PREPARATION, CHARACTERISATION AND MICROWAVE DIELECTRIC PROPERTIES OF
$A_nB_{n-1}O_{3n}$ (n=5, 6, 8) TYPE PEROVSKITE COMPOUNDS

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Thiruvananthapuram

Dated: 9/08/02

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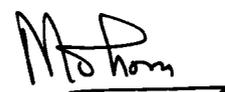
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PREFACE

Dielectric Resonators (DR) are ceramic pieces that can act as frequency determining components at microwave frequencies. DRs should have high dielectric constant (ε_r) in the range 20 to 100 for better miniaturization, high Q factor (Q > 2000) for better frequency selectivity and nearly zero temperature coefficient of resonant frequency (τ_f) for frequency stability with temperature. In addition to the above characteristics, their low cost of production and excellent integrability to microwave integrated circuits (MICs) make them indispensable components in microwave oscillators, filters, duplexers used in cellular phones and in dielectric resonator antennas. Dielectric resonators increasingly replace the conventional resonators such as metallic cavities or micro strip circuits. Though several temperature-stable DRs are available at present, investigation is still going on to find new materials having better dielectric resonator properties. In this work we investigate the microwave dielectric properties of (1) $A_5B_4O_{15}$ (A = Ba, Sr, Mg, Ca, Zn; B = Nb, Ta) ceramics, their solid solutions and mixtures (2) $MO-La_2O_3-TiO_2$ (M = Ba, Sr, Ca) ceramics which mainly consists of $A_nB_{n-1}O_{3n}$ (n = 5, 6, or 8) type cation deficient hexagonal perovskites and (3) a novel method of achieving temperature compensation by stacking positive and negative τ_f resonators. Dielectric resonator properties were studied in terms of phases, crystal structure, crystal symmetry, polarisability of ions, lattice parameters and characterization techniques such as XRD and SEM are employed.

Chapter 1 is a general introduction about material, scientific and technological aspects of DRs. Three important parameters, ε_r, Q and τ_f, used for the DR

characterization are described. The relationship of the above parameters with the fundamental material characteristics is discussed. Different modes are excited when a DR is excited with suitable microwave spectrum of frequencies. A description of analytical determination of frequencies and construction of mode charts used for sample design and mode identification are also discussed.

Chapter 2 presents the methods used for the preparation of ceramics and the various techniques used for the microwave characterization of dielectric properties. The ceramic samples were prepared through the solid-state ceramic route. The dielectric constant of the ceramics at microwave frequencies are measured using the end shorted dielectric post resonator. The quality factors are determined by a transmission mode cavity. The temperature coefficient of resonant frequency (τ_f) is measured by heating the end shorted dielectric post resonator set up in the temperature range 20 to 75°C and by noting the variation of the resonant frequency with temperature. The crystal structure of the samples is analysed using powder X-ray diffraction pattern and surface morphology and grain size is observed using Scanning Electron Microscopy.

Chapter 3 describes the investigation of microwave dielectric properties of $A_5B_4O_{15}$ (A = Ba, Sr, Mg, Ca, Zn; B = Nb, Ta) ceramics. The ceramics show dielectric constant in the range 11 to 51. The hexagonal perovskites show higher dielectric constants than the orthorhombic phases $Mg_5Nb_4O_{15}$ and $Mg_5Ta_4O_{15}$. The FIR and submillimeter techniques are used to study the above compounds. The basic theory of the techniques is discussed. An indirect estimation of the lower limit of dielectric loss and upper limit of dielectric constant at microwave frequencies can be obtained by the extrapolation of real part and imaginary part of the dielectric function down to

microwaves from data obtained through far infra-red spectroscopy. The solid solution phase of the type $Ba_{5-x}Sr_xTa_4O_{15}$, $Sr_5Nb_xTa_{4-x}O_{15}$, $Ba_5Nb_xTa_{4-x}O_{15}$ are prepared and microwave dielectric properties are characterized. The $Sr_5Nb_xTa_{4-x}O_{15}$ phases show abnormal dielectric properties where a decrease in dielectric constant is observed for the intermediate compounds when compared to the end members. This is attributed to the possible structural changes and is evident from analysis of X-ray diffraction data. The $Ba_5Nb_xTa_{4-x}O_{15}$ show linear behaviour for solid solution and intermediate dielectric properties of the end members are obtained. In section of the chapter $xZn_3Nb_2O_8$-$(1-x)ZnNb_2O_6$ mixture phases are discussed. The results are interpreted based on the method of mixtures. The mixture phases show good sinterability and higher quality factors than the end members. The substitution of Zn at Mg site in $Mg_5Nb_4O_{15}$ also gave mixture phases and the compositions $xZnO$-$(5-x)MgO$-$2Nb_2O_5$ showed high quality factors (Q x f up to 89000 GHz) with ε_r in the range 11 to 22. The mixture phases showed intermediate dielectric properties of the end compounds.

Chapter 4 describes the microwave dielectric properties of MO-La_2O_3-TiO_2 (M = Ba, Sr, Ca) ceramics. All the ceramics, except $CaLa_4Ti_5O_{17}$ and $CaLa_8Ti_9O_{31}$, which are orthorhombic structured, belong to the cation deficient hexagonal perovskites belonging to the $A_nB_{n-1}O_{3n}$ (n=5, 6) type compounds. The ceramics show high dielectric constant in the range 41 to 54 with high quality factors and small temperature coefficients of resonant frequencies. The applicability of Claussius - Mossotti equation to these ceramics is discussed. These ceramics are suitable for low frequency applications requiring narrow bandwidth and low insertion loss. The orthorhombic phases show comparatively higher dielectric constants than the hexagonal phases.

Chapter 5 describes a novel method of achieving temperature compensation by stacking positive and negative τ_f resonators. The stack acts as a single resonator. The τ_f of the resultant stack depends on the volume fraction of the positive τ_f and negative τ_f DR materials. The τ_f can be tuned to zero or to a desired value by adjusting the volume fraction of the positive and negative τ_f materials. The dielectric constant and quality factor also change depending on the volume fraction of the two different DR materials. The experiment is performed with varying volume fraction of $Ba_5Nb_4O_{15}$ as the positive τ_f DR and $Sr(Y_{1/2}Nb_{1/2})O_3$ and $5ZnO\text{-}2Nb_2O_5$ as the negative τ_f DR materials. The DR material in the bottom of the stack has greater influence on the τ_f of the resultant stacked resonator.

Chapter 6, gives a summery and conclusion of the present investigation and also discusses the scope for further work in this field.

Chapter 1

INTRODUCTION

1. 1 DIELECTRIC RESONATORS

Dielectric resonators (DRs) are frequency determining components in filters and oscillators used in modern communication systems. Until recently quartz resonators were used to generate, stabilize and filter frequencies in the communication devices. The piezoelectric quartz crystal resonators can be used only up to a few hundred MHz. The recent advances in the communication system increased the number of transmitters and receivers in a particular geographical area, which led to crowding of channels. The only way to prevent interference due to crowding of the channels is to go towards higher frequency range (microwave range). One can use quartz resonators at high frequencies by a frequency multiplication process but leads to high noise and are expensive. Metallic cavity resonators were tried but were very large in size and not integrable in a microwave integrated circuit. Microstrip resonators were also tried but they have low Q with large temperature variation of the resonant frequency. In 1939 Richtmeyer theoretically predicted [1] that a suitably shaped dielectric material could behave as an electromagnetic resonator. In 1960 Okaya [2] found that a

piece of rutile acted as a resonator and later in 1962 Okaya and Barash [3] for the first time analyzed the different modes of a dielectric resonator. In 1968 Cohen [4] for the first time experimentally determined the microwave dielectric properties of a rutile resonator with dielectric constant $\varepsilon_r=104$, quality factor $Q=10,000$ and coefficient of temperature variation of the resonant frequency $\tau_f=+400$ ppm/$^\circ$C. The τ_f of rutile resonator is too high for practical applications. A real breakthrough for dielectric resonators occurred in early 1970's with the development of the first temperature stable, low loss barium tetra-titanate ($BaTi_4O_9$) resonator [5]. Since then extensive work has been carried out on microwave ceramic dielectric resonators.

The recent progress in microwave telecommunication and satellite broadcasting has resulted an increasing demand for Dielectric Resonators (DRs). Technological improvements in DRs have contributed to considerable advancements in wireless communications [8-22]. Ceramic dielectric resonators have advantage of being more miniaturized as compared to traditional microwave cavities, while having a significantly higher quality factor than transmission lines and microstrips. DRs are advantageous in terms of compactness, light weight, stability and relatively low cost of production as compared to the conventional bulky metallic cavity resonators. In addition temperature variation of the resonant frequency of dielectric resonators can be engineered to a desired value to meet circuit designers requirements. Table 1.1 gives comparison of the properties of metallic cavities, microstrips and dielectric resonators. Functioning as

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important components in communication circuits, DRs can create, filter [31-39] and select frequencies in oscillators [24-30], amplifiers and tuners.

Table 1.1: Comparison of the properties of metallic cavity, microstrip and dielectric resonator

Component	Size	Q factor	τ_f	Integabiltiy in a MIC
Metallic Cavity (Cu, Brass, invar etc)	Large	High	Low	Nonintegrable
Microstrip resonators	Very small	Very low	Very High	Integrable
DR .	Small	Very high	Very Low	Integrable

DRs are important components in duplexers, multiplexers, combiners, radar detectors, collision avoidance systems, automatic door opening systems, telemetry, cellular radio, cordless phones or personnel communication systems, global positioning systems, TVRO, satellite and military communication systems.

1. 1. 1 Resonance

A dielectric resonator should have maximum confinement of energy within the resonator when used at a particular resonant frequency. The resonance occurs by total multiple internal reflections of microwaves at the boundary or dielectric-air interface (see Fig. 1. 1). If the transverse dimensions of the dielectric

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are comparable to the wave length of the microwave, then certain field

distributions
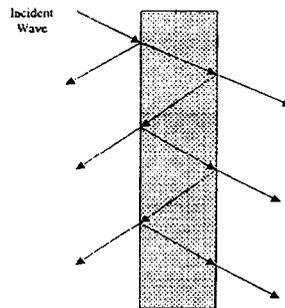
Fig.1. 1 Schematic sketch showing total multiple internal
reflections at the air-dielectric interface

or modes will satisfy Maxwell's equations and boundary conditions. It was found

that through multiple total internal reflections, a piece of dielectric with high

dielectric constant can confine microwave energy at a few discrete frequencies,

provided that the energy is fed in the appropriate direction. The reflection

coefficient approaches unity when the dielectric constant approaches infinity. In

the microwave frequency range free space wavelength (λ_o) is in centimeters and

hence the wavelength (λ_g) inside the dielectric will be in millimeters only when

the value of the dielectric constant ε is in the range 20-100. Hence the dimensions

of the dielectric sample must be of the same order (in millimeters) for the

resonance to occur. Still larger values of the dielectric constant gives better

confinement of energy, reduced radiation loss and further miniaturization but will

result in higher dielectric losses because of the inherent material properties. A

high dielectric constant material can confine most of the standing electromagnetic

wave within its volume due to reflections at the air dielectric interface. The

frequency of the standing wave depends on the dimensions and dielectric constant of the dielectric. The electromagnetic fields outside the dielectric sample decay rapidly. One can prevent radiation losses by placing the DR in a small metallic enclosure. Since only a small radiation field sees the metallic surface, the resulting conduction loss will be too small and can be neglected.

1. 1. 2 Types of Dielectric Resonators

The disk shaped dielectric material is the simplest form of a dielectric resonator. The usual geometries of DRs are discs, rings and parallelpipeds. By inserting a metal or ferrite screws into the central hole of a ring resonator, the resonant frequency of modes can be tuned. Similar techniques are used to suppress the modes adjacent to the desired mode, to avoid interference and to reduce the dielectric loss. The mode spectrum and resonant frequencies of DRs greatly depend on the aspect ratio (diameter D/length L). The dimensions of the specimen are important to achieve wide separation of modes. The proper aspect ratios are 1.0 to 1.3 and 1.9 to 2.3. In practice the specimen diameters in the range 7 to 25 mm have been found most suitable.

There are two main types of resonators, coaxial and dielectric resonators, employed in the frequency range 500MHz to 30 GHz using the available materials today ($10 < \varepsilon_r < 120$). The coaxial resonators which are tubular

in appearance are used for frequencies up to 3 GHz. The coaxial resonators are also called λ/4 resonators. Their length is determined by

$$l = \frac{\lambda_0}{4\varepsilon_r}$$ where λ_0 is the vacuum wave length at the resonant frequency

They have four times more size reduction than the dielectric resonators. The tubular coaxial resonators are given a thin metallic coating and the resonance is by the total multiple internal reflections at the dielectric-metal interface. The quality factor of coaxial resonators is limited to values less than 1500 by the finite conductivity of the metallic surface of the tubular resonator. These types of resonators are commonly used in cellular telephone systems at about 800MHz where miniaturization is very important. At higher frequencies cylindrical dielectric resonators (DRs) are used. For a cylindrical resonator the required diameter is proportionally reduced as follows

$$D = \lambda_0 \frac{1}{\sqrt{\varepsilon}}$$

1. 1. 3 Analytical Determination of Frequencies

Practical circuits employing DRs are of different types. DRs placed between two parallel conducting plates, DRs enclosed by metal shields, DR enclosed in substrate-box system, open dielectric resonators are some of the common structures. When the DR enclosed structure is fed with microwaves different modes gets excited. The $TE_{01\delta}$ mode is the most commonly used mode

for practical applications. It is of great importance if the resonant frequency of the DR enclosed structure can be analytically determined. An exact analysis usually lead to complex solutions, which is very difficult to implément. Hence using some simple models we can compute the resonant frequencies with a small percentage of error.

One of the first model to suggest was the magnetic wall model [74-76]. Here the cylindrical surface containing the circumference of the resonator is replaced with a fictitious open circuit boundary (magnetic wall). The tangential magnetic field component and normal field component vanish at the DR-air boundary. Some of the field leaks out of the DR and if not taken into account results in discrepancies with the measured results. The method often leads to an error of less than 10 %. The variational method developed by Konishi et al. [77] has an error of less than 1 %. The method is computationally complex. Itoh Rudokas [78] Model is less complex and gives accuracies very near to the variational method. Guillon and Garault [79] proposed a method where all the surfaces are simultaneously considered as imperfect magnetic walls. The method has an accuracy of better than 1 %. Some rigorous analytical formulation is also found which determine the complex resonant frequencies of isolated cylindrical dielectric resonators. Glisson et al. [80] have applied a surface integral formulation and the method of moments. Tsuji et al. [81] have presented an alternative method, in which the resonator fields are expanded into truncated series of solutions of the Helmholtz equation in spherical polar coordinates, and the boundary condition on the resonator surface is treated in the least square

sense. Both these methods are reported to give highly accurate values of resonant frequencies and Q factors, substantiated by experiment. Mongia et al. [82] have reported an effective dielectric model that is a simple, analytical technique to determine the resonant frequencies of isolated dielectric resonators. The method yields results as accurate as those reported using rigorous methods. Tobar et al. [83] has developed an improved method, which allows the determination of mode frequencies to high accuracy in cylindrical anisotropic dielectric resonators. Yousefi et al. [84] have applied the GIBC (generalised Impedance boundary condition) formulation for the determination of resonant frequencies and field distribution of a substrate mounted dielectric resonator. Apart from that one can find several other different methods for finding the resonant frequencies reported during the last two decades.

1. 1. 4 Mode Chart

If one can analytically determine frequencies corresponding to various modes mode charts can be constructed. A mode chart helps to find out how different modes behaves with resonator parameters. It helps to find out sample dimensions of dielectric resonator filter circuits corresponding to aspect ratio where maximum mode separation with adjacent modes are obtained.

As an example a typical mode chart is constructed (Fig.1. 2). The mode chart is constructed for the end shorted dielectric rod configuration based on the

theory developed by Pospieszalski [85]. The dielectric resonator in the shape of a cylinder with diameter D and length L placed between two conducting plates constitute the resonant structure. For very high ε_r and D/L>0, the solution for the characteristic equation corresponding to the HE_{111}, HE_{211} modes in the broad
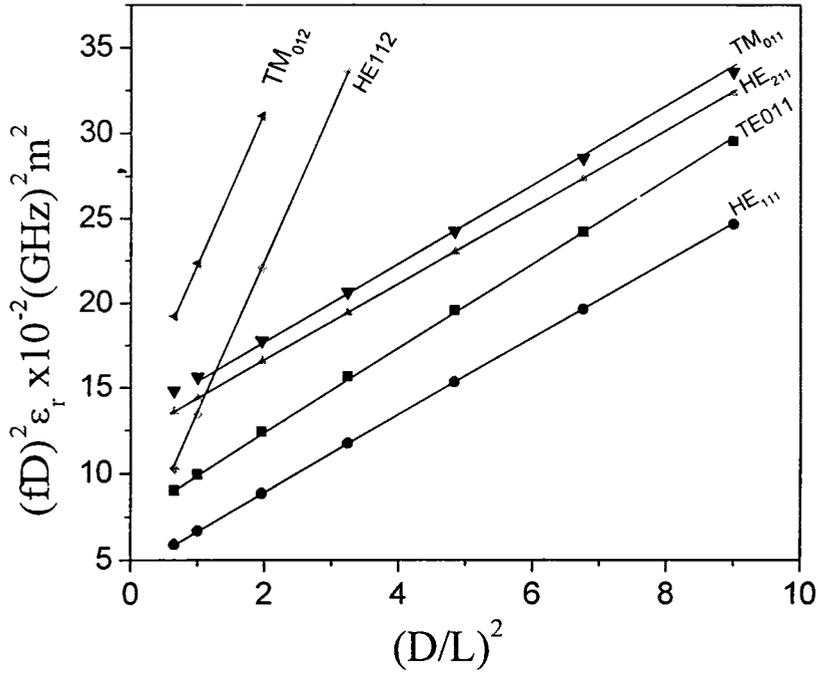
Fig. 1. 2 The mode chart constructed for a material with $\varepsilon_r = 22$

range of ε_r and D/L are almost straight lines from which simple approximate formulas can be derived.

For HE_{111} modes if $\varepsilon_r > 500$ and $1 \leq (D/L)^2 \leq 15$

$$F_0^{\ 2} = (2.21)^2 + (\frac{\pi Dl}{2L})^2$$

(1. 1)

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where $F_0 = (\dfrac{\pi D}{\lambda_0})^2 \varepsilon_r$ with accuracy better than 0.7%. D is the diameter of the dielectric rod, L its length and l is the no of field variations along the axis.

Similarly for HE_{211} mode

$$F_0^{\ 2} = (3.65)^2 + (\frac{\pi D l}{2L})^2 \qquad\qquad (1.2)$$

Accuracy of expression (2) is better than 0.7% if $\varepsilon_r \geq 500$ and $2 \leq (D/L)^2 \leq 15$.

For circularly symmetric modes the characteristic equation has very simple forms.

For the TM_{0ml},

$$\frac{J_1(u)}{u J_0(u)} = -\frac{1}{\varepsilon_r}\frac{K_1(w)}{w K_0(w)} \qquad\qquad (1.3)$$

For the TE_{0ml}

$$\frac{J_1(u)}{u J_0(u)} = -\frac{K_1(w)}{w K_0(w)} \qquad\qquad (1.4)$$

If ε_r is large enough the solution of (3) for w>0 can be approximated by the solution of

$$J_1(u) = 0 \qquad\qquad (1.5)$$

Therefore for TM_{0ml}

$$F_0{}^2 = \rho_{1m}{}^2 + (\frac{\pi Dl}{2L})^2 \qquad\qquad (1.6)$$

where ρ_{1m} is the mth greater than zero solution of (5). For $\varepsilon_r > 20$; $(D/L)^2 \geq 1$, the accuracy is better than 2%. The accuracy is better than 0.5% if $\varepsilon_r > 100$ and $(D/L)^2 \geq 1$.

For the TE_{0ml} modes the approximate solution is

$$F_0{}^2 = \rho_{1m}{}^2 (1 - \frac{1}{\sqrt{\rho_{1m}{}^2 + (\frac{\pi Dl}{2L})^2}})^2 + (\frac{\pi Dl}{2L})^2 \qquad\qquad (1.7)$$

Accuracy is better than 0.5% if $\varepsilon \geq 500, 3 \leq (D/L)^2 \leq 15$.

Computer program is developed such that the resonant frequency can be obtained as a function of dielectric constant, length and diameter of the sample. As an example mode chart is constructed for a material with dielectric constant $=22$ by plotting different values of $(fD)^2 \varepsilon_r$ for different values of $(D/L)^2$ for different modes (See Fig. 1. 2). It is evident from the graph that $(D/L)^2$ around 4 is good for maximum mode separation. For low value of D/L various modes tends to interfere to destroy the quality of resonance.

1.2. MATERIALS REQUIREMENTS

The important characteristics required for a dielectric resonator material for practical applications are

1. 2. 1. High dielectric constant.

High dielectric constant (ε_r) in the range 20-100 or more are needed for applications. High dielectric constant facilitates miniaturization of the devices and the miniaturization is proportional to $1/(\varepsilon)^{1/2}$. According to classical dispersion theory [6,7] the crystal is approximated as a system of damped oscillators having an appropriate frequency and dipole moment. The real and imaginary parts of the complex dielectric constant (ε',ε'') as functions of ω where $\omega=2\pi\nu$) are given by

$$\varepsilon'(\omega) = \varepsilon_\infty + \sum_j \frac{4\pi\rho_j(\omega_j^{\ 2} - \omega^2)\omega_j^{\ 2}}{(\omega_j - \omega^2)^2 + (\gamma_j\omega)^2} \qquad (1.\ 8)$$

where $4\pi\rho_j$ is the oscillator strength, ω_j is the resonant angular frequency of frequency of the j th oscillator, ε_∞ is the dielectric constant caused by electronic polarization at higher frequencies and γ_j is the damping constant which is given

by the width of the peak. The summation is over the j resonances in the spectrum.

Each resonance is characterized by its dispersion parameters.

For $\omega_j >> \omega$,   .

$$\varepsilon'(\omega) = \varepsilon_\infty + \sum_j 4\pi\rho_j \qquad (1.9)$$

From the above equation it is clear that the dielectric constant is independent of

frequency in the microwave frequency region.

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1. 2. 2 High quality factor (low dielectric loss).

Any type of defects such as grain boundaries, stacking faults, chemical or

structural disorder, point defects, planar defects, line defects, inclusions,

secondary phases, twinning, porosity etc contribute losses. For an ideal crystal

quality factor is approximately equal to the reciprocal of the dielectric loss (tan δ).

In the microwave region the loss is mainly due to the interaction of the applied

field with phonons [8].   The microwave energy is transferred to transverse

optical phonons. These optical phonons can then generate thermal phonons. This

leads to damping of the optical lattice vibrations and therefore causes dielectric

loss. Hence there is a linear increase of loss with frequency and is a characteristic

phonon effect. From the classical dispersion theory,

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$$\varepsilon^{''}(\omega) = \sum_j \frac{4\pi\rho_j(\gamma_j\omega)\omega_j^2}{(\omega_j^2 - \omega^2)^2 + (\gamma_j\omega)^2} \qquad (1.10)$$

For $\omega_j \gg \omega$

$$\varepsilon^{''}(\omega) = \sum_j \frac{4\pi\rho_j(\gamma_j\omega)}{\omega_j^2} \qquad (1.11)$$

The above equation shows that loss is frequency dependent. Several phonon processes contribute to intrinsic losses in a dielectric depending on the ac field frequency, temperature range and the symmetry of the crystal under consideration. In general the losses are lower for Centro symmetric crystals than the non-Centro symmetric crystals.

In the case of a resonator the unloaded quality factor can be expressed as

$$1/Q_u = 1/Q_d + 1/Q_r + 1/Q_c \qquad (1.12)$$

where 1/Qd, 1/Qr and 1/Qc are the dielectric loss, radiation loss and conduction loss respectively. Normally a quality factor greater than 2000 is required for practical applications

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1. 2. 3 The coefficient of temperature variation of the resonant frequency (τ_f).

The coefficient of temperature variation of the resonant frequency(τ_f) determines the frequency stability. τ_f is defined as

$$\tau_f = (1/f)(\Delta f/\Delta T) \tag{1. 13}$$

where f is the resonant frequency at room temperature, Δf is the variation of resonant frequency from room temperature for a change in temperature ΔT. The τ_f depends on the temperature variation of ε_r and coefficient of linear thermal expansion according to the expression

$$\tau_f = -\alpha_L - \varepsilon_r/2 \tag{1. 14}$$

The value of τ_f should be near to zero for practical applications. However often the device engineer requires a low positive or negative τ_f to compensate for the temperature variation of the resonant frequency due to the circuit.

The extensive research in last three decades has provided several suitable DR materials like $(Mg,Ca)TiO_3$ [42-47] , complex perovskites [48-52], $BaTi_4O_9$-$Ba_2Ti_9O_{20}$ [53-59], $(Zr,Sn)TiO_4$ [60-62], BaO-Ln_2O_3-TiO_2 [63-72] system

1. 3 POLARISATION MECHANISMS IN DIELECTRICS

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Under the influence of an electric field on a dielectric material four types polarisation mechanisms can occur, i.e. interfacial, dipolar, ionic and electronic. Piling up of mobile charge carriers at physical barrier such as grain boundary causes for interfacial polarisation or space charge polarisation. At low frequencies the mechanism gives rise to high dielectric constant and in some cases may extend up to 10^3 Hz.

In zero field the permanent dipoles will be randomly oriented and the system has no net polarisation, but an electric field will tend to align the dipoles and the materials will acquire a net moment. This is called orientational polarisation. In other words, the perturbation of thermal motion of the ionic or molecular dipoles, producing a net dipolar orientation in the direction of the applied field. Two mechanisms can be operative in this case. [16]. (a) In linear dielectrics (non-ferroelectrics) dipolar polarisation results from the motion of the charged ions between the interstitial positions in ionic structures parallel to the applied field direction. The mechanism is active in the 10^3-10^6 Hz range. (b) Molecules having permanent dipole moment may be rotated about an equilibrium position against an elastic restoring position. Its frequency of relaxation is very high of the order of ~10^{11}Hz. The dipolar polarisation contributes to the dielectric constant in the sub-infrared range of frequencies.

The displacement of positive and negative ions with respect to each other gives rise to ionic polarisation. The mechanism contributes to the dielectric constant at infrared frequency range ($\sim 10^{12}$-10^{13} Hz).

When an electric field is applied the valence electron cloud shifts with respect to the nucleus the atom acquires a dipole moment. This occurs at high frequencies of about 10^{15} GHz. The refractive index (η) and electronic polarisability (α_e) are related by the relation as α_e are related by the equation

$$\alpha_e = [\frac{4\pi V_m}{3}(\frac{\eta^2 - 1}{\eta^2 + 2})]^{-1} \qquad (1.15)$$

where V_m is the molar volume of the material and η the refractive index is equal to $\varepsilon_r^{1/2}$ which is valid for all materials well at optical frequencies, but there is disagreement when dipolar polarisation is present. At microwave frequencies the mechanisms due to ionic and electronic polarisation contribute to the dielectric properties.

1. 4 BEHAVIOUR OF DIELECTRIC WITH RESPECT TO FREQUENCY

From Maxwell's equations, it can be shown that the refractive index (η) of a material is equal to $\varepsilon_r^{1/2}$. But experiments show much difference between two

values for most of the materials. The measured ε_r usually found to decrease with frequency. It follows fairly sharp drops at certain frequencies. Associated with each of these drops there is a region of energy dissipation or dielectric loss. This indicates the switching of one of these polarisation mechanisms because polarisation can no longer keep in step with the applied field. There are two different types of mechanisms, which can give rise to this kind of behaviour. They are resonance absorption and dipole relaxation.

1. 4. 1 Resonant absorption

A dipole will have a natural frequency of oscillation. The dipole follows the variation of an applied field of frequency ω, only if $\omega < \omega_0$. IF x is the relative separation of charges on a dipole, then mathematically

$$m\frac{d^2x}{dt^2} + \gamma\frac{dx}{dt} + \omega_0^2 x = -eE\exp(-i\omega t) \qquad (1.16)$$

The steady state solution of the above equation is

$$x = -\frac{eE}{m\{(\omega_0^2 - \omega^2) - i\gamma\omega\}}\exp(-i\omega t) \qquad (1.17)$$

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The displacement x is proportional to polarisability α_D.

Polarisability can be written as

$$\alpha_D = e\frac{x}{E}$$

(1. 18)

Then the susceptibility is

$$\chi = \frac{Ne}{\varepsilon_0}(\frac{x}{E})$$

(1. 19)

Hence

$$\chi = \frac{Ne^2}{m\varepsilon_0}\{\frac{\omega_o^2 - \omega^2}{(\omega_o^2 - \omega^2)^2 + \gamma^2\omega^2} - \frac{i\gamma\omega}{(\omega_o^2 - \omega^2)^2 + \gamma^2\omega^2}\}$$

(1. 20)

The dielectric constant is related to susceptibility as

$$\varepsilon_r - 1 = \chi \quad \text{or} \quad \varepsilon_r = 1 + \chi$$

(1. 21)

The complex dielectric constant $\quad \varepsilon^* = \varepsilon_r - i\varepsilon_r^{"}$

(1. 22)

Hence we can write

$$\varepsilon_r = 1 + \frac{Ne^2}{m\varepsilon_0}\{\frac{{\omega_0}^2 - \omega^2}{({\omega_0}^2 - \omega^2)^2 + \gamma^2\omega^2}\} \qquad \cdot \qquad (1.23)$$

$$\varepsilon_r" = \frac{Ne^2}{m\varepsilon_0}\{\frac{\gamma\omega}{({\omega_0}^2 - \omega^2)^2 + \gamma^2\omega^2}\} \qquad (1.24)$$

The above equations are known a dielectric dispersion formula. The peaking of ε"

shows that the loss is maximum at ω = ω₀.

The following dispersion relations can be obtained from the classical theory

$$\varepsilon_r = \varepsilon_\infty + \sum_i \frac{4\pi\rho_i{\omega_i}^2({\omega_i}^2 - \omega^2)}{({\omega_i}^2 - \omega^2)^2 + {\gamma_i}^2{\omega_i}^2} \qquad (1.25)$$

$$\varepsilon_r" = \sum_i \frac{4\pi\rho_i\omega_i\gamma_i\omega}{({\omega_i}^2 - \omega^2)^2 + {\gamma_i}^2{\omega_i}^2} \qquad (1.26)$$

where $4\pi\rho_i$ is the strength of oscillation of the ith dipole, ω_i its natural frequency

of oscillation and ω is the frequency of the external field.

1. 4. 2 Relaxation Absorption

The phenomenon of dielectric relaxation arises only within polar molecules. If a permanent dipole is oriented in an electric field and is then displaced, it will vibrate about the field direction and eventually by interaction with its surroundings it will relax back to the original position. The Debye expressions for the relaxation mechanism are [15]

$$\varepsilon = \varepsilon_\infty + \frac{\varepsilon_s - \varepsilon_\infty}{1 + \omega^2 \tau^2} \qquad (1.27)$$

and

$$\varepsilon'' = \frac{(\varepsilon_s - \varepsilon_\infty)\omega\tau}{1 + \omega^2 \tau^2} \qquad (1.28)$$

where ε_s is the low frequency(static) dielectric constant and ε_∞ is the dielectric constant at very high frequency and τ is the relaxation time. These two phenomena gives rise to a frequency dependant dielectric constant in materials.

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1.5 DIELECTRIC CONSTANT

Consider unit volume of a material containing N atoms. When a dielectric is subjected to an external electric field E_{ext} dipole moments are induced inside the material. Let P be the dielectric polarisation, which is equal to the total dipole moment induced in the material by the electric field. The total polarisation

$$P = N\alpha E_{local} \qquad (1.29)$$

.

It can be shown that the local field acting on the dipoles

$$\cdot\ E_{local} = E_{Ext} + 4\pi\frac{P}{3} \qquad (1.30)$$

where E_{local} is the local electric field, E_{ext} is the external electric field, P-polarsation

The dipole moment of a single atom p is proportional to the field. $p = \alpha E_{local}$ where α is the electrical polarisability of the atom. The total polarisation of an insulator containing N atoms is

$$\sum_{i=1}^{n} n_i \alpha_i\ E_{local} \qquad (1.31)$$

where n_i is the number of i atoms having polarisabilities α_i and acted on by local field E_{local}. Hence from equations (22) and (23), by rearranging terms we can obtain the Claussius - Massotti equation

$$\frac{\varepsilon_r - 1}{\varepsilon_r + 2} = \frac{4\pi}{3} \sum_i n_i \alpha_i \qquad (1.32)$$

From the expression polarisabilities are additive. The additive relation is valid for electronic, ionic and dipolar polarisation. Hence expression can be written in the total poalrisability form. If N is the number of atoms or ions per unit volume

$$\frac{\varepsilon_r - 1}{\varepsilon_r + 2} = \frac{4\pi}{3} N\alpha \qquad (1.33)$$

The above formula gives Clausius-Massotti relation.

In other words

$$\varepsilon_r = \frac{3 + 8\pi N\alpha}{3 - 4\pi N\alpha} \qquad (1.34)$$

But N = 1/V_m

Hence

$$\varepsilon_r = \frac{3 + \dfrac{8\pi\alpha}{V_m}}{3 - \dfrac{4\pi N\alpha}{V_m}} \qquad (1.35)$$

which simplifies to

$$\varepsilon_r = \frac{3V_m + 8\pi\alpha}{3V_m - 8\pi\alpha} \qquad (1.36)$$

The above expression gives the dielectric constant in relation with molar volume in $\mathring{A}^3$ and α gives the total dielectric polarisability of individual ions using Shannon's dielectric polarisability [77].

1.5.1 Determination of ε_r

From the classical dispersion equation, at frequencies very much less than the oscillation frequency of dipole, i. e, when $\omega \ll \omega_i$, it can be shown that ε_r is independent of frequencies in the microwave range. Hence we can write

$$\varepsilon' = \varepsilon_\infty + \sum 4\pi\rho_i \qquad (1.37)$$

where ε_∞ is the dielectric constant caused by electronic polarisability at very high frequencies and $4\pi\rho_i$ is the strength of oscillation. It can be understood that ε_r is a constant in the microwave region since it is independent of frequency. Using Far-Infrared spectroscopy the reflectance as well as transmittance can be recorded. From the reflection band, ε_r is calculated using Krammer- Kronig Analysis. The method gives an indirect estimation of the dielectric constant. In the microwave frequency region ε_r is measured from the resonance spectra using the resonance method. The methods used will be discussed in the next chapter.

1.6 QUALITY FACTOR (Q FACTOR)

The figure of merit for assessing the performance or quality of a resonator

is Q factor. It is a measure of energy loss or dissipation per cycle as compared to the energy stored in the fields inside the resonator. Q factor is defined by

$$Q = \frac{\text{Maximum energy stored per cycle}}{\text{Average energy-Dissipated per cycle}} \qquad (1.38)$$

$$Q = \frac{2\pi W_0}{PT} = \frac{\omega_0 W_0}{P} \qquad (1.39)$$

where W_0 is the stored energy, P is power dissipation, ω_0 is resonant radian frequency and period $T = \dfrac{2\pi}{\omega_0}$

To a very good approximation, it can be shown that

$$Q = \frac{\omega_o}{\Delta\omega} = \frac{f_o}{\Delta f} \qquad (1.40)$$

When a resonant circuit or cavity is used as a load in a microwave circuit, several different Q factors can be defined. First Q accounts for internal losses. It is the unloaded Q factor Q_0. Next external Q factor Q_e, accounts for external losses. When the resonator is used in actual circuit there arises the loaded Q factor, Q_L which is the overall Q factor and includes both internal and external losses.

The dielectric Q factor Q_d for homogeneous dielectric material is given by

$$Q_d = \frac{1}{\tan\delta} \qquad (1.41)$$

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For a dielectric loaded cavity where cavity containing an aperture

$$\frac{1}{Q_0} = \frac{1}{Q_c} + \frac{1}{Q_d} + \frac{1}{Q_r} \qquad (1.42)$$

where Q_c is the conduction Q factor, Q_d is the dielectric Q factor and Q_r the radiation Q factor. When the resonator is connected to load

$$\frac{1}{Q_L} = \frac{1}{Q_e} + \frac{1}{Q_o} \qquad (1.43)$$

where Q_L is the loaded Q factor, Q_e the external Q factor and Q_0 the unloaded Q factor.

The Q factor can be determined by the resonance method, which will be described in the next chapter.

1. 6. 1 Determination of Q factor

It follows form classical dispersion relation [7] where $\omega < \omega_i$

$$\varepsilon''(\omega) = \sum_i \frac{4\pi\rho_i\gamma_i\omega}{\omega_i^2} \qquad (1.44)$$

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The above equation shows that $\varepsilon''(\omega)$ is directly proportional to applied

frequency. That means loss factor increases with frequency. At microwave

frequencies, for an ideal DR material $Q \approx \dfrac{1}{\tan \delta}$. Hence a decrease in Q factor

with respect to frequency in the microwave range.

ε' and ε'' can be determined by FTIR reflectance spectra using Krammer-

Kronig analysis. However far infrared reflectivity spectra has limited sensitivity

to the weak modes in the submillimeter (SMM) region ($10\text{-}100\text{cm}^{-1}$). Hence a

generalised factorised four parameter oscillator model of complex permittivity is

used.

$$\varepsilon^*(\omega) = \varepsilon_\infty \prod_j \frac{\omega^2_{LOJ} - \omega^2 + i\omega\gamma_{LOj}}{\omega^2_{TOJ} - \omega^2 + i\omega\gamma_{TOj}}$$

(1.45)

Here reflectivity spectra are fitted together with the transmission spectra.

The reason for simultaneous fitting is that in many cases the parameters of a good

reflectivity spectra do not fit satisfactorily with transmission spectra.

1.7 TEMPERATURE COEFFICIENT OF RESONANT
   FREQUENCY

The stability of the resonant frequency of a resonator with temperature is an

important parameter for practical applications. The temperature coefficient of

resonant frequency is defined as

$$\tau_f = \frac{1}{f} \frac{\Delta f}{\Delta T}$$

(1. 46)

.

The unit of τ_f is in parts per million per Kelvin. It can be shown that in the case of

cavity resonators, the temperature coefficient of resonant frequency $\tau_f = -\alpha_T$.

In the case of a dielectric resonator we can write $\lambda = \lambda_0 \varepsilon_r^{-1/2} = 2L$. WE can

show that $\tau_f = -(\tau_\varepsilon / 2 + \alpha_T)$. In order to get $\tau_f = 0$, $\tau_\varepsilon = -2\alpha_T$.

1. 7. 1 Determination of τ_f

The τ_f can be determined by finding τ_ε at low frequencies from the change

in capacitance with temperature and also by finding out linear expansivity α_T. In

the cases of dielectric resonators we can determine τ_f directly by noting the change

in resonant frequency with temperature. The method we have used is described in

the next chapter.

1.8 DIELECTRIC RESONATORS AT MICROWAVE

FREQUENCIES

For practical applications DRs are developed in different shapes. The most popular one is disk shaped DRs. When a spectrum of microwave frequencies is applied several modes get excited .DRs supports not only the transverse electric (TE) and transverse magnetic (TM) modes but also a family of hybrid (HEM) modes. Among the various modes the $TE_{01\delta}$ is the most commonly used mode. The subscripts denote the azimuthal, radial and axial modes respectively. Due to the finite dielectric constant a part of the field exists outside the DR. Hence the subscript δ is used, which is not a whole number.

In $TE_{01\delta}$ mode, the magnetic field lines are confined in the meridian plane. The electric field lines are concentric circles around z-axis. An exact solution to Maxwell's equation to such simple shapes such as cylindrical DR is not simple. Hence numerical techniques are employed to get exact solutions.

In the case of DRs the presence of evanescent fields helps for coupling or tuning with adjacent DRs. Different types of coupling are employed in DR circuits. DR coupled to a micro strip line is most commonly used configuration. Coupling between adjacent DRs is made easy because of the presence of evanescent field. The resonant frequency of a DR can be controlled by changing its length, diameter or both. Once DR is put into use tuning will be required to get the desired frequency. DRs can be tuned using mechanical structures, varactors or other electronic devices.

1. 9 DIELECTRIC RESONATOR MATERIALS

There are a number of research works for developing new ceramics and also for improving the dielectric properties. (Mg, Ca)TiO$_3$ [42-47] has been attempted as DR material. Complex perovskite [48-52] materials showed excellent microwave dielectric properties. Ba(Mg$_{1/3}$Ta$_{2/3}$)O$_3$ showed high dielectric constant of 25 with high Q factor as high as Q x f=350000 and near to zero τ_f. Ba(Zn$_{1/3}$Ta$_{2/3}$)O$_3$ showed high dielectric constant 29 with high Q x f as high as 150000 and near to zero τ_f., BaTi$_4$O$_9$.Ba$_2$Ti$_9$O$_{20}$ [53-59] (Zr, Sn)TiO$_4$ [60-62]ceramics also gained commercial acceptance. Complex perovskite type materials showed excellent microwave dielectric properties. All these ceramics possess ε_r<45 and for applications near 1 GHz their size will be large. Hence tungsten Bronze type BaO-RE$_2$O$_3$-TiO$_2$ [63-72] mixed oxide with high dielectric constant >80 is used in this range. Still the search for new materials is going on to get ceramics with improved properties.

1. 10 APPLICATIONS OF DIELECTRIC RESONATORS

Coupling to the dielectric resonator is easy because of field existing outside the DR. The strength of coupling is usually determined by the geometrical placing of the resonator.

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