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Research Progress of Piezoelectric Ceramics

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Abstract

Piezoelectric ceramics (PZT) have been widely used in the fields of sensors, transducers, nondestructive testing and communication technology, etc. After more than 50 years of development, the application of piezoelectric ceramics has been widely used in every corner of People's Daily life, and plays an extremely important role in military equipment. All countries in the world attach great importance to the research and development of piezoelectric ceramic materials. The research focuses on discovering new effects and exploring new applications from known materials, as well as finding new piezoelectric materials by controlling the structure and structure of materials. This paper mainly describes the research progress of piezoelectric ceramics.

Keywords

Piezoelectric ceramics; Lead-free piezoelectric ceramics; Ultra-high temperature piezoelectric ceramics; Ferroelectric ceramics.

1. Introduction

Piezoelectric ceramic is an important functional material which can convert mechanical energy and electrical energy into each other. Because of its stable chemical properties, excellent physical properties, easy preparation of various shapes and arbitrary polarization direction of material properties, are widely used in based on the equivalent circuit of the piezoelectric oscillator, filter and sensors, and various types of acoustic, ultrasonic, acoustic transducer, etc., in daily life, industrial production, military and other fields.

With the rapid development of electronic information technology, the miniaturization, functionalization, low cost and high stability of electronic components are required. Research into piezoelectric materials and their applications is also deepening. We expect to obtain piezoelectric ceramic materials with the advantages of good performance, variety, high value-added and less pollution [1].

2. Lead-free Piezoelectric Ceramics

Lead-free piezoelectric ceramics, also known as environmental coordination piezoelectric ceramics, require that the ceramic materials have the highest possible piezoelectric performance and good environmental coordination. Since the 1960s, researchers at home and abroad have started to study lead-free piezoelectric ceramics with perovskite structure mainly composed of niobate and titanate. In recent years, the research, development and application of lead-free piezoelectric ceramics have made substantial progress, which has become one of the hot spots of ferroelectric materials and their applications. And competition for intellectual property rights in lead-free piezoelectric ceramics is fierce.

At present, according to the crystal structure classification, lead-free piezoelectric ceramics mainly have the following three series of research, including tungsten bronze system, bismuth layered structure system and perovskite system. Tungsten bronze is lead-free piezoelectric ceramics, which can be used in electro-optical, photorefractive, pyroelectric and other fields. However, the application of tungsten bronze lead-free piezoelectric ceramics is relatively narrow. Bismuth layered structure is lead-free piezoelectric ceramics with low dielectric constant, high Tc and large anisotropy. Perovskite lead-free piezoelectric ceramics have excellent piezoelectric properties, and the preparation process is compatible with the traditional lead-based piezoelectric ceramics. It is expected to be widely used in information detection, conversion, processing and storage, especially in sensors, drivers, high-power devices and high-temperature devices. Therefore, perovskite lead-free piezoelectric ceramics are the focus of current research[2].

2.1. Perovskite Series Lead Free Piezoelectric Ceramics

Perovskite series of lead-free piezoelectric ceramics includes several subclasses, including lead-free piezoelectric ceramics of barium titanate series ($BaTiO_3$, called BT), lead-free piezoelectric ceramics of alkali metal niobate series ($(K_{1/2}Na_{1/2})NbO_3$, called KNN), and what's more, lead-free piezoelectric ceramics of bismuth sodium titanate series ($(Bi_{0.5}Na_{0.5})TiO_3$, called BNT). Recently, the research of perovskite series lead-free piezoelectric ceramics mainly focuses on the alkali metal niobate series, among which KNN base niobate series is the main one. Lead-free piezoelectric ceramics and bismuth sodium titanate lead-free piezoelectric ceramics, especially the alkali metal niobate series lead-free piezoelectric ceramics, have been studied most deeply.

2.2.1 $BaTiO_3$-base lead-free piezoelectric ceramics

The development of piezoelectric ceramics began with $BaTiO_3$ ceramics. Barium titanate ceramics are lead-free piezoelectric ceramics with high dielectric constant, high mechanical coupling coefficient, medium mechanical quality factor and low dielectric loss. At present, the research on $BaTiO_3$-based piezoelectric ceramics is mainly focused on binary or multi-component ceramic systems based on BT. Some achievements have been made in the research of these systems. For example, $Ba(Ti1\text{-}xZrx)O_3$ piezoelectric ceramics have low sintering temperature, small and dense grain (relative density up to 95%), wide operating temperature range (-30-80°C), and greatly improved piezoelectric performance (d33 up to 340pc/N, k33 up to 65%). However, compared with the existing lead-based piezoelectric ceramic materials, there is still a big gap, and the overall performance is still in urgent need of continuous improvement [3].

2.2.2 BNT-based lead-free piezoelectric ceramics

BNT is a perovskite-type relaxation ferroelectric body replaced by A-position ion complex discovered by Smolenskii et al in 1960. BNT has complex phase transition sequence, strong ferroelectric phase (room temperature residual polarization Pr=38 μC/cm2), large piezoelectric coefficient, large electromechanical coupling coefficient, small dielectric constant and good acoustic performance, and low sintering temperature, sintering belongs to the medium temperature sintering (about 1050°C -1100°C), it is easy to obtain good ceramic sintered body. As a result, it is widely regarded as one of the most attractive lead-free piezoelectric ceramic systems.

BNT has been studied extensively over the years. In the late 1980s, especially in the 1990s, Japanese scholars represented by Tadashi were very active in the research of BNT-based lead-free piezoelectric ceramic system. By introducing various elements and materials such as Pb, Ba, Ca, Sr, Mn, Sc, $NaNbO_3$, etc., the polarization of BNT ceramics has been successfully solved, and lead-free piezoelectric ceramics with better performance have been obtained. The research at home and abroad mainly focuses on three aspects, such as oxide doping modification, solid solution modification and preparation process modification. At the same time, new techniques have been explored to prepare lead-free piezoelectric ceramics with good piezoelectric properties. Grain orientation growth method is the focus of research.

2.2.3 Alkali metal niobate (KNN) series lead-free piezoelectric ceramics

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The alkali metal potassium sodium niobate lead-free piezoelectric ceramics is a solid solution formed by the combination of ferroelectric body $KNbO_3$ and anti-ferroelectric body $NaNbO_3$, denoted as $(K, Na)NbO_3$ (called KNN).

In 2004, Saito et al. reported the breakthrough of KNN series lead-free piezoelectric ceramics in Nature, and the system has been widely studied since then. However, compared with commercial PZT, KNN based lead-free piezoelectric ceramics have significant disadvantages, that is, the phase stability temperature is low, resulting in a narrow range of sintering temperature, and it is difficult to densize them by traditional ceramic preparation process. In addition, because the alkali metal element is easy to volatilize in the sintering process, it is easy to cause the deviation of the stoichiometric ratio and produce heterofacies, thus the performance and stability of ceramics become worse. These factors greatly limit the application of KNN ceramics.

In recent years, scholars at home and abroad have done a lot of research on the sintering process and performance enhancement of KNN based lead-free piezoelectric ceramics, which are summarized in the following three aspects. First, new preparation techniques are adopted, such as texture method, hot pressure sintering method, discharge plasma sintering method, etc. Second, add new elements and ion substitution. For example, new phase structures were constructed in KNN by the addition of tripartite inducers $BaZrO_3$, $CaZrO_3$ and tequel inducers. Meanwhile, the introduction of new components led to the substitution of A and B ions in the perovskite structure A, B and O by introduced ions, which significantly improved the electrical performance of ceramics. Thirdly, add sintering flux to form low eutectic point with KNN. According to the liquid phase sintering mechanism, the liquid phase will be formed in the early stage of sintering to reduce the sintering temperature, and then in the later stage of sintering as a dopant into the lattice to improve the performance of ceramics [4].

3. Ultra-high Temperature Piezoelectric Ceramics

Ultra-high temperature piezoelectric ceramics refer to a class of materials whose Curie temperature is higher than 900°C. At present, the research of high performance and ultra-high Curie temperature piezoelectric ceramic material system is being carried out at home and abroad, and the development of ultra-high Curie temperature (Tc) and stable piezoelectric ceramic material has become one of the research hotspots. The common characteristic of ultra-high temperature piezoelectric ceramics is that the Curie temperature is very high, but the piezoelectric performance is poor (d33 < 20pC/N), and the higher the Curie temperature is, the worse the piezoelectric performance is. Typical ultra-high temperature piezoelectric ceramics mainly include $Sr_2Ni_2O_7$, $La_2Ti_2O_7$, $Ca_2Nb_2O_7$ and other perovskite structures, and $CaBi_2Nb_2O_9$, Bi_3TiNbO_9 and other bismuth layered structures.

$A_2B_2O_7$ perovskite and bismuth-layered ultra-high temperature piezoelectric ceramics have very high cury temperature, but very low piezoelectric performance, which severely limits their application in special high temperature fields such as aerospace, petroleum and geological exploration. Meanwhile, their high pre-firing and sintering temperatures make polarization difficult. Through doping, its dielectric, ferroelectric and piezoelectric properties can be improved, but Curie temperature and operating temperature can be reduced. The influence of doping on its sintering process and polarization process remains to be further studied [5].

4. Conclusion

The application of piezoelectric ceramics, on the one hand, will continue to improve the performance ratio of the practical piezoelectric ceramics products, so as to expand the large market sales. On the other hand, in order to adapt to the development of modern science and technology, new piezoelectric applications are being developed. Among them, the development

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of high quality and high-grade sensors such as voltage and electroacoustic sensitive, force sensitive, thermal sensitive and gas sensitive will still be the focus. In addition, the development of bio-piezoelectric ceramics and piezoelectric intelligent ceramics has a wide range of applications. In a word, the future of piezoelectric ceramics has a bright future. With the improvement of new piezoelectric materials and production technology, piezoelectric ceramics will develop faster and more rapidly.

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