

## Microwave Electromagnetic Characteristics of New Substituted M-Hexaferrites $\text{BaFe}_{12-2x}\text{A}_x\text{Me}_x\text{O}_{19}$ (A = Ru, Ir; Me = Co, Zn)

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**Abstract.** The substitution of  $x(\text{Ti-Co})$  for  $2x$  Fe cations reduces the strong axial anisotropy of the BaM-hexaferrite, which becomes in-plane for  $x = 1.3$ . Then M(Ti-Co) substituted hexaferrite have been extensively investigated as promising material for magnetic recording or hyperfrequency wave absorption. But saturation magnetization and ordering temperature are unfortunately reduced in proportion to the substitution ratio. Doping barium hexaferrite with A-Me elements (A = Ru, Ir ; Me = Co, Zn) produces the same effects for a much smaller substitution ratio. Ordering temperature and saturation magnetization are consequently much higher. Large saturation magnetization associated with in-plane magnetic anisotropy allows to obtain high magnetic permeability values on a wide frequency range at high values of some GHz. Complex electromagnetic characteristics :  $\mu'$ ,  $\mu''$ ,  $\epsilon'$  and  $\epsilon''$  of these new M(A-Me) substituted hexaferrites have been simultaneously measured at high frequency (0.5 to 18 GHz) using coaxial wire device on ceramic samples. The ferromagnetic resonance observed in these ceramics strongly depends on both their magnetic and microstructural properties. Results are discussed in terms of substitution ratio and microstructure depending on preparation conditions.

### 1. INTRODUCTION

Permittivities and permeabilities of ferrites are frequency dependent. Since the complex permittivity of a ferrite is roughly constant in the GHz frequency range, the permeability related to the gyromagnetism and the domain wall motion strongly depends on the frequency.

With M(Ti-Co) substituted hexaferrites, the substitution ratio allows to control within a wide range the magnetic anisotropy which even becomes in-plane at  $x = 1.3$ . Such materials opens up the possibility both a strong domain wall relaxation and a strong FMR in a frequency range between 100 MHz and several GHz. Unfortunately the substitution makes the saturation magnetization decrease which proportionally limits the magnetic permeability, and also the order temperature which reduces the use range in temperature.

Many years ago TAUBER [1] showed new BaM(Ir-Zn) substituted hexaferrites with in-plane magnetic anisotropy possess high  $M_s$  and  $H_{A\theta}$  values for a weak substitution ratio. We have begun a systematic study of new BaM(A-Me) substituted hexaferrites, and showed that with A = Ru, Ir and Me = Co, Zn, the same effects are obtained as with Ti and Co elements but for much smaller substitution ratios [2, 3]. In this work the complex electromagnetic characteristics are investigated in ceramic samples of these new ferrites with composition  $\text{BaFe}_{12-2x}\text{A}_x\text{Me}_x\text{O}_{19}$  (A = Ru, Ir ; Me = Co, Zn).

### 2. EXPERIMENTS

#### 2.1 Elaboration conditions

Polycrystalline hexaferrites with composition  $\text{BaFe}_{12-2x}\text{A}_x\text{Me}_x\text{O}_{19}$  (A = Ru, Ir ; Me = Co, Zn) were prepared by standart ceramic techniques for  $x$  varying from 0 to 1.

In a first stage sintered samples of  $\text{M}(\text{Ru-Zn})_x$  substituted hexaferrites have been obtained with  $x = 0, 0.1, 0.2, 0.3, 0.4, 0.6, 0.8, 1.0$ . Stoichiometric amounts of dried  $\text{BaCO}_3$ ,  $\alpha\text{-Fe}_2\text{O}_3$ ,  $\text{RuO}_2$  and  $\text{ZnO}$  raw materials have been mixed, grinded in a ball mill, calcinated at  $850^\circ\text{C}$  to allow the decarbonation reaction, and finally compressed into pellets to be sintered at  $1275^\circ\text{C}$  for 24 h under air. In these samples particles were not oriented and the density values was included between 85% and 90% of the single crystal one. Chemical composition has been controlled by means of microprobe and X-ray analysis.

In a second time, in order to improve the electromagnetic characteristics of the ceramic samples, we have tried both to increase their density values and to produce textured samples in which the c-axis of each grain is perpendicular to the surface of the sample. Besides samples have been sintered under  $\text{O}_2$  atmosphere to avoid the "evaporation" of the Ru and Ir cations. Among the different tested processes to increase density, hot pressure at  $1200^\circ\text{C}$  before sintering has allowed us to reach more than 95% of the single crystal density. Concerning textured samples, we have used acicular  $\alpha\text{-FeOOH}$  particles as a raw

material instead of the  $\alpha\text{-Fe}_2\text{O}_3$  oxide. Texture is induced by the orientation effect of these acicular particles under an axial pressure. These processes have been applied to the different compositions  $\text{BaFe}_{12-2x}\text{A}_x\text{Me}_x\text{O}_{19}$  ( $\text{A} = \text{Ru, Ir}$  ;  $\text{Me} = \text{Co, Zn}$ ).

## 2.2 Electromagnetic measurements

The complex permeability and permittivity dispersions have been investigated in the oriented and non oriented ceramic samples of  $\text{M}(\text{A-Me})$  doped hexaferrites.  $\epsilon^*$  and  $\mu^*$  have been simultaneously obtained by using the coaxial line technique in the frequency range 0.5 GHz to 18 GHz measuring the S parameters with a network analyzer. NRW [4] and also Superjv [5] procedures have been used with the APC7 standard and have given similar results. The experimental error for the complex characteristics is less than 5%.

## 3. ELECTROMAGNETIC CHARACTERISTICS

For all the studied samples the complex permittivity is roughly constant with the frequency ( $\epsilon' \cong 19$  and  $\epsilon'' \cong 2$  for the samples sintered under air ;  $\epsilon' \cong 15$  and  $\epsilon'' \cong 1$  for the samples sintered under  $\text{O}_2$ ). The stoichiometry in oxygen reduces both the electric polarization and the conductivity of the samples. As a general behaviour we note  $\epsilon^*$  gets higher with the substitution ratio  $x$  which is to be linked with an increasing lack of stoichiometry.

The permeability spectra reveal these hexaferrites present a damped FMR in the GHz range. Figure 1 shows the  $\mu^*$  spectra of  $\text{M}(\text{Ru-Co})$  doped hexaferrites with  $x$  varying from 0.2 to 0.8. These measurements have been performed on oriented ceramic samples whose density was only about 90% of the single crystal one. The other  $\text{M}(\text{A-Me})$  substituted hexaferrites have a similar behaviour. As expected the FMR frequency is very high for a low substitution ratio, decreases with  $x$  as the axial anisotropy field gets lower, reaches a minimum when this axial field disappears, then increases again with the in-plane anisotropy field. Magnetic losses ( $\mu''$ ) are the highest ( $\approx 5-6$ ) when the FMR frequency reaches a minimum (of about 2 GHz). The corresponding substitution ratio is about 0.4 for  $\text{M}(\text{Ru-Me})$ , 0.5 for  $\text{M}(\text{Ir-Zn})$ , and 0.6 for  $\text{M}(\text{Ir-Co})$ . Note these values are in good agreement with other magnetic and neutron studies of  $\text{M}(\text{A-Me})$  doped ferrites [2, 3].

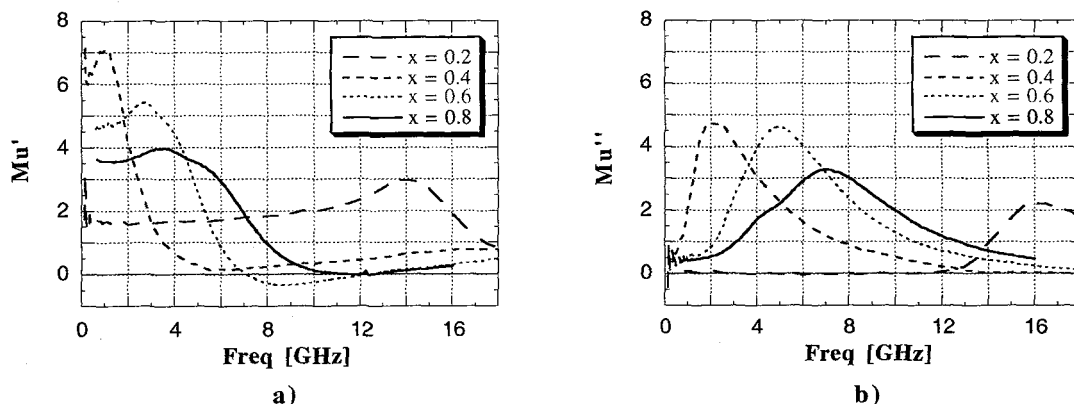


Figure 1 : Complex permeability spectra of  $\text{BaM}(\text{Ru-Co})$  oriented ceramic samples with  $x = 0.2 / 0.4 / 0.6 / 0.8$   
 a) real part of the permeability      b) imaginary part of the permeability

Oriented and hot pressed samples have not obviously increased the measured permeabilities. This behaviour is due to several factors. First the orientation effect in the samples is perfectible (the mean dispersion is about  $10^\circ$ ). Secondly in the hot pressed samples we have noted a smaller grain size (only 10  $\mu\text{m}$  on average) and stronger internal stresses. Other factors as local chemical and crystalline defaults probably limit the permeability values. A conclusion can be drawn that microstructural parameters have a predominant influence and only a very good orientation, a better chemical homogeneity and larger grain sizes may significantly increase the permeability.

The  $\text{M}(\text{A-Me})$  doped hexaferrite samples presents equal or higher permeabilities than non oriented  $\text{M}(\text{Ti-Co})$  samples with order temperatures 150°C higher on average.

## REFERENCES

- [1] A. Tauber, J.A. Kohn, R.O. Savage, J. of Appl. Phys. **14** (1963) 1265
- [2] E. Brando, Thesis of Nat. Polytech. Inst. of Grenoble (1995)
- [3] E. Brando, H. Vincent, "Synthesis, X-ray, neutron and magnetic studies of new M-hexaferrites", Proc. ICF7 (1996) to be published
- [4] W.B. Weir, Proc. IEEE **62** (1974) 33-36
- [5] N. Belhadj-Tahar, A. Fourier-Lamer, H. de Chanterac, IEEE Trans. on Micro. Th. and Tech. **38** (1990) 1-7