piezoelectric conversion) (95 / 5) at 750℃. SEM observation showed that with the increase of T1, the accumulated flour gradually became thinner and more uniform. At 750℃, the average particle size of PZT nano powder (95 / 5) is about 100 nm. This study is of great value to this paper [6].

1.4. Innovation

The preparation method used in this paper has the following advantages: 1) the preparation method is simple and has large output, which can be used in construction industry. 2) The thickness of the material of the invention is adjustable, and the minimum material thickness can reach 80 microns. 3) It can be used in many applications based on PZT antiferroelectric materials. 4) Due to the high shear force and extrusion capacity between rollers, the preparation method of this material can significantly increase the density of the material, so as to improve the breakdown resistance and energy storage density.

2. PZT-based Antiferroelectric Material Based on Building Roof Rolling Process and Its Preparation Method

2.1. Relevant Theories of PZT Ferroelectric Materials

Ferroelectric materials are materials that spontaneously polarize in two or more possible directions [7]. Pb(Zr,Ti)O_3 (lead titanate, PZT) is a ferroelectric material with high tensile strength, piezoelectric, dielectric and pyroelectric properties. The hysteresis loop can be used to determine the relationship between the polarization intensity of ferroelectric materials and the external electric field. As shown in Figure 1.

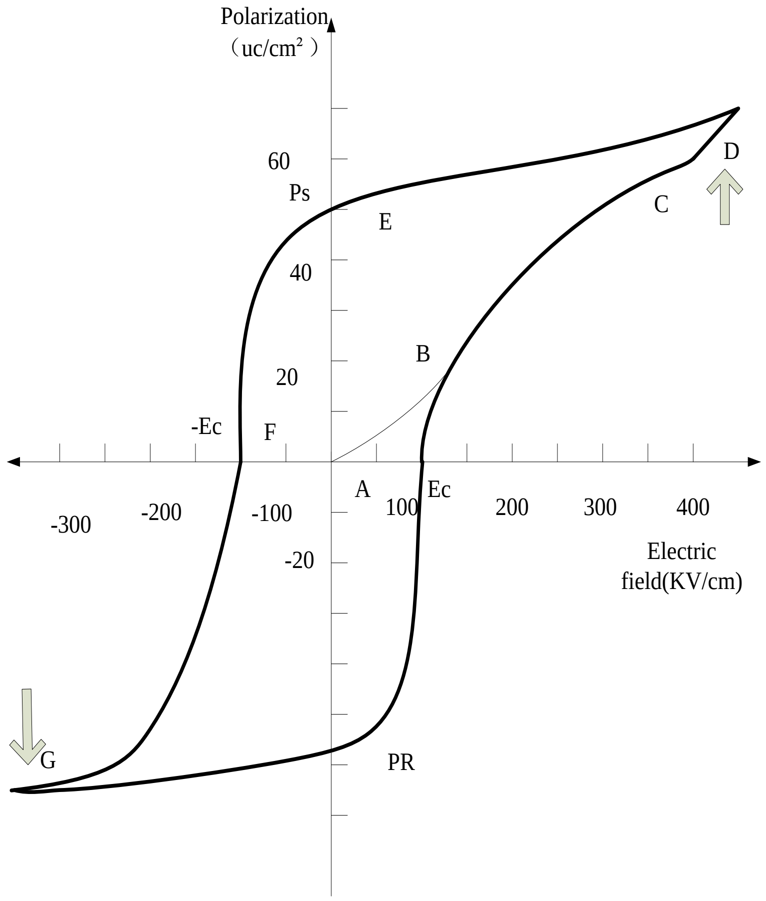

Figure 1. Hysteresis loop of ferroelectrics

When the external electric field is not used, although ferroelectrics have spontaneous polarization, the electric field direction is inconsistent and the external polarization force is strong. When the applied electric field reaches a certain value, the orientation remains unchanged. If the electric field energy continues to increase, the electrons and ions contained in the material will cause the displacement of the polarization line and finally reach the highest point. If the external electric field decreases gradually at the pole, the polarizability will decrease slightly. If you continue to use the reverse electric field until the polarization intensity reaches this point, the polarization intensity is called the residual polarization intensity. When the opposite light field increases and the polarization intensity reaches the opposite direction, the direction of the pole is the same as that of the opposite light field. When the directional electric field decreases again, the change of intensity polarization is similar to the previous process [8].

Ferroelectric material is a kind of dielectric material. The properties of dielectric materials are generally characterized by dielectric spectrum. Dielectric spectrum includes dielectric spectrum and dielectric spectrum. Dielectric temperature spectrum is used to reflect the changes of dielectric constant and loss with frequency and temperature. The dielectric constant measured by dielectric spectrum under alternating electric field is called dynamic dielectric constant. This physical quantity is complex, also known as complex dielectric constant. The magnitude of this physical quantity is related to frequency and temperature, and its functional relationship spectrum with electric field frequency and ambient temperature is called dielectric spectrum [9]. The dielectric spectrum whose frequency is higher than the dielectric spectrum is called the optical frequency dielectric spectrum, which is also called the resonant dielectric spectrum. When the frequency is lower than the dielectric spectrum, it is called the relaxation dielectric spectrum. The dielectric spectrum can well reflect the properties of the dielectric under the electric field. For the dielectric response under the optical frequency, it is also called the resonant response. In this frequency band, the internal polarization of the material is ion and electron polarization, and the electron polarizability is independent of the change of temperature, Ion polarizability has little relationship with temperature change [10]. The main research here is the relaxation dielectric response process. The dielectric relaxation process is usually analyzed by model functions in the form of Debye (semicircle arc), Cole-Cole (ARC), Davison-Cole (skew semicircle arc) [11].

(1) Cole-Cole equation

The complex permittivity of a system with relaxation time distribution can be described by equation. It can be seen from the formula that the equation was equation at that time. The real part and imaginary part of the dielectric constant are expressed as

$$\varepsilon(\theta) = \varepsilon_a + \frac{\Delta\varepsilon\{1+(\theta t)^{1-b}\cos[\pi(1-b)/2]\}}{1+2(\theta t)^{1-b}\cos[\pi(1-b)/2]+(\theta t)^{2(1-b)}} \quad (1)$$

$$\varepsilon''(\theta) = \frac{\Delta\varepsilon\{1+(\theta t)^{1-b}\sin[\pi(1-b)/2]\}}{1+2(\theta t)^{1-b}\cos[\pi(1-b)/2]+(\theta t)^{2(1-b)}} \quad (2)$$

(2) For low molecular materials, Debye equation and Cole-Cole equation are not good to represent the real and imaginary parts of dielectric function on the complex plane. For this, we introduce Davidson-Cole equation[12]. Which can be expressed as::

$$\varepsilon(\theta) = \varepsilon_a + \Delta\varepsilon\cos(c\varphi)\cos^c\varphi \quad (3)$$

$$\varepsilon''(\theta) = \Delta\varepsilon\sin(c\varphi)\cos^c\varphi \quad (4)$$

Among $\varphi = \tan^{-1}(\theta t)$

(3) Havriliak-Negami equation

Havriliak-Negami equation is proposed to explain the dielectric relaxation and mechanical relaxation processes of some polymer materials [13], also known as HN equation, which can be expressed as:

$$\varepsilon(\theta) = \varepsilon_a + h^{-c/2}\Delta\varepsilon\cos(c\varphi) \quad (5)$$

$$\varepsilon''(\theta) = h^{-c/2}\Delta\varepsilon\sin(c\varphi) \quad (6)$$

$$h = [1 + (\theta t)^{(1-b)} \sin(b\pi/2)]^2 + [(\theta t)^{(1-b)} \cos(b\pi/2)]^2 \quad (7)$$

The characteristic of dielectric spectrum is that it can measure a wide range of frequency and temperature, ranging from 10-4Hz to 1012hz, and the temperature range is - 270 ℃ to 1650 ℃ [14]; The measured sample can also be plated with an electrode and directly connected to the impedance analyzer for measurement. Because the added test voltage is not very large, it does not need to consider the change of the physical structure and properties of the material, which is superior to other test methods; At the same time, the measurement of more than 100 frequency points of 480 orders of magnitude can be completed in one hour, which is faster [15].

When the dielectric is in the external electric field, assuming that the external electric field is $E(t) = E_m \cos \theta t$, its point displacement vector will change harmonically, but there will be later phase $\delta(\theta)$, that is:

$$S(t) = S_m \cos[\theta t - \delta(\theta)] \quad (8)$$

That is:

$$S(t) = \varepsilon'(\theta) E_m \cos \theta t + \varepsilon'' E_m \sin \theta t \quad (9)$$

Among

$$\varepsilon'(\theta) = \frac{S_o}{E_o} \cos[\delta(\theta)], \varepsilon'' = \frac{S_o}{E_o} \sin[\delta(\theta)] \quad (10)$$

At this time, the potential shift vector can be decomposed into two components, one is in the same phase with the external electric field, and the other is in the 270^0 phase first by the external electric field, which can be expressed as:

$$S^*(t) = \varepsilon^*(\theta) \bullet E^*(t) \quad (11)$$

The complex permittivity is

$$\varepsilon^*(\theta) = \varepsilon'(\theta) - i\varepsilon''(\theta) \quad (12)$$

The dielectric response of ferroelectrics can be analyzed from two aspects [16]: (1) Dielectric adjustability ;(2) Dielectric loss.

It is an important characteristic of ferroelectrics that the dielectric constant of ferroelectrics changes nonlinearly under the action of external electric field. When the electric field exceeds the ferroelectric coercive field, the dielectric constant was decreased with the increase of the electric field. The typical dielectric constant electric field relationship is shown in Fig. 2. Using this nonlinear dielectric electric field characteristic, the phase velocity of electromagnetic wave is controlled by changing the capacitance of ferroelectrics, so as to meet the related applications such as microwave phase shifter [17]. Dielectric tunability of Ferroelectrics α It can be expressed as follows::

$$\alpha = \frac{\varepsilon_r(0) - \varepsilon_r(E)}{\varepsilon_r(0)} \times 100\% \quad (13)$$

Where $\varepsilon_r(0)$ is the dielectric constant without bias, and $\varepsilon_r(E)$ is the dielectric constant under the action of bias electric field E. In order to produce large dielectric tuning effect in ferroelectrics, it is necessary to exceed the coercivity field of the material, which is fatal for bulk materials, because breakdown is easy to occur under high electric field and it is difficult to get practical application [18]. With the continuous maturity of thin film preparation technology, ferroelectric thin films can withstand high breakdown electric field. The application of ferroelectric thin films to realize dielectric tuning has aroused a wide research upsurge.

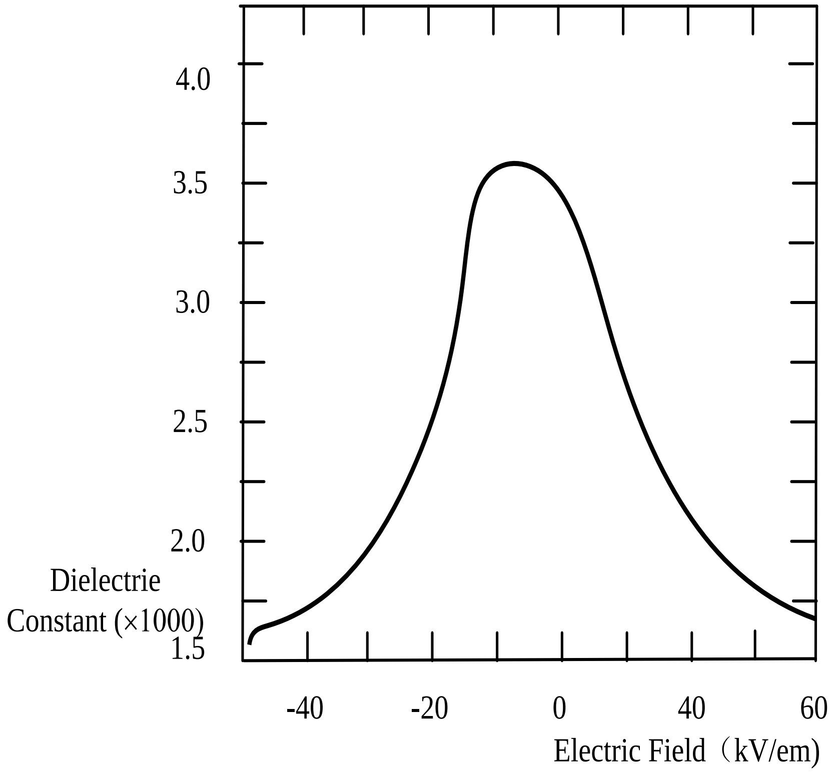

Figure 2. Typical dielectric constant (ε) -electric field (E) of ferroelectrics

Under the action of external electric field, ferroelectric materials not only produce favorable changes such as dielectric constant, but also have certain leakage conductivity and dielectric loss, which are not conducive to the reliability and stability of devices, so they are also an important parameter in the research of ferroelectric thin film materials [19]. The tangent of loss angle can be expressed as:

$$\tan\varphi = \frac{\varepsilon''}{\varepsilon'} \quad (14)$$

Where ε' is the real part of the dielectric constant and ε'' is the imaginary part of the dielectric constant [20]. The loss tangent $\tan\varphi$ reflects the energy consumed by storing a specific charge in the material. Because the ferroelectric domain reversal of ferroelectric materials in ferroelectric phase will bring certain energy loss to the preparation of YIG / PZT based magnetoelectric composites and the study of magnetoelectric effect, it is more inclined to choose dielectric tuning materials in paraelectric phase in practical application [21].

The structure of PZT ferroelectric material is complex. It is a dielectric with ABO_3 perovskite structure. The A position is occupied by Pb^{2+}, and the B position is occupied by Zr^{4+} and Ti^{4+}. PZT is a solid solution of ferroelectric $PbTiO_3$ and antiferroelectric $PbZrO_3$. Near the quasi homomorphic phase boundary (MPA), the performance of PZT will change greatly by changing the chemical composition (ratio of Zr to Ti) or heat treatment [22]. PZT thin films have high dielectric constant and polarization intensity, and are often used to make devices with high storage and high memory function (DRAM and FeRAM).

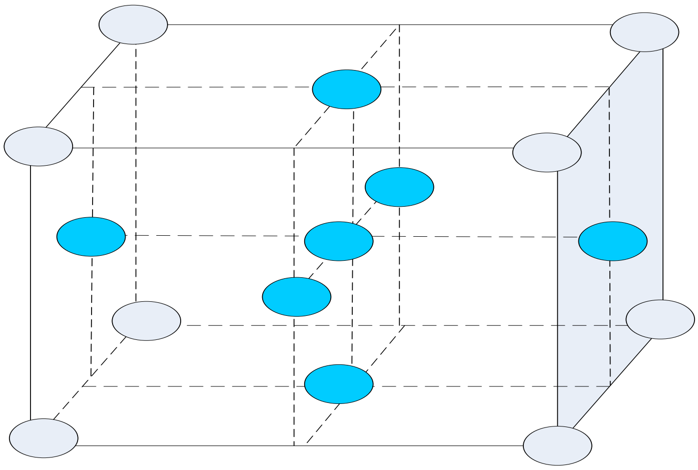

Figure 3. Schematic diagram of perovskite structure

Fig. 3 is a schematic diagram of perovskite structure. A is located at eight vertices, O^{2-} is located at the face center, and B is located at the body center. The structure determines the nature. At high temperature, the positive and negative charge centers of perovskite structure coincide and do not show polarity. However, when the temperature decreases, B ions begin to move towards one of the eight vertices, and the positive and negative charge centers shift, resulting in the generation of spontaneous polarization. Therefore, materials with perovskite structure belong to ferroelectric materials [23].

As shown in Fig. 4, $PbTiO_3$-$PbZrO_3$ is the phase diagram of binary solid solution. It can be seen from Fig. 4 that at room temperature, the content of titanium is greater than 0.47, which is the tetragonal ferroelectric phase ft, and the point group is 4mm; If the content of titanium is greater than 0.05 and less than 0.47, it is triangular ferroelectric phase FR, with a point group of 3m, including high-temperature ferroelectric phase FR (HT) and low-temperature ferroelectric phase FR (LT); If the content of titanium is less than 0.05, it is orthogonal antiferroelectric phase Ao [24].

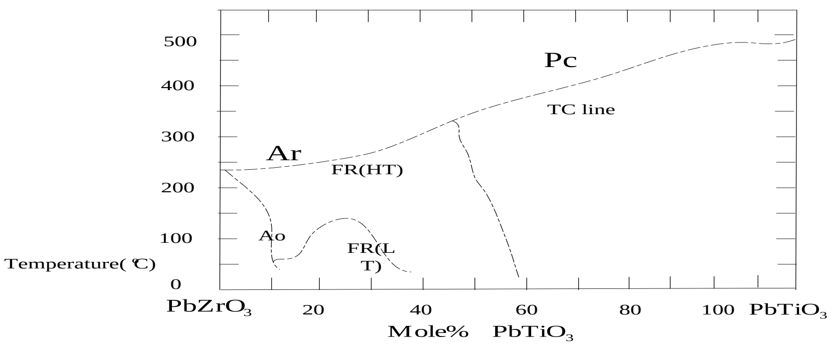

Figure 4. PZT phase diagram

Where: Pc:Paraelectric cubic phase, Ar:Antiferroelectric phase, Ao:Antiferroelectric orthogonal phase, FR: Ferroelectric triangular phase, FT:Ferroelectric tetragonal phase.

You can see a TC line from the figure. TC is called Curie temperature. When the actual value is greater than the Curie temperature, ferroelectrics will behave as paraelectric phase, and ferroelectrics will not have spontaneous polarization. When the real value is less than the Curie temperature, ferroelectrics will not have the ferroelectric effect of piezoelectric effect, and ferroelectrics have spontaneous polarization [25]. The TC line in the figure separates the dielectric part from the ferroelectric phase. It is obvious that the Curie temperature will change with the change of zirconium titanium ratio. Above the phase transition temperature, the change of dielectric constant with temperature follows Curie Weiss law. Close to the change of phase transition temperature, the temperature change method of ferroelectric properties is abnormal at Curie temperature, and the dielectric and dielectric loss are often at the maximum. Below the phase transition temperature, the MPB type is uniformly close to the Zr / Ti ratio Zr / Ti = 52 / 48. Quasi homogeneous boundary is a special case where square and triangular planes exist at the same time. Its dielectric constant is the largest. This is because the crystals in the quasi homogeneous boundary layer are sensitive to square changes and three-dimensional planes. Therefore, it is conducive to the conversion of oxygen ions or titanium ions, so the dielectric conversion rate is the largest [26].

When a good electric field is used, PZT films will produce great deformation and viscosity at the interface due to the good piezoelectric effect. The application of ferroelectric materials is shown in Figure 5 [27].

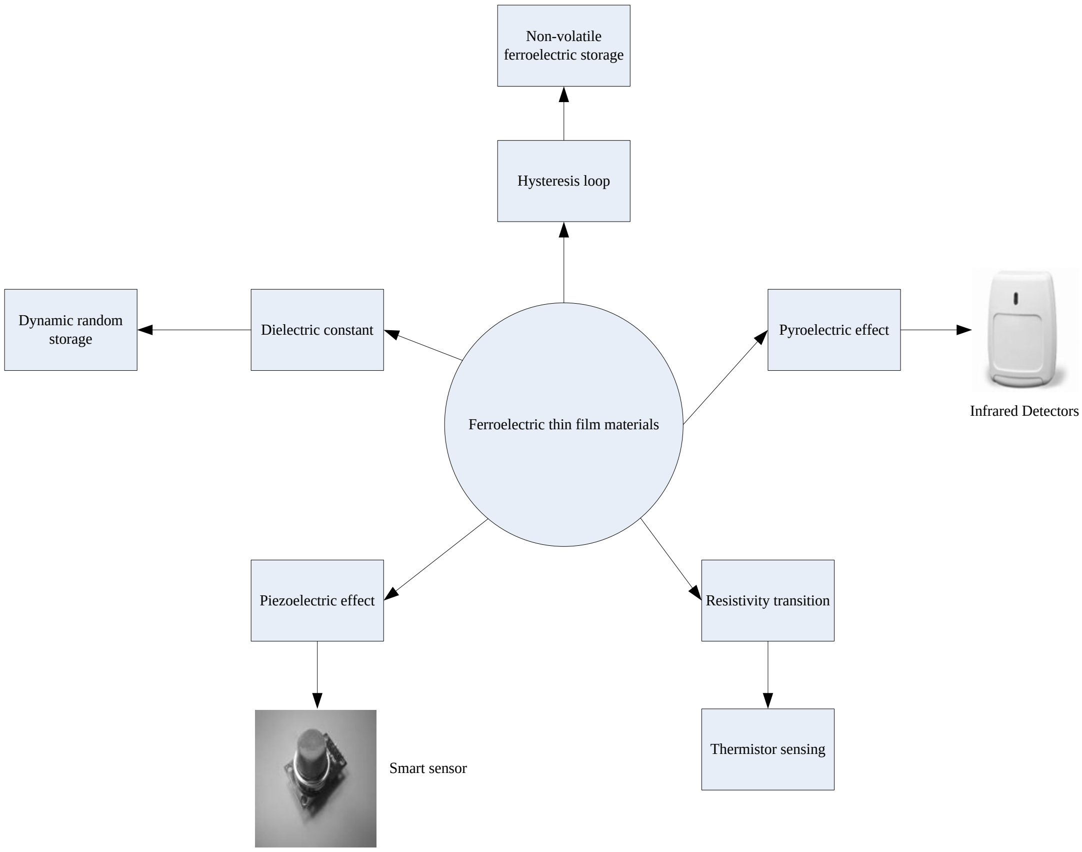

Figure 5. Application of ferroelectric materials

As a representative of tunable ferroelectric elements, ferroelectric phase shifters use the microwave constant transfer characteristics of ferroelectric materials with dielectric constant to achieve the purpose of phase transition. It has other advantages such as low-voltage control power supply and semiconductor technology.

2.2. Preparation Method of PZT Based Antiferroelectric Material

The solid-state reaction method is to prepare samples through high-temperature reaction phase after mixing and grinding crude oxide raw materials. Solid phase method has the advantages of simple process, low cost and leading design industry. But it also has many weaknesses. For example, every part of the raw material begins to react in the form of strong layer. Even after grinding with a sphere, it is difficult to achieve a completely uniform composition. Moreover, the reaction efficiency is low and it is difficult to control the reaction process. The final flour is mixed, not easy to separate, and it is easy to mix with other impurities. PZT piezoelectric ceramic products can generally reach 1200 ℃ when fired, which is very easy to cause the loss of lead composition and change Zr/Ti in PZT. Therefore, some new solid loading methods have been developed, such as mechanical activation method, shock wave high pressure synthesis method, microwave radiation method and so on.

1) Mechanical activation method

Mechanical activation method, also known as high-energy ball milling method or mechanochemical method, is a new type of ultra-fine material made by rolling and grinding high-pressure balls into objects. It is used to initiate chemical reaction or change the structure and properties of materials. The new test method of PZT preparation is to prepare high-density nanocrystalline perovskite PZT powder by mixing oxides in the production process. The stoichiometric alkali with particle size of 10nm-30nm was determined by high-strength ball milling with oxide as raw material. However, the manufacturing process has obvious disadvantages such as long decay time, large pollution mixing elasticity and easy agglomeration of dust, so its application scope is small.

2) Shock wave high pressure synthesis

Shock wave high pressure synthesis is a new experimental method for preparing PZT by using the process characteristics of shock wave absorption technology, such as high pressure, high temperature and short operation time. Piezoelectric ceramic materials are synthesized by shock wave high pressure synthesis method, which have the characteristics of high density, high dielectric frequency, high voltage generator and low dielectric loss. However, shock wave high pressure synthesis has the disadvantages of small volume, high cost and not suitable for mass production.

3) Microwave radiation synthesis

PZT material will produce dielectric loss in electric field. The preparation method using the heat generated in this process to determine the amount is called microwave radiation method. The PZT grinding sheet obtained by this method has better performance, which is reflected in compactness and dielectric coefficient. At the same time, it is a good method for energy conservation and environmental protection to a certain extent.

On the basis of reading a large number of documents, this work adopts the methods of theoretical calculation, field investigation and data classification, combined with the practical application of the proposed building roof rolling process system, in order to improve the technical and economic benefits. The technical path of this paper is shown in Figure 6.

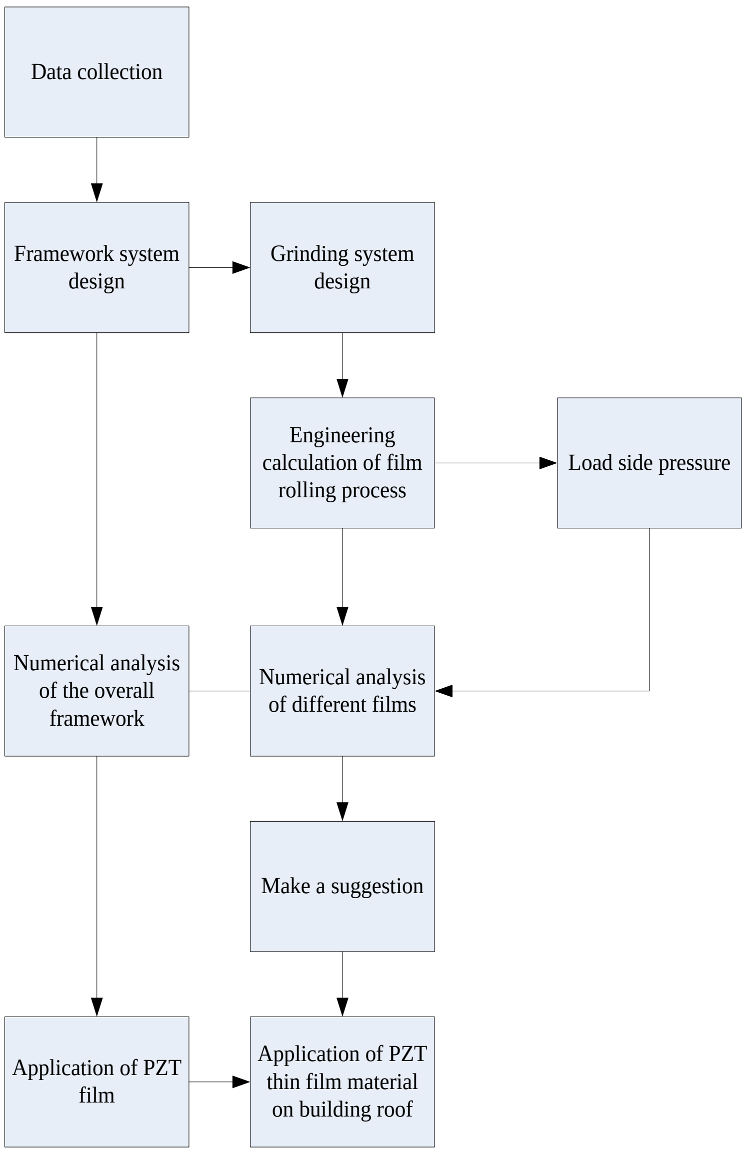

Figure 6. Technology roadmap

3. Preparation of Materials

In order to overcome the weakness in the current research, PZT ferroelectric material based on rolling film technology is suitable for industrial production. It has the characteristics of high tensile strength, high energy storage density, simple operation and low cost. Solid state reaction methods are usually used to combine polycrystalline or solid materials with large crystals and fine synthetic fibers. In this paper, a powerful solid-state method was used to prepare $Pb(Zr_{1-x},Ti_{x})O_3$ substrate. Weigh the raw materials according to the formula, grind, mix, dry, press and mix with balls. After grinding the synthetic powder, add good composite granulation, pre press and cool isostatic pressing to a pressure of 200 MPa. Then select the appropriate temperature for sintering. The process flow is shown in Figure 7, some information of raw materials is shown in Table 1, and some preparation process parameters are shown in Table 2.

The specific steps of the experiment are as follows:

(1) The raw materials such as lanthanum oxide, lead oxide, zirconia, tin oxide and titanium oxide used in the reaction are weighed and ground according to the specified proportion.

(2) The treated powder is washed, ball milled, and then dried.

(3) Mix the treated powder with a mixer, roll it repeatedly in the film roll for 2 hours, and then roll it to the required thickness.

(4) After drying and mixing, put it into a preheated baking oven for mixing. Finally, the antiferroelectric material of PZT was obtained by silver plating on the electrode.

(5) The heat treatment system has a great influence on the structure of the film. When the heating rate is fast, it is easy to make the film crystallize faster, so the film is easier to crack. During heat treatment, because the raw material used is Pb, it is very volatile at high temperature, resulting in loss, so the annealing temperature cannot be too high.

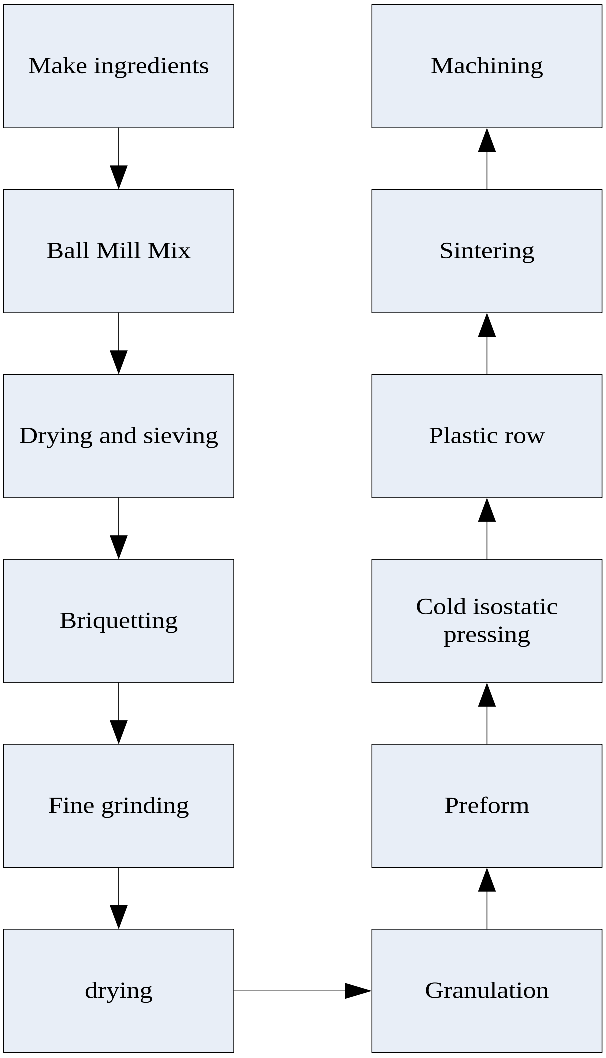

Figure 7. Process flow

Table 1. Raw materials

Raw material	Lead tetroxide	Zirconium dioxide	Titanium dioxide
Chemical expression	ZrO_2	98.3%	Sinopharm Chemical Reagent CO., Ltd
Purity	Pb_3O_4	99.9%	Sinopharm Chemical Reagent Co., Ltd
Manufacturer	TiO_2	99.62%	Sinopharm Chemical Reagent Co., Ltd

Table 2. Partial preparation process parameters

	Temperature(℃)		Time	Heating rate(℃/min)
	Maximum	Minimum	-	-
Drying conditions	100	120	1	-
Pre burning treatment	800	950	2-4	-
Glue discharge treatment	600	700	2-5	1-3
Sintering treatment	1000	1200	2-6	

4. Experimental Analysis of Preparation Method of PZT Based Antiferroelectric Material Based on Building Roof Rolling Process

In the preparation process, the sputtering speed of each part is different, resulting in the composition difference between the film and the bulk material. Therefore, the composition and quality of the bulk material directly affect the composition and quality of the film. The preparation of bulk materials with good stoichiometric composition is the first step to obtain good film materials. In this paper, a rectangular PZT material was designed and prepared by solid-state reaction method (the process is shown in Fig. 7). The clamping length is Φ= 120mm, and then processed to match the sputtering instrument. The standard material size is Φ= 100mm, with a thickness of about 4 ~ 5mm. Then the electrical properties are studied.

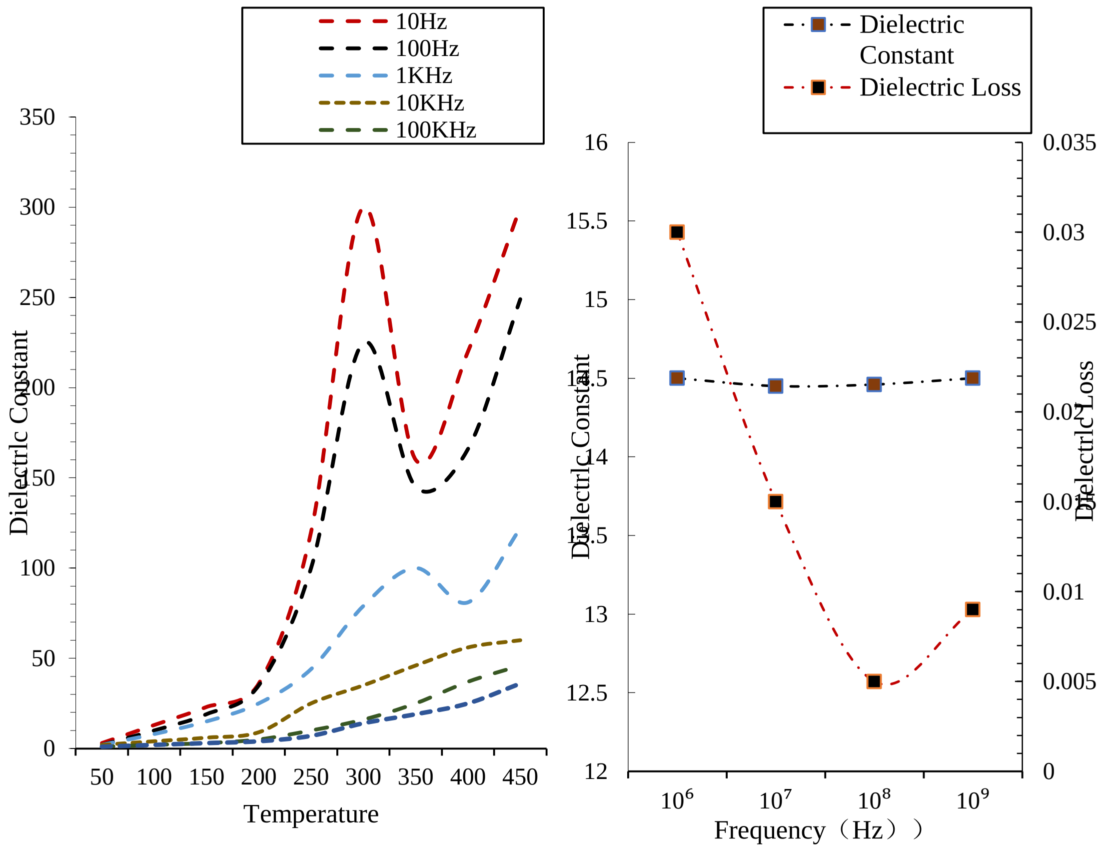

Figure 8. Dielectric temperature spectrum and dielectric spectrum

We can see from Fig. 8 that the dielectric response varies with frequency. The phase change temperature at 100 Hz is very close to the sensor value 550 K. The dielectric constant and dielectric loss at room temperature are very constant between 0.1 and 100 MHz. The relative dielectric constant at room temperature is about 14.5, and the dielectric loss at 20 MHz is 10^{-3} orders of magnitude. The general index of the test is similar to the theoretical estimation of PZT based antiferroelectric materials, which is a necessary condition for the preparation of high-strength PZT ferroelectric materials.

In addition, we can clearly see from the figure that the dielectric frequency increases abnormally at a temperature of about 380 ℃, that is, 380 ℃ is the Curie temperature range of the PZT based antiferroelectric material. Select four different temperatures, take the real part of the dielectric constant as the abscissa and the imaginary part as the ordinate, and draw its Cole Cole diagram, as shown in Figure 9:

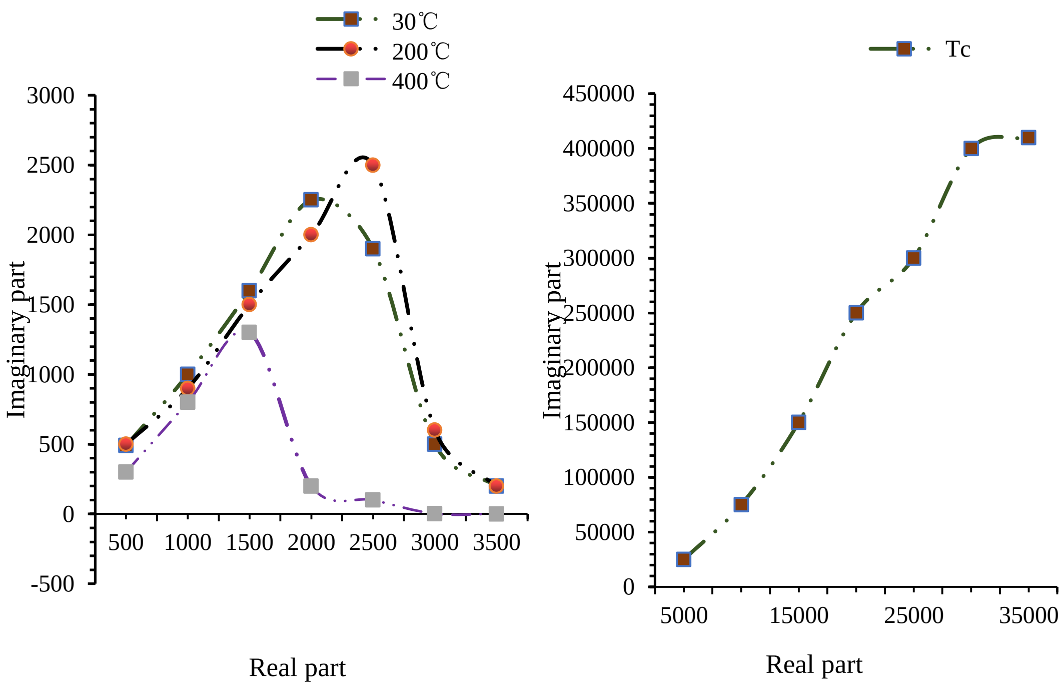

Figure 9. Cole-Cole diagram of samples at different temperatures

According to the classical model of dielectric spectrum, it can be known that all samples are non Debye relaxation except Curie temperature. The curve shape is asymmetric and the relaxation time is unevenly distributed. It can be seen from Fig. 9 that ferroelectric relaxation occurs before and after Curie temperature. At Curie temperature, the size of coordinates is very different from others. At Curie temperature, both dielectric frequency and dielectric loss are abnormal. From the distribution trend of relaxation time, the symmetry at Curie temperature is the worst. Before the Curie temperature, the sample is in the ferroelectric phase, and the ferroelectric relaxation phenomenon is mainly caused by the pole and polarization of the lattice. Under the action of external electric field, the dipole will not only be affected by the energy of electric field, but also prevent viscous vibration. The equilibrium function goes through a period called relaxation time. In terms of polarization, especially in heterogeneous media, there will be a certain amount of cumulative loads in the phase view, which exist in the changing AC field. This process can also be regarded as the process of macro dipole orientation. This polarization process includes applications with irregular phase interfaces and applications with defects and impurities. When the temperature is 400 ℃, the material is at paraelectric level. When the temperature is 400 ℃, the material is in the paraelectric phase. Because the local components in the sample are slightly uneven, the Curie temperature of the sample will also be distributed. Therefore, the paraelectric phase does not appear immediately after the Curie temperature. The reason for material relaxation is the low-energy transition of titanium ions and zirconium ions in different energy related states at B position in perovskite structure. The transition activation energy of charged particles covers a wide energy range, resulting in the phenomenon of dielectric spectrum of non Debye type multi relaxation time distribution.

5. Conclusion

In this paper, a variety of preparation methods for preparing PZT antiferroelectric materials are introduced. Finally, PZT-based antiferroelectric materials suitable for building roofs based on film rolling process are prepared by solid-state reaction method. After sintering, PZT-based antiferroelectric materials prepared by film rolling process have ideal structure in detail, uniform density distribution and sufficient three-phase interface. This work mainly studies the dielectric temperature spectrum and dielectric spectrum of PZT-based dielectric antiferroelectric materials, which provides an important design method for the preparation of PZT-based ferroelectric materials with appropriate properties. However, due to the problem of personal ability and time, there are still many deficiencies, and even many contents have not been deeply studied, which need to be improved. For example, when testing the dielectric temperature spectrum of PZT materials, it is necessary to select several more temperature points, which is conducive to specific analysis.

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Data Availability

Data sharing is not applicable to this article as no new data were created or analysed in this study.

Conflict of Interest

The author states that this article has no conflict of interest.

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