Research on Preparation and Properties of Interfacial Sintering Agent for Piezoelectric Ceramics

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Abstract: The purpose of this paper is to give solutions to the problem that the bonding layer is prone to crack when high power is made as spherical transducers are utilized in broadband applications. In this paper, the technology of low temperature sintering is applied to the integrated preparation of piezoelectric ceramic spheres. After exploration on the suitable low temperature sintering agent, the interface sintering agent system similar to the composition of ceramic spheres is constructed, so that the ceramic spheres become an organic integrity at large. In this study, ceramic powder and three kinds of glass powder with low melting point were used as raw materials to study the influence of ceramic phase composition change on the morphology, electrical and mechanical properties of ceramic matrix at different temperatures. The results indicate that the electrical and mechanical properties of piezoelectric ceramics with bonding layer are improved by the preparation of low temperature sintering agent, and the interface morphology proves with good performance. Compared with the epoxy bonded sample, the piezoelectric coefficient, dielectric constant and three-point bending strength of the optimal composition are increased by 46.13%, 45.75% and 8.37% respectively. The spectrogram of the spherical piezoceramic sample prepared by sintering technology through the admittance/impedance test in air shows that the curve of the sintered PZT piezoceramic spherical shell is smoother than that of the piezoceramic spherical shell bonded by epoxy resin. The resonance peak is more simple, the radial vibration energy is more concentrated, and the electromechanical coupling performance is better, which is beneficial to improve the use power of the spherical transducer. This research will provide conditions for the realization of wide-band and high-power spherical transducer.
***Keywords*:** spherical piezoelectric ceramics, high power, low temperature sintering agent, electrical properties, mechanical properties

1. Introduction

As a treasure house of resources not fully exploited by human beings, the ocean attracts people's attention [1]. Advanced marine instruments and equipment are bound to become an important guarantee for development of marine resources and national strategic security[2]. Therefore, acoustic wave, as the only information carrier capable of transmitting information and energy over long distances in the ocean, determines that sonar has become the most widely used ocean detection system [3]. As the core component of sonar, underwater acoustic transducer, known as "eyes and ears of underwater acoustic equipment", has attracted great attention [4].

Spherical transducer is one of the transducers universally used in the field of underwater acoustics. The piezoelectric ceramic ball is taken as a functional element and its positive and negative piezoelectric effects are utilized to realize the transformation of acoustic and electrical signals[5,6]. Underwater acoustic transducer has become a hotspot in research due to its simple mechanical structure, easiness in production, low cost, easy array and good consistency[7]. However, the maximum radiated sound power of the spherical transducer is characterized with limitations, and its size is determined by the diameter, wall thickness, thickness to diameter ratio and material properties of the piezoelectric ceramic sphere.

Therefore, in order to make it possible in the application in the engineering field, piezoelectric ceramic balls with specific diameter and thickness are frequently used for transmission in a wide frequency band. At this time, the purpose of high power is generally achieved by increasing the voltage at the poles of the ceramic ball. If the voltage at both ends of the transducer is increased unlimitedly, the transducer will be damaged. It is manifested as a serious decline in the radiated sound power of the transducer or a change in the performance index, which is mainly caused by the depolarization or rupture of the piezoelectric ceramics [8]. In addition, even if the degree of depolarization has not been reached, when the mechanical alternating stress of piezoelectric ceramics exceeds a certain value, the material will lead to fracture. Even if the value is lower than this value, the repeated changes of strain will also lead to mechanical fatigue[9], thus affecting the operation of the transducer.

As for the spherical transducer, its working effect is related to the design and manufacture, but the bonding process also plays an important role in the ultrasonic transducer. The bonding effect will directly affect the performance of the spherical transducer, and strive to achieve the optimal interface coupling, improve the extreme value of mechanical fatigue as much as possible, and achieve the purpose of wide band and high power spherical transducer. Therefore, the bonding of spherical transducer is different from ordinary bonding, which requires both good mechanical properties and excellent acoustic transmission performance [10].

However, epoxy resin or organic adhesive is generally adopted in the traditional spherical transducer. Due to the difference between the composition, performance and structure of the organic adhesive and the ceramic substrate, there is not enough adhesive between the adhesive and the adhesive, and the interface coupling is at poor level, and the adhesive layer is subject to damage when working under high power. In order to solve the problem of the piezoelectric ceramic ball interface coupling, this topic decided to low temperature sintering technology is applied to in the preparation of piezoelectric ceramic ball integration build similar to ceramic ball component interface of sintering system, realizes the interface and the structure, mechanical and electrical properties of ceramic ball, ceramic ball as an organic whole.

At the same time, the low temperature co-firing ceramic technology is introduced into the piezoelectric ceramic sintering, can effectively reduce the sintering temperature of piezoelectric ceramic, reduce the volatilization of PbO in lead-based piezoelectric ceramic, avoid ceramic components stoichiometric ratio offset, in guarantee good performance of the piezoelectric material original reduce pollution to the environment on the basis of [11,12]. Therefore, it will be the primary task to explore the suitable composition system of low temperature interfacial sintering agent.

Low temperature co-firing ceramic technology is made in the ceramic base so as to add glass powder as a firing aid, an appropriate amount of organic solvent as a dispersant, the use of glass powder low fusing, sintering glass softening, viscosity decline, so as to reduce the sintering temperature, to achieve sintering at 900℃ technology. To low temperature sintering ceramics accurate thickness and density of ribbons,laser drilling can be used, micro hole grouting materials, precision conductor size printing technology can also be adopted to produce need circuit graphics, they will be more passive component embedded in the multilayer ceramic substrate, and then the overlying together, both inside and outside the electrode can be achieved with the use of silver, copper, gold and other metals, respectively to make a high density circuit which fails to interfere with each other in three-dimensional space.Three-dimensional circuit board with built-in passive components can also be made, and IC and active devices can be affixed on its surface to make functional modules with passive/active integration [13]. It has been widely used in base station, automotive electronics, Bluetooth, aerospace and other aspects [14-15]. In this paper, it is applied to the interfacial coupling of spherical piezoelectric ceramics for the first time. The traditional bonding process is transformed to sintering process with better electrical and mechanical properties, and the broadband and high power performance of spherical transducer is improved.

2. Experiment

2.1 Materials: ceramic and low-temperature glass powder

PZT ceramic powder is determined as functional ceramic phase.Three kinds of low melting point glass powder were used as combustion aid respectively and terpineol as dispersant to form low temperature sintering agent. The matrix of the flake piezoelectric ceramics was PZT-43 purchased from Zibo Yuhai Electronics Co., LTD. The electrical properties of the piezoelectric ceramics with sintered Ag electrode interface were tested with a piezoelectric coefficient of 286 pF/N and a relative dielectric coefficient of 1361.Two pairs of PZT piezoelectric ceramic hemispheres (hemispheres 1,2 and 3,4) with an outer diameter of Ø=25.7mm and a wall thickness of t=2 mm were selected for the spherical piezoelectric ceramic sample matrix. Each pair of two piezoelectric ceramic hemispheres had opposite polarity of internal and external electrodes, and the hemispheres were half-opened along the wall with a size of Ø=4mm, which were used for wire thread and mounting holes.

2.2 Process: preparation of sintering agent and sample preparation test

First of all, different levels of PZT ceramic powder were added into different types of glass powder combustion AIDS on the basis of temperature exploration, mixed right amount terpineol as flux to the viscous state (relative maximum viscosity), the lower ceramic substrate evenly daub low temperature sintering agent, bonding with the upper ceramic substrate, 2 h at room temperature, dry, Then it was placed in muffle furnace and heated to 700℃ at a rate of 5℃/min [17] under a certain pressure, kept for 2 h, and sintered with the cooling of the furnace. The sintered piezoelectric ceramics were polarized at 120℃ for 30 min at a voltage of 3.0 kV /mm. After polarization, the piezoelectric coefficient and dielectric constant of the piezoelectric ceramics were tested. The electrical properties of the sintered piezoelectric ceramics were compared with those of the original ceramic matrix samples and epoxy bonded samples without secondary sintering.

Then according to the national standard (GB/T 17671-1999), the ceramics-bending mechanical properties of the low-temperature sintering agent of the above components are tested. The cross beam rate of the testing machine is 0.5 mm/min, the sample size is 3*4*45 mm, and the Angle of bevelling is 45°. No less than 10 samples in each group were compared with the original ceramic matrix samples and epoxy bonded samples without secondary sintering. The process flow is shown in Figure 1.

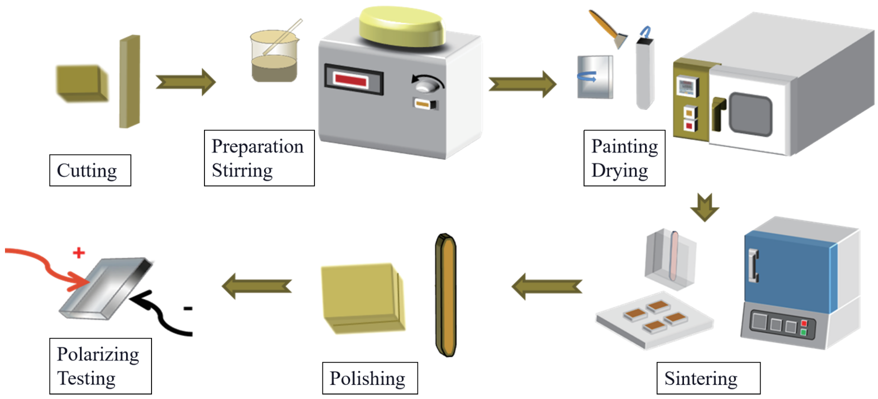

Figure 1. Flow chart of experimental process

Finally, PZT ceramic powder, low melting point mixed glass powder and terpineol were mixed in proportion to prepare low temperature co-sintering agent of piezoelectric ceramics. It was applied to the bond of the upper and lower hemispheres of piezoelectric ceramics and dried at room temperature for 4 hours. Then the dried PZT spheres were sintered at low temperature at 750℃ for 2 hours. After both inside and outside the piezoelectric ceramic ball across the electrodes on the surface of welding line in series and lead wire should be cleaned, through the anode lead to sintering is made after the high voltage polarization in the piezoelectric ceramic ball, comparison can be made in epoxy adhesive of piezoelectric ceramic ball performance comparison. The piezoelectric ceramic ball sample is shown in Figure 2.

Figure 2. Piezoelectric ceramic ball sample (Sintered spherical shell sample on the left; glued spherical shell sample on the right)

The softening temperature of low melting point glass powder was measured by DSC scanning calorimeter. The microstructure of the sample was observed by SEM scanning electron microscope. The piezoelectric constant (d33) and relative dielectric constant (Ɛr) of the electrical sample and the equivalent impedance (Z)/admittance (G)/impedance (R) of the sphere were measured by impedance analyzer. The three-point flexural strength (σf) of the specimens was measured using an electronic universal testing machine.

3. Results and discussion

3.1 Effects of temperature and phase composition on properties

Low temperature sintering process can reduce the sintering temperature most effectively by adding sintering additives. Generally, in piezoelectric ceramic system, sintering temperature can be reduced by adding sintering additives such as low melting point glass. Low melting glass has both properties of glass and low melting point. The sintering temperature can be reduced by forming solid solution, forming liquid phase and transition liquid phase sintering. So the temperature parameters of low melting point glass powder are very important. The sintering temperature is closely related to the softening temperature, and the size of glass powder has an important influence on the softening temperature, and then affects the sintering temperature. The softening temperature of the glass powder with large particle size is higher, and the sintering temperature is also increased, while the softening temperature and sintering temperature of the glass powder with small particle size are decreased synchronously. As shown in Figure 3, PZT powder and 1, 2 and 3 glass powder with average particle sizes of 2.286μm 2.369μm, 2.490μm and 2.170μm were obtained by ball milling process. Compared with before ball milling, particle size of powder is significantly reduced, particle size of powder is more concentrated, shape is more consistent, and powder does not contact each other before dispersant mixing, and no agglomeration phenomenon occurs.

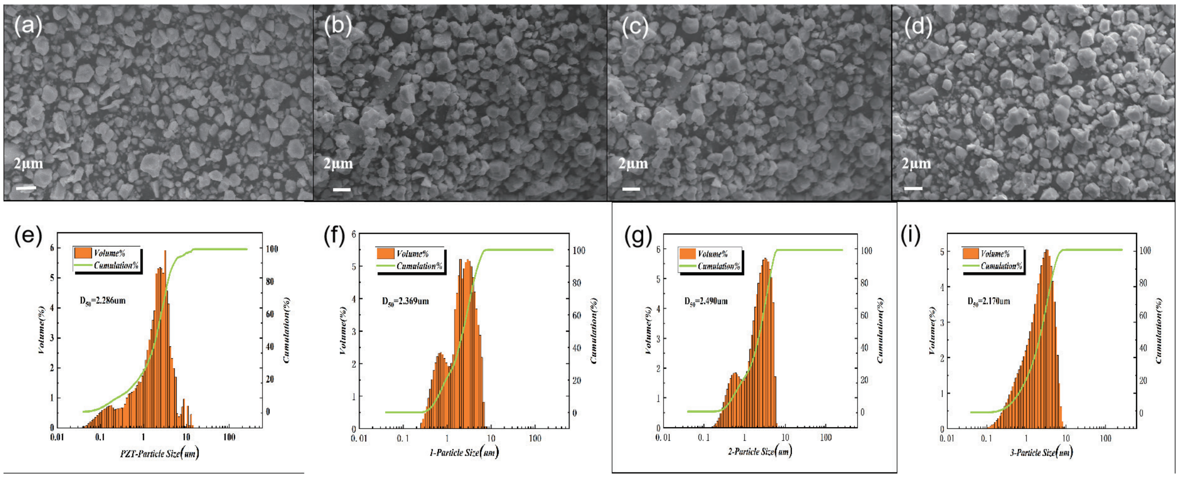

Figure 3. Powder morphology and particle size

Note: (a), (b), (c) and (d) are PZT and 1, 2, 3 respectively. Model glass powder morphology.
(e), (f), (g) and (i) are PZT and 1, 2, 3 respectively. Model glass powder particle size diagram.

That of low-melting point glass powder of three types is shown in Table 1, the softening temperature Tg of type 1, 2 and 3 low melting point glass powder manufacturer is 580,570,560 respectively. The DSC test curve of the ball-milled glass powder is shown in Figure 4. It can be seen from the figure that the softening temperature Tg of low melting point glass powder of type 1 and 2 and 3 decreased significantly, being 477,499 and 491 respectively. This fully indicates that the reduction of powder size after ball milling effectively reduces the softening temperature of glass powder, which means that the sintering temperature can be reduced [18].

Table 1. Performance index of low melting point glass powder

No.	SiO_2 (wt%)	B_2O_3 (wt%)	Na_2O (wt%)	ZnO (wt%)	ZrO_2 (wt%)	coefficient of thermal expansion (30-400℃ /℃ $^{-1}$)	Softening temperature (℃)	Melt flow temperature (℃)
1	27	30	5	19	17	$6.34*10^{-6}$	580	750
2	40	18	10	22	10	$7.02*10^{-6}$	570	720
3	45	15	6	24	10	$7.13*10^{-6}$	560	720

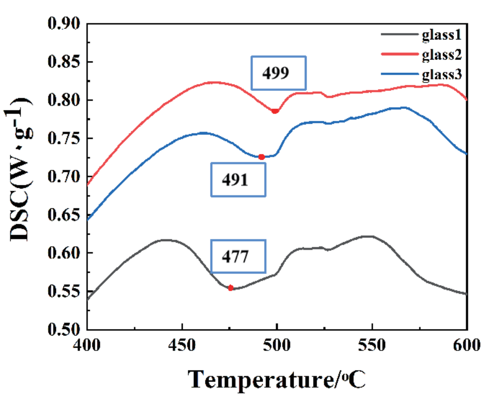

Figure 4. Sample DSC test curve

In order to meet the conditions for the formation of solid solution, liquid phase and transition liquid phase, we set the sintering temperature at 550℃, 600℃, 650℃, 700℃ and 750℃ for preliminary test exploration by referring to the temperature parameters of 640℃, 650℃, 650℃ and 650℃ of low-melting glass powder given by the manufacturer model 1, 2, 3. Type 1 glass powder with the highest softening temperature was selected for single-side sintering, and the sintered sample is shown in Figure 5. The samples obtained good sintering results at 700℃ and 750℃. The glass layer presents translucent and transparent state, uniform distribution, consistent thickness, no bubble generation.

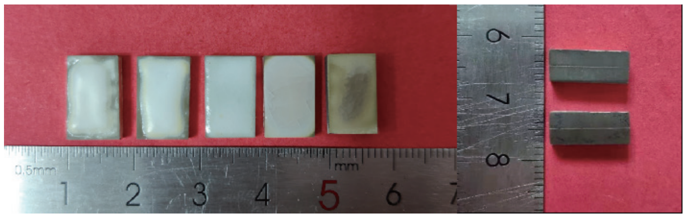

Figure 5. Single-side sintering sample of glass powder

In low temperature sintering process,and in addition to adding low-temperature glass and dispersant, obtaining powder materials that are easy to sinter is also a key link, and making powder suitable for sintering is an important content of current exploration and research[16]. Based on the successful preparation of PZT ceramic powder and the exploration of sintering temperature, the sample sintering interface morphology obtained by changing the addition amount of PZT ceramic powder at the sintering temperature of 700 is shown in Figure 6. Among them, the low temperature sintering system composed of type 1 glass powder with the addition of 20% showed good effect of sintering, while the samples sintered with the low temperature sintering system composed of type 1 glass powder with the addition of 60% showed poor binding ability and all cracked. The results showed that as the content of PZT ceramic powder increased to 60%, the density of sintering interface decreased significantly, the defects were obvious, and the bonding effect could not meet the requirements. Therefore, in order to ensure the mechanical properties and further improve the binding ability, this paper decided to reduce the content of PZT ceramic powder below 60% for further experimental exploration.

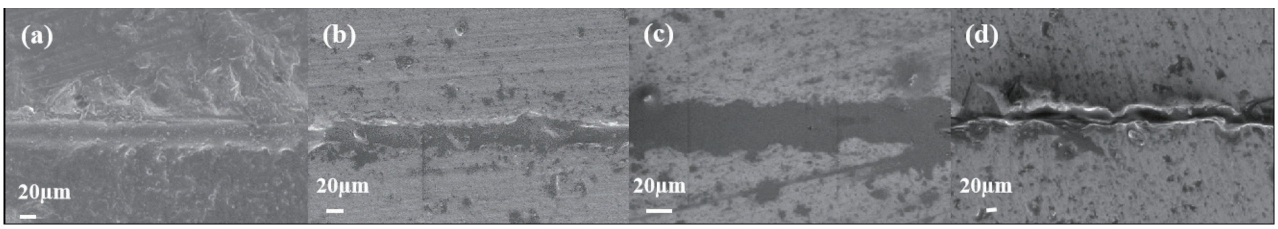

Figure 6. SEM diagram of the sintering interface of different components of 1-PZT

Note: (a), (b), (c) and (d) are the interface morphologies of low-temperature sintering agent prepared by adding 0%, 20%, 40% and 60% PZT ceramic powder and type 1 glass powder, respectively.

The performance index of ceramic material is shown in Table 2, the electrical properties of piezoelectric ceramic matrix with sintered Ag electrode interface were tested. The piezoelectric coefficient was 286 pF/N, and the relative dielectric coefficient was 1361; the thermal expansion coefficient measured in the temperature range of 30-400 is 6.54* 10-6-1, and the difference between the thermal expansion coefficient and the three types of low melting point glass powder is 0.20*10-6,0.48*10-6,0.59*10-6. At the same time, the electrical properties of PZT ceramic powder were tested. The piezoelectric coefficient was 620 pF/N, and the relative dielectric coefficient was 3200.The results show that the electrical properties of PZT ceramic powder are better than that of PZT piezoelectric ceramic matrix, which provides a prerequisite for the effective improvement of electrical properties of low temperature sintering interface.

Table 2. Performance index of ceramic materials

Materials	Piezoelectric coefficients ($d33/10^{-12}$m/v)	Relative dielectric constant (Ɛr)	The Curie temperature (TC/℃)	Coefficient of thermal expansion (30-400℃ /$℃^{-1}$)
PZT ceramic powder	620	3200	230	-
PZTPiezoelectric ceramic matrix	309	1361	320	$6.54*10^{-6}$

The low-temperature sintering agent formed at 700℃ has the appearance of transparent glass. At this time, the low melting point of glass phase and the properties of glass can be reflected, so it can be used as sintering temperature. The addition amount of PZT powder was 0%, 10%, 20%, 30%, 40% and 50%, respectively. The powder was added to model 1 glass powder to make low-temperature sintering agent, and X-ray diffraction analysis was conducted. As the silicon content is less than 40%, the glass system does not produce obvious phase separation[19]. It can be seen from Figure 7 that PZT powder after secondary sintering in low-temperature sintering agent still has good crystal phase structure. X-ray diffraction was used to determine its spectra, and the diffraction peaks of the pure perovskite PZT from (100), (101), (111), (200), (210), (211) and (022) surfaces (PDF #33-0784) could be reflected by changing the addition amount of PZT powder. The intensity of diffraction peak increases step by step with the increase of PZT content. The results show that the prepared PZT ceramic powder is pure phase, and there is no change of crystal phase after secondary sintering. It can be used as functional ceramic phase in low temperature interfacial sintering agent. Therefore, in order to achieve the interface coupling between the sintering layer and the piezoelectric ceramic sphere and to match and improve the electrical and mechanical properties, it is particularly critical to select PZT powder with good electrical properties when constructing the interface binder system similar to the composition of the ceramic sphere (P4/P5).

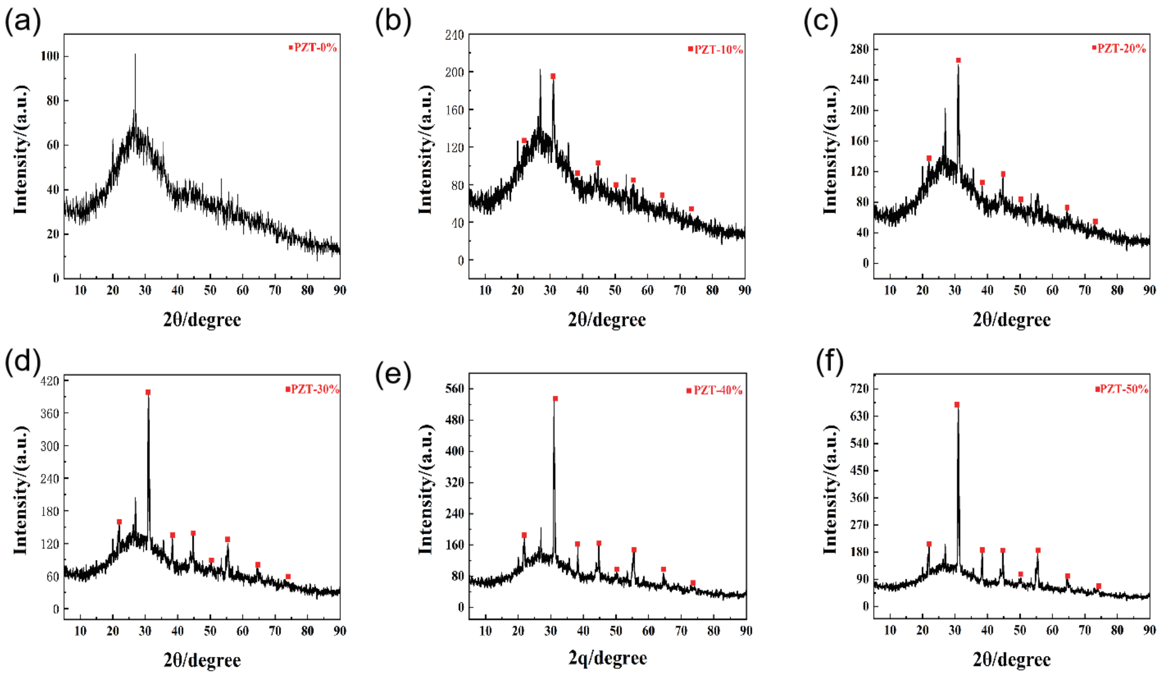

Figure 7. XRD pattern of 1-PZT low temperature sinter

Note: (a), (b), (c), (d), (e), (f) respectively for PZT ceramic powder adding quantity 0%, 10%, 20%, 30%, 40%, 50% and type 1 glass powder XRD for preparation of low temperature sintering agent

3.2 Influence of different ceramic phase components on electrical properties at 700℃ and 750℃

Test results of sintered samples obtained by changing sintering temperature are shown in Figure 8. Compared with the test results of epoxy bonding sample shown in Figure 9, the piezoelectric ceramics sintered under 700 samples with 20% PZT ceramic powder and the low temperature sintering system composed of type 2 glass powder showed good electrical properties, which is selected as component I. The piezoelectric coefficient is 177 pF/N,which is 57.10% of pure PZT ceramic, 29.04% higher than that of epoxy bonded sample, and the relative dielectric coefficient is 909, which is 67.04% of pure PZT ceramic, 48.51 %higher than that of epoxy bonded sample. The piezoelectric sheets sintered at 750℃ in a low temperature sintering system consisting of type 1 glass powder with 10% doping showed good electrical properties. The piezoelectric coefficient was 223 pF/N, which was 74.19 % of pure PZT ceramics. Compared with the epoxy bonded sample, the relative dielectric coefficient is 872, 64.28% of pure PZT ceramic, 45.75% higher than the epoxy bonded sample, which is selected as component II. The electrical properties of the bonding interface of piezoelectric ceramics can be significantly improved by preparing the appropriate low temperature sintering system, which makes up for the poor electrical properties of traditional adhesives.

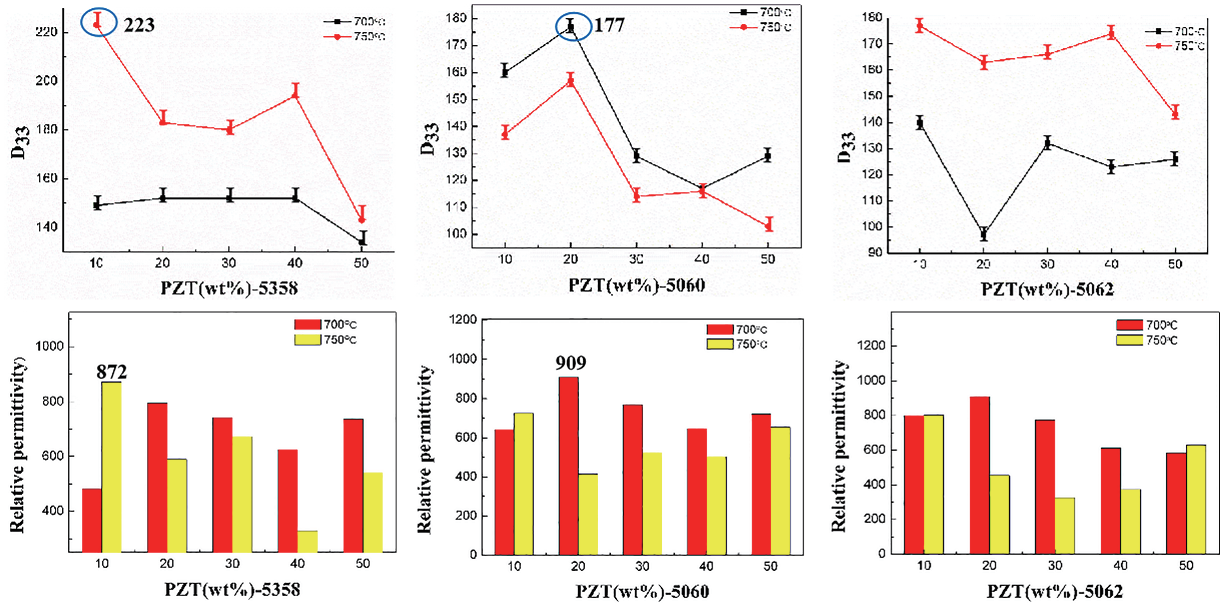

Figure 8. Piezoelectric coefficient and dielectric constant of sintered sample

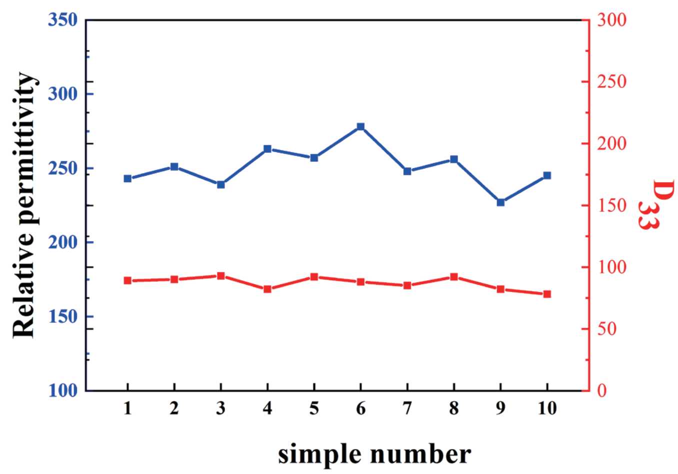

Figure 9. Piezoelectric coefficient and dielectric constant of the bonded sample

Figure 10 shows the morphology analysis of the sintering interface and epoxy bonding interface of the two components. The interface morphology shows that the sintering interface has better compactness than the bonding interface, and there is no porosity. At the interface of sintering, the diffusion of molten liquid is uniform, and the bonding interface has obvious

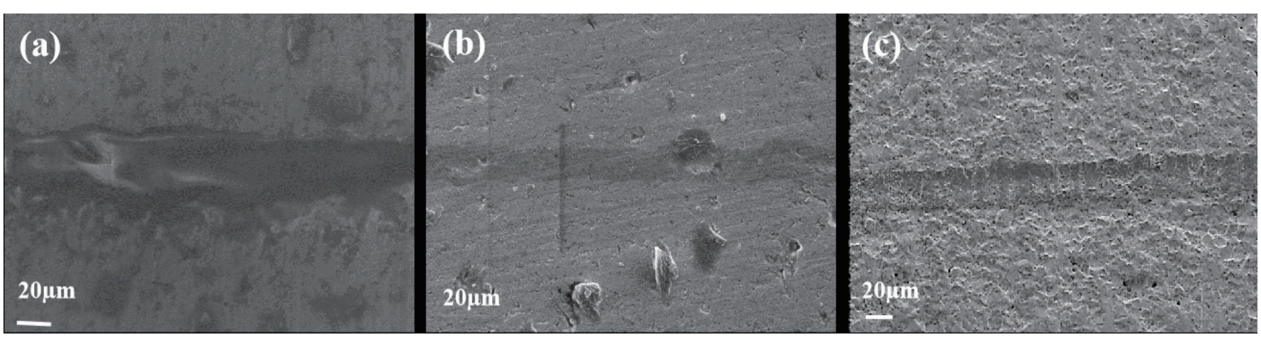

Figure 10. SEM images of the sintering interface and epoxy bonding interface of the two components

Note: (a) (b) (c) are the sintering interface of component 1 sintering agent, the sintering interface of component 2 sintering agent, and the epoxy bonding interface, respectively.

boundary. At the interface of sintering, the diffusion of molten liquid is uniform, and the bonding interface has obvious boundary. By controlling the size and thickness of the ceramic slice and the coating method, the bonding interface with uniform thickness of about 40 μm is obtained. And shows the component analysis diagram of the sintering interface of the two components. It shows that the glass powder component (manufacturer's reference value) in the sintering interface uniformly exists at all points and the proportion of each component is almost the same, which fully indicates that the sinter is evenly distributed in the interface of the ceramic matrix, and each component is evenly dispersed and fully mixed. The zirconium content of the three low melting point glass powders is greater than or equal to 10, belongs to crystallization glass, and has better toughness and strength.

3.3 Influence of different sinter components on mechanical properties

The mechanical properties of component I and component II sintered samples and the original ceramic matrix samples and epoxy bonded samples without secondary sintering were tested. The test results are shown in Figure 11. The results show that the average flexural strength of the original ceramic sample is 84.50 MPa, the average flexural strength of the sintered sample is 36.76 MPa, and 60% of the samples fracture in the sintered layer, and the mechanical properties are poor, far lower than the flexural strength of the original ceramic sample. The average flexural strength of the sintered samples of the second component is 74.89 MPa, and the ceramic itself has internal fracture, sintering layer does not crack, and has good flexural strength. Compared with the original ceramic sample, its flexural strength is slightly reduced, which is caused by the abnormal growth of individual grains and the increase of defects caused by the second sintering. The average bending strength of epoxy bonded samples is 67.82 MPa, and 70% of the samples have fracture in the bonding layer. The mechanical properties are between component I and component II, and the mechanical properties are average.

Figure 12 shows the Weibull analysis of mechanical properties of samples. β is a shape factor used to characterize the dispersion of flexural strength. The larger the β value is, the narrower the distribution of flexural strength is, and the better the consistency of mechanical properties of materials. It can be seen from the figure that β is greater than 6, with good consistency. The coefficient value of component II is the largest and the fitting is the best. Based on the above analysis, the sinter system prepared by component II has the best mechanical properties. This is based on the fact that the PZT matrix and the model 2 low-melting glass powder have the highest matching degree of thermal expansion coefficient, and the strain difference is small in the process of rising and cooling, so as to reduce internal stress and prevent cracking.

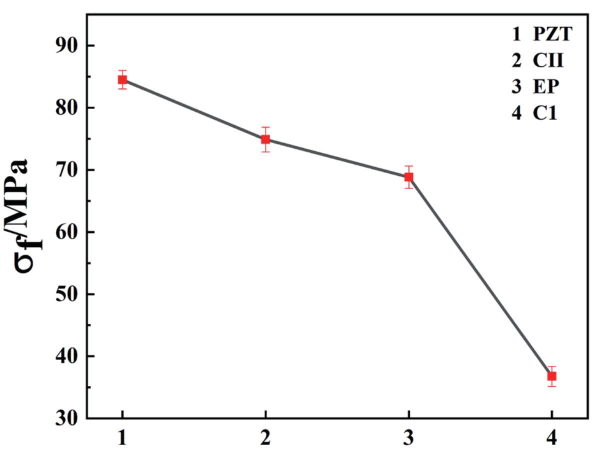

Figure 11. Three point bending strength

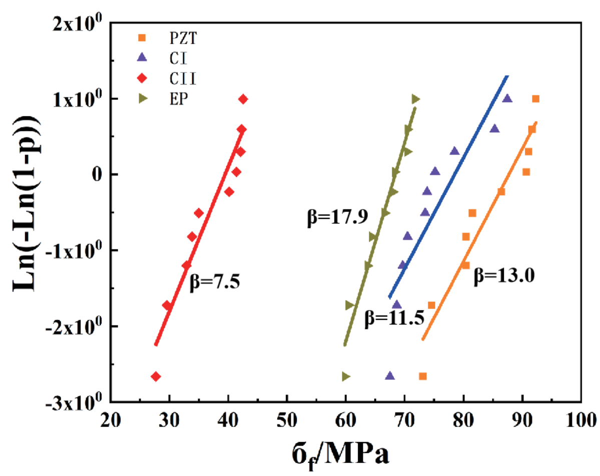

Figure 12. Weibull analysis

3.4 Influence of sintering agent on interfacial coupling performance of spherical piezoelectric ceramics

Through a series of experiments, component two was selected as the best sintering agent system to determine the best sintering process. Both the sintered sphere and the bonded sphere ensure that the wall thickness error of the hemispherical shell is less than 0.05mm. The performance of the piezoelectric ceramic spherical shell combined with these two hemispherical shells has good symmetry in three dimensional space. The electrical properties of sintered PZT piezoelectric ceramic ball shell bonded with epoxy resin in air are tested, as shown in Figure 13. The resonant frequency of the sintered PZT ball is 68.75khz, and the antiresonant frequency is 85.25khz. Compared with the bonded PZT ball, both resonant frequency and antiresonant frequency move to high frequency. In addition, the radiation impedance of the piezoelectric ceramic ball prepared by sintering is greatly improved compared with the ceramic ball prepared by bonding, indicating that the amount of sound energy generated by the system has been substantially improved . The comparison also shows that the PZT PZT ball shell sintered is smoother and flatter than the PZT ball shell bonded with epoxy resin, the resonance peak is more simple, the radial vibration energy is more concentrated, and the electromechanical coupling performance is better.In conclusion, the piezoelectric ceramic spheres prepared by sintering have better electrical properties,which is beneficial to improve the transmission power of the spherical transducer. It also makes it possible to work at higher frequencies [20].

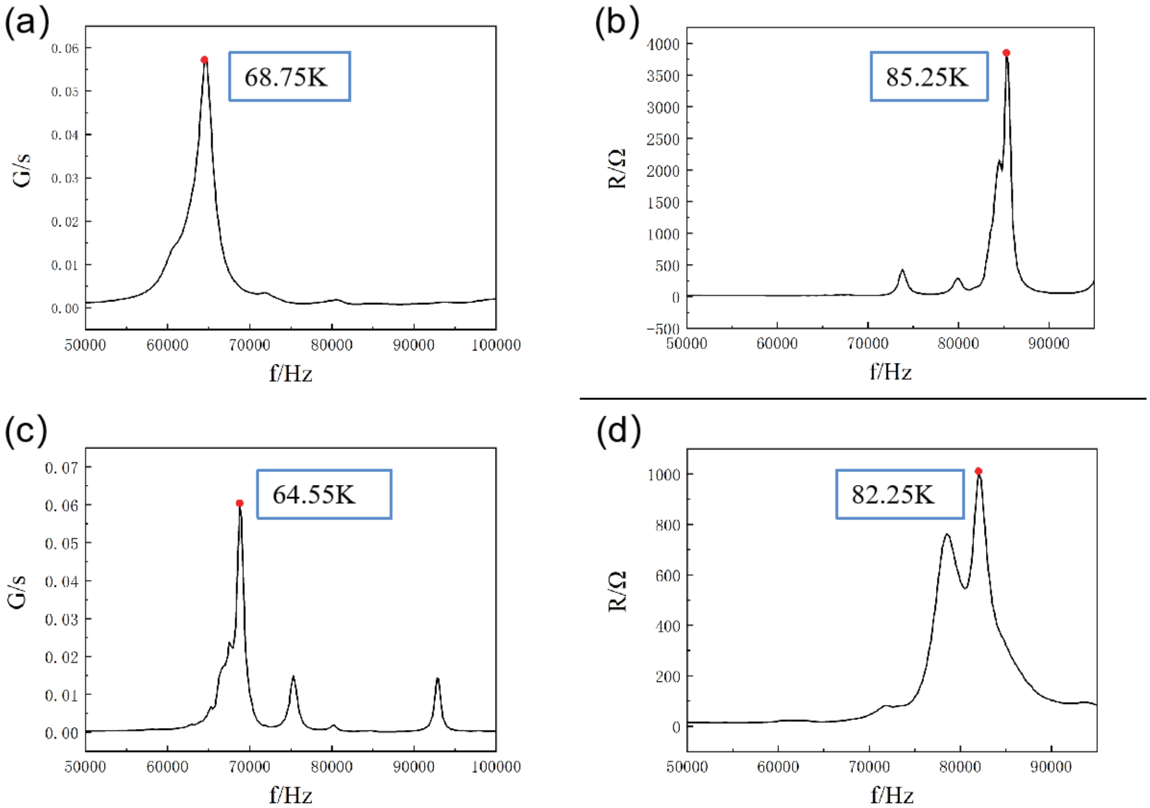

Figure 13. Equivalent impedance/admittance/impedance curve (a, b are the conductivity/resistance diagrams of sintered ceramic spheres; c, d are the conductivity/resistance diagrams of epoxy bonded ceramic ball)

4. Conclusion

In the existing technology, for the piezoelectric ceramic spherical shell with less than the outer diameter, the whole spherical shell is sintered at 1300-1350 high temperature. However, due to the high temperature sintering deformation under the action of gravity, the wall thickness of the shell is not uniform, especially for the piezoelectric ceramic shell with an outer diameter of 35mm or more. With the increase of outer diameter, the sintering deformation also increases, so that the inhomogeneity of shell wall thickness becomes more serious.Therefore, for piezoelectric ceramic spherical shells with an outer diameter greater than 35mm and a wall thickness unevenness less than 0.05mm, the whole shell high-temperature sintering method can not be used[21]. In this paper, by exploring the influence of different parameters such as composition and temperature of low temperature sintering agent system on properties, the mechanical properties, piezoelectric properties and dielectric properties were tested at the interface to determine the appropriate sintering system. In other words, a low temperature sintering system consisting of 10% PZT ceramic powder and type 1 glass powder was selected to be sintered at 750℃, which was applied to the integrated preparation of spherical piezoelectric ceramics to further verify its sintering performance, and a spherical transducer sample with excellent performance was obtained.

Proceeding from the perspective of improving the bonding process of spherical piezoelectric ceramic and in order to make up for the previous single organic binder slash is insufficient, low temperature sintering technology was used to improve the electrical and mechanical properties of piezoelectric ceramics so as to better match the hemispheres and sintering interfaces. As a result, the goal of high frequency power can be realized and the demand of spherical transducer with higher performance can be satisfied.

Acknowledgments

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