I O U R N A L O F

Ceramic Processing Research

# Low loss dielectric ceramics for microwave applications: a review

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Microwave dielectric ceramics have been used for a wide range of applications from wireless communication (including mobile communication, ultra high speed local area networks, intelligent transport system, satellite communication) to consumer electronic products. The need for portable, light weight and multifunctional electronic components has forced scientists to search for new advanced microwave materials. During the past 25 years, a large number of dielectric materials have been developed and the properties of existing materials have been improved. However, the data on these very useful materials are scattered. The main purpose of this review is to bring together the data of low loss microwave dielectric ceramics and classify them according to their applications. This should be of immense help to researchers and technologists all over the world.

Key words: Dielectric, Ceramics, Permittivity, Quality factor, Temperature coefficient of resonant frequency.

### Introduction

The early microwave systems, consisted of bulk metallic cavities, were huge and could not be integrated with microwave integrated circuits (MICs). Microwave devices were traditionally machined from metal, and coaxial RF connections were provided with connectors generally leading to expensive, heavy and bulky packages [1, 2]. These systems cannot meet the market demand for portable and low cost modules. However, electronic circuits for telecommunications, entertainment electronics and the automobile industry have to handle a steadily increasing amount of functions occupying as tiny a space as possible. Dielectric ceramics have replaced bulky metallic cavities in most microwave applications for reasons of cost, dimension, mass, stability, efficiency, ruggedness and ease of use [3]. Also, the temperature coefficient of the resonant frequency can be engineered to a desired value to meet circuit designer's requirements.

Wireless communication has been a fast growing industry in recent years. Various applications include wireless FAX, cellular phones, global position satellite (GPS), military radar systems, intelligent transport system (ITS) and direct broadcast satellites. Dielectric ceramics are being used to make a variety of components such as resonators, filters and oscillators for these systems. Although a large numbers of dielectric materials have been developed, it has been difficult to have all the properties needed for practical applications at various operating frequencies in a single material at a reasonable cost. The advantages of these materials are that they are relatively cheap compared with some of the compounds currently used and in the future they can be improved even further by suitable additives and by optimizing the preparation conditions.

The main purpose of this paper is to gather together the research carried out, classify dielectric ceramics according to their application and discuss the performances of different low loss dielectric ceramic materials. Research work in this area is considerable and therefore it is difficult to present an exhaustive description of the subject in a single review. This review gives briefly the dielectric properties of some well known ceramics reported by various researchers.

### Important characteristics of dielectric ceramics

The dielectric properties of a ceramic material determine its functionality. These properties include relative permittivity or dielectric constant ( $\varepsilon_r$ ), quality factor Q which is the inverse of the dielectric loss (tan $\delta$ ) and the temperature coefficient of the resonant frequency ( $\tau_f$ ).

### Relative permittivity $\varepsilon_r$

The relative permittivity is an important parameter deciding the application of the ceramic. It should be high (20-100) for miniaturization of mobile equipment. The permittivity is related to the resonant frequency (f<sub>0</sub>) of the dielectric resonator by the relation [3]:  $f_0 \approx c/(\lambda_d \epsilon_r^{1/2}) \approx c/(D\epsilon_r^{1/2})$ , where c is the speed of the light in a vacuum and  $\lambda_d$  is the wavelength of the standing wave along the diameter (D) of the resonator. Consequently, if the permittivity is increased, the size of the resonator may be decreased while still maintaining a specific resonant frequency, i.e. larger permittivities enable miniaturization.

Low relative permittivity materials in the range of 4-12 are used for millimetrewave communications and also as substrates for microwave integrated circuits. High speed signal propagation with minimum attenuation is an important aspect which is a direct function of the relative

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permittivity. In these applications, the relative permittivity  $(\varepsilon_r)$  governs the propagation delay  $t_d$ , which is given by the relation [4] :  $t_d = (\sqrt{\varepsilon_r})/c$ , where c is the speed of light in vacuum. Thus materials with low relative permittivity are required to increase the speed of the signal.

### **Quality factor**

One of the important characteristics required for a dielectric material is a high quality factor Q (> 1,000 GHz) which is the inverse of the dielectric loss, tan\delta. The quality factor is determined by the ratio of resonant frequency ( $f_0$ ) to the bandwidth,  $\Delta f_0$ , measured at 3db below the maximum height at resonance, Fig. 1 [3]. This shows that Q is a measure of the selectivity of a resonator to a given frequency. Higher Q values reduce the risk of cross talk within a given frequency range. Q changes with change in frequency. The theoretical relationship between the two is such that  $Q \times f_0$  should be constant and often  $Q \times f_0$  are quoted when comparing materials [3].

### Temperature coefficient of the resonant frequency

The variation of the dielectric properties of ceramics with temperature is very crucial for practical applications. The temperature coefficient of the resonant frequency  $(\tau_f)$  is a measure of the drift in the resonant frequency with respect to temperature. It is evident that a material with a significantly large  $\tau_f$  is not useful in a microwave circuit as it cannot maintain its resonant frequency with changes in the operating temperature [3]. The  $\tau_f$  with a value of 10 ppm/K causes a 0.11% shift of the resonant frequency (5.7 MHz at 5.2 GHz) within the temperature range from -30 to +80 °C, a common temperature range of operation for mobile communication. Large  $\tau_f$  values are undesirable as temperature compensation requires additional mechanical structures or electrical circuits [5]. The temperature coefficient of the resonant frequency



Fig. 1. Schematic showing a resonant peak and associated parameters.

 $\tau_f$  of the resonator material should be close to zero for thermal stability of the device frequency. The  $\tau_f$  is related to temperature coefficient of relative permittivity ( $\tau_\epsilon$ ) and the coefficient of thermal expansion ( $\alpha$ ) by the relation [3]:  $\tau_f = -(\tau_\epsilon/2 - \alpha)$ . It is also related to the temperature coefficient of capacitance ( $\tau_c$ ) by the relation [3]:  $\tau_f = -(\tau_c - \alpha)/2$ .

### **Classification of dielectric ceramics**

The low loss dielectric ceramics developed for various applications can be classified into three categories. The first category includes ceramics with low permittivity and ultra high Q used for millimetrewave and substrate applications. The second class with medium permittivity (25-50) and high Q are used for satellite communication and in cellular phone base station direction. The third category consists of high  $\varepsilon_r$  materials used in mobile phones where miniaturization of a device is very important. These categories of microwave dielectric materials are shown in Fig. 2, in which the quality factor Q.f is plotted as a function of the dielectric constant [6]. Curves in the figure shows an outline of the upper limit of Q.f obtained up to now for a given  $\varepsilon_r$ . The following sub-sections classify the important compositions researched and give the microwave properties reported.

### Ceramics with low permittivity and ultra high Q

Wireless communications have been tremendously developed in the recent ubiquitous age. The utilizable frequency range has been extended to millimetrewaves because of a shortage of the conventional frequencies region. For ultra high frequencies of millimetrewaves and substrate applications, dielectrics with a ultra high quality factor Q and low dielectric constant are desired. Alumina, forsterite and willemite are some of the candidates for this category of dielectrics with ultra high Q and low  $\varepsilon_r$ .

#### Alumina

Alumina is a well-known ceramic packaging material. Powder purity is an important factor in the production of low-loss alumina [7-10]. In polycrystalline alumina, the presence of a very small amount of  $TiO_2$  considerably improved the quality factor [7]. It was found [7] that



Fig. 2. Three categories for research and development of microwave dielectrics [6].

addition of 0.5 wt% TiO<sub>2</sub> lowers the sintering temperature to about 1500 °C with a considerable increase in the quality factor up to 50,300 at 9 GHz which is close to that of single-crystal sapphire. A Qf of 680,000 GHz with  $\varepsilon_r = 10.05$  and  $\tau_f = -60$  ppm/°C was obtained in alumina ceramics sintered at 1550 to 1650 °C for 5 h [11].

Huang *et al.* [12, 13] found that the use of nano particlesized starting material significantly improved the densification and microwave dielectric properties of alumina. The  $\varepsilon_r$  of 10, Q f value of 521,000 (at 14 GHz) and  $\tau_f$  of -48.9 ppm/K were reported for  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> ceramics without a sintering aid at 1550 °C for 4 h [12]. A sample containing 8 wt% of nano-TiO<sub>2</sub> sintered at 1350 °C for 4 hours had  $\varepsilon_r = 10.8$  with Qf = 338,000 GHz and  $\tau_f = 1.3$  ppm/K [13].

Forsterite ceramics

Ohsato and coworkers [14-16] reported forsterite (Mg<sub>2</sub>SiO<sub>4</sub>) as a high Q material for millimetre wave communication as well as a substrate for the MICs. Tsunooka *et al.* [15] reported that high-Q plain forsterite ceramics with Q.f = 240,000 GHz were developed by the usual solid sintering process using highly purified MgO and SiO<sub>2</sub> as raw materials. The composition prepared with an addition of 1 wt.% of TiO<sub>2</sub> exhibited a high Q.f value = 230,000 GHz with  $\varepsilon_r$  = 7.0 and  $\tau_f$  = -65 ppm/K [15]. Latter on the Q.f of forsterite (Mg<sub>2</sub>SiO<sub>4</sub>) has been reported as 270,000 GHz with  $\varepsilon_f$  = 6.8,  $\tau_f$  = -67 ppm/K by Ohsato *et al.* [16].

Willemite ceramics

Willemite (Zn<sub>2</sub>SiO<sub>4</sub>), another low-loss silicate useful as a substrate and in millimetre wave communication, has been successfully prepared by a solid state reaction in the temperature range from 1280 °C to 1340 °C [17]. The zine-silicate has an ultra-high Qf of 219,000 GHz and low  $\varepsilon_r = 6.6$  and  $\tau_f = -61$  ppm/K. Guo *et al.* [17] tailored the high negative  $\tau_f$  of willemite by the addition of TiO2. The willemite-rutile mixture ceramics have excellent dielectric properties. It was found that an addition of 11 wt% TiO<sub>2</sub> sintered at 1250 °C resulted in a temperature-stable ceramic with  $\varepsilon_r = 9.3$ , Qf = 113,000 GHz and  $\tau_{\rm f}\,{=}\,1$  ppm/K [17]. Zou et al. reported that it was difficult to acquire dense ceramics at 1380 °C without sintering aids in the ZnO-SiO<sub>2</sub> system. The samples of ZnO-0.6SiO<sub>2</sub> with Li<sub>2</sub>CO<sub>3</sub> and Bi<sub>2</sub>O<sub>3</sub> additions sintered at 910 °C for 2 h showed excellent properties of  $\varepsilon_r = 6.65$ , Q.f = 33,000 GHz (at 11 GHz) and  $\tau_{\rm f}$  = -70 ppm/K. [18].

### Ceramics with medium permittivity and high Q

Recently, as the number of base stations for telecommunications is increasing rapidly, the resonator is expected to reduce the size for installation and maintenance. So, new materials with medium-to-high permittivity are expected for miniaturization of resonators.

TiO<sub>2</sub> based systems

The  $A_nLa_4Ti_{3+n}O_{12+3n}$  (A = Ba, Sr, Ca) [19-20] homol-

ogous compounds are excellent candidates for dielectrics of base stations with  $\epsilon_r = 44.4$ , Q.f = 410,008 GHz,  $\tau_f = -26$  ppm/K for Ba ;  $\epsilon_r = 43.7$ , Q.f = 46,220 GHz,  $\tau_f = -8.4$  ppm/K for Sr and  $\epsilon_r = 41.1$ , Q.f = 50,246 GHz,  $\tau_f = -25.5$  ppm/K for the Ca analogue.

A few more candidate materials such as BaO- TiO<sub>2</sub>-ZnO :  $\varepsilon_r = 36$ , Q.f = 42,000 GHz [21] and ZnNb<sub>2</sub>O<sub>8</sub>-TiO<sub>2</sub>:  $\varepsilon_r = 37$ , Q.f = 29,000 GHz have also been reported [22].

### Zinc niobates

 $MNb_2O_6$  (M = Ca, Co, Mn, Ni or Zn) ceramics are known [23] as useful microwave dielectric materials. ZnNb<sub>2</sub>O<sub>6</sub> sintered at 1150 °C for 2 h has a Qf of 83,700 GHz and  $\varepsilon_r$  of 25.

### (Mg,Ca)TiO<sub>3</sub>

(Mg,Ca)TiO<sub>3</sub> is a very useful dielectric material for high frequency antenna applications. When sintered at ~1350 °C,  $\varepsilon_r$  is ~20, Qf is 86,000 GHz and  $\tau_f$  is -3 ppm/ K [24]. Its decomposition does not adversely affect the dielectric properties since MgTi<sub>2</sub>O<sub>5</sub> has  $\varepsilon_r = 17.4$  and Qf = 47,000 GHz and Mg<sub>2</sub>TiO<sub>4</sub> has  $\varepsilon_r = 14.4$  and Qf = 55,000 GHz.

#### ZnO-TiO<sub>2</sub> system

The ZnO-TiO<sub>2</sub> system contains ZnTiO<sub>3</sub> (hexagonal), Zn<sub>2</sub>TiO<sub>4</sub> (cubic) and Zn<sub>2</sub>Ti<sub>3</sub>O<sub>8</sub> (cubic) [25]. The preparation of ZnTiO<sub>3</sub> from a mixture of ZnO and TiO<sub>2</sub> is difficult because the compound decomposes into Zn<sub>2</sub>TiO<sub>4</sub> and rutile at ~945 °C [26]. Haga *et al.* [27] in 1992 reported for the first time the microwave dielectric properties of ZnO-TiO<sub>2</sub> ceramics. Later in 1998 Golovochanski *et al.* [28] reported that ZnTiO<sub>3</sub> can be sintered at the relatively low temperature of ~1100 °C with Qf = 30,000 GHz and  $\varepsilon_r = 19$ .

(Zr,Sn)TiO<sub>4</sub> having excellent high frequency properties ( $\epsilon_r = 40$ , Qf = 53,000 GHz and  $\tau_f = 50$  ppm/K) has been used for microwave applications.

### Tellurium based systems

Most tellurium based oxide materials can be synthesized and sintered at temperatures below 900 °C. Udovic *et al.* [29] reported that TeO<sub>2</sub> has a poor sinterability and the ceramic sintered at 640 °C for 15 h has 20% porosity with  $\varepsilon_r = 19.3$ , Q.f = 30,000 GHz and  $\tau_f = -119$  ppm/K. Many authors [29-30] have reported the synthesis of TiTe<sub>3</sub>O<sub>8</sub> with a cubic unit cell from oxides at 700 °C. Kwon *et al.* [31] reported that it is difficult to prepare it as a dense ceramic. Udovic et al. [29] prepared single phase dense TiTe<sub>3</sub>O<sub>8</sub> by muffling in TeO<sub>2</sub> powder and the samples showed  $\varepsilon_r$  of 50 and Qf of 30,600 GHz, but the  $\tau_f$  was +133 ppm/K.

### Bi<sub>2</sub>O<sub>3</sub>-Nb/Ta based compounds

Mixed dielectric structures combine the attributes of high and low relative permittivity materials. Low  $\varepsilon_r$  substrates are useful for impedance matching transmission lines

and for minimizing cross-talk. The incorporation of high  $\varepsilon_r$  bismuth pyrochlore capacitors in low  $\varepsilon_r$  substrates was found [32] to decrease the band pass filter dimensions with an improvement in the overall filter characteristics. Kagata *et al.* [33] also investigated the microwave dielectric properties of bismuth-based compounds. They reported that BiNbO<sub>4</sub> ceramics is difficult to sinter without additives. An addition of small amounts of CuO or V<sub>2</sub>O<sub>5</sub>, CuV<sub>2</sub>O<sub>6</sub>, B<sub>2</sub>O<sub>3</sub> to BiAO<sub>4</sub> (A = Nb/Ta) resulted in dense ceramics with  $\varepsilon_r$  in the range 40-45 and Qf up to 30,000 GHz and low  $\tau_f$  when sintered at a temperature of 950 °C [34-39].

### Ceramics with high permittivity

This sub-section will discuss the systems based on well known dielectric ceramics having high  $\varepsilon_r$  and high Qf for applications in mobile phone handsets.

### TiO<sub>2</sub> systems

TiO<sub>2</sub> is widely researched as a basic ceramic for high permittivity ceramics. Several approaches including monosized [40] or nanosized [41] TiO<sub>2</sub> have been attempted to sinter TiO<sub>2</sub> ceramics at temperatures lower than 1000 °C. Kim *et al.* reported that the addition of CuO to anatase significantly lowered the sintering temperature. The final composition was a mixture of CuO and rutile [42] and with 2 wt-% addition of CuO sintered at 900 °C for 2 h, the  $\varepsilon_{r}$  Qf and  $\tau_{f}$  of 98, 14,000 GHz and 374 ppm/K were achieved.

Detvos *et al.* [43] studied the permittivity-frequency response of compressed TiO<sub>2</sub> being calcined under different conditions in the frequency range 20 Hz-1 MHz. The rutile microcrystal structure almost attains a relaxation-free permittivity response, with low loss characteristics in the frequency range examined. A calcination temperature of 1180 °C provides ceramics with a high packing microcrystal density, having stable  $\varepsilon_r = 12.3$  and  $\tan \delta = 2.3 \times 10^{-3}$  (at 1 MHz).

### BaO-TiO<sub>2</sub> system

The BaO-TiO<sub>2</sub> system is also widely used in microwave applications. The typical phases BaTi<sub>4</sub>O<sub>9</sub> and Ba<sub>2</sub>Ti<sub>9</sub>O<sub>20</sub> are commonly sintered at about 1250 and 1300 °C, and  $\varepsilon_r$  and Qf are close to 36 and 50,000 GHz respectively. Additionally, the  $\tau_f$  value depends on the formation of secondary phases, but can be very close to 0 ppm/K [44]. BaO-TiO<sub>2</sub> with 0.1 wt-%WO<sub>3</sub> (N-35) was found to show excellent microwave dielectric properties with  $\varepsilon_r$  = 35, Qf = 52,000 GHz and  $\tau_f$  close to 0 ppm/K [45]. The sintered N-35 consists of BaTi<sub>4</sub>O<sub>9</sub>, Ba<sub>2</sub>Ti<sub>9</sub>O<sub>20</sub> and BaWO<sub>4</sub> phases. However, its sintering temperature is relatively high (1360 °C).

Wu *et al.* [46] prepared the BaO-TiO<sub>2</sub> system ceramics by a conventional mixed oxide route and reported the dielectric properties at 1 GHz as  $\varepsilon_r = 37$ , Q = 12,500 and  $\tau_f = -29$  ppm/K. ZnO and Nb<sub>2</sub>O<sub>5</sub> were added as sintering agents to lower the sintering temperature to 1260 °C. Weng *et al.* [47] successfully synthesized single phase BaTi<sub>4</sub>O<sub>9</sub> powders and ceramics at a low processing temperature (1250 °C) by a polymeric precursor route. The dielectric properties of BaTi<sub>4</sub>O<sub>9</sub> ceramics were dielectric constant = 35.6; a high Q.f value of 42,600 and the temperature coefficient of resonant frequency of 12 ppm/K.

### BaO-R<sub>2</sub>O<sub>3</sub>-TiO<sub>2</sub> Tungsten bronze type system

The BaO-R<sub>2</sub>O<sub>3</sub>-TiO<sub>2</sub> (R = rare earth, Sm, Nd, Gd, Pr, La) in 1:1:4 or 1:1:5 compositions are excellent dielectric materials having high  $\varepsilon_r$  and high Qf especially suitable for dielectric resonators in mobile phone hand sets [6].

The tungsten bronze-type like Ba<sub>6-3x</sub>R<sub>8+2x</sub>Ti<sub>18</sub> O<sub>54</sub> (R = rare earth) solid solutions sintered at ~1350 °C are well known microwave compositions having moderately high relative permittivity (70-100), high Qf (> 2,000 GHz) and low  $\tau_f$  [47-52]. Similar results for Sm and Nd systems have been reported by Negas *et al.* [53]. The characteristic phenomenon is that Qf values varied non linearly as a function of composition, although  $\varepsilon_r$  and  $\tau_f$  vary proportionally to the composition.

The Ba<sub>6-3x</sub>R<sub>8+2x</sub>Ti<sub>18</sub> O<sub>54</sub> ceramics with x = 2/3 shows the highest Qf values: 10,549 GHz in Sm analogue, 10,010 GHz in the Nd analogue and 2,024 GHz in the La-analogue.

#### Bi<sub>2</sub>O<sub>3</sub> based compounds

Among the bismuth-based ceramics, Bi<sub>2</sub>O<sub>3</sub>-ZnO-Nb<sub>2</sub>O<sub>5</sub> (BZN) ternary oxides have received considerable attention. Several research groups [54-57] studied the structure and properties of BiO-ZnO-Nb<sub>2</sub>O<sub>5</sub> ceramics. There are two main phases in BZN, a cubic pyrochlore phase Bi<sub>1.5</sub>ZnNb<sub>1.5</sub>O<sub>7</sub> ( $\alpha$ -phase) with  $\varepsilon_r \sim 150$  and  $\tau_f$  about -400 ppm/K, and a monoclinic zirconolite like phase  $Bi_2Zn_{2/3}Nb_{4/3}O_7~(\beta\text{-phase})$  with  $\epsilon_r\sim 80$  and  $\tau_f$  about +150 ppm/K. Several authors [58-61] reported that BZN are temperature-stable dielectrics suitable for the capacitor industry with a sintering temperature of ~1000 °C. Yan et al. [59] proposed that the sintering temperature can be further lowered below 950 °C by incorporating Bi<sub>2</sub>O<sub>3</sub>-NiO-Nb<sub>2</sub>O<sub>5</sub> into BZN ceramics. Several authors investigated the microwave dielectric properties of Bi<sub>2</sub>O<sub>3</sub>-ZnO-Nb<sub>2</sub>O<sub>5</sub>/ Ta<sub>2</sub>O<sub>5</sub> ceramics [62-65].

Finally, Bi can be partially substituted by La, Nd, Sm, Ce and Nb/Ta by Mo, Sb, etc. [66-69].

#### Li based systems

Borisevich and Davies reported [70-71] that  $Li_{1+x-y}$ M<sub>1-x-3y</sub>Ti<sub>x+4y</sub>O<sub>3</sub> (M=Nb/Ta; x=0.1; y=0.05-0.175) solid solutions are potential candidate materials for microwave applications with a quality factor up to 10,500 GHz when sintered at 1100 °C. However, volatile Li has a deleterious effect on dielectric properties in Li-based ceramics and the deficiency of Li lead to a decrease in density and lattice defects.

#### Ag based systems

The AgNbO<sub>3</sub> and AgTaO<sub>3</sub> compounds represent a class

of materials with high relative permittivity greater than 400 with Qf in the range 600-900 GHz [72-74]. In general, these compounds undergo a series of structural phase transitions as they cool from the prototypic cubic perovskite phase and AgNbO<sub>3</sub> exhibits a weak ferroelectric behavior at room temperature.

## Conclusions

Microwave dielectric ceramics are being developed for a variety of applications such as miniaturization for mobile phones, a transmitter and receiver with high performance for base station, and millimetrewave applications for ultra speed wireless LAN and ITS. They have been characterized according to these major applications. Microwave dielectric properties reported by various researchers have been summarized for important ceramic systems. This review will provide a vital guideline for further development.

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