Investigations of a Ferrimagnetic Spinel-Garnet Composite System

N. Kumar, P. Venugopal Reddy, P. Kishan

► To cite this version:

N. Kumar, P. Venugopal Reddy, P. Kishan. Investigations of a Ferrimagnetic Spinel-Garnet Composite System. Journal de Physique IV Proceedings, 1997, 07 (C1), pp.C1-551-C1-552. 10.1051/jp4:19971227 . jpa-00254900

HAL Id: jpa-00254900

<https://hal.science/jpa-00254900>

Submitted on 1 Jan 1997

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Investigations of a Ferrimagnetic Spinel-Garnet Composite System

N. Kumar, P. Venugopal Reddy* and P. Kishan*

Solid State Physics Laboratory, Lucknow Road, Delhi - 110054, India

* Department of Physics, Osmania University, Hyderabad - 500007, India

Abstract: A composite system comprising of two ferrimagnetic oxide compounds, a spinel and a garnet, has been investigated. A series of materials with varying proportion of the contents of magnesium manganese ferrite and yttrium iron garnet, both of quality suitable for microwave applications, have been prepared and studied in relation to the parent materials. Results obtained from examination of samples by XRD and EPMA coupled with electrical and magnetic measurements have been used to understand the observed behaviour.

1. INTRODUCTION

Ferrimagnetic oxide materials having the spinel and garnet structures are extensively employed in a variety of components operating in microwave frequency ranges. Magnesium-manganese ferrites and yttrium-iron garnets are among the families of such materials which are being used for almost four decades. Substitutions of different metal ions in the basic ferrite/garnet chemical formula unit are exploited to tailor and control the properties of the material. Composites of magnetic oxides with other dielectric materials have been of interest to researchers working in this field[1,2]. System of the type described in this paper has not been found in literature. The aim of the present work is to prepare the spinel-garnet composites containing the individual compounds in different proportions and study their properties in relation to the parent materials to find out whether their salient features could be exploited in the composites.

2. MATERIALS AND METHODS

Ferrite of chemical composition $\text{Mg}_{0.9}\text{Cu}_{0.1}\text{Mn}_{0.1}\text{Fe}_{1.8}\text{O}_4$ (MF) and garnet of $\text{Y}_3\text{Mn}_{0.03}\text{Fe}_{4.92}\text{O}_{12}$ (YIG) were prepared separately by standard ceramic methods. AR grade oxide/carbonate of Mg, Mn and Cu were used while Fe_2O_3 of purity 3N+ and Y_2O_3 of 4N+ were employed. The mixed raw materials were repeatedly ball-milled and calcined. The calcining temperatures used were 1000 and 1100 °C respectively for MF and YIG powders. From XRD patterns of the calcined powders almost complete formation of the desired spinel phase was observed while the garnet phase formation was not so. Samples in the form of a disc and a toroid from both the powders were pressed and sintered at 1275 and 1450 °C for 4 hours. Dense samples so obtained were tested by XRD and for other properties. The measurements indicated the formation of single phase material with desired properties, (samples C1 and C13 listed in Table I). The composites of MF and YIG were prepared by thoroughly mixing the two sets of calcined powders in ball mills in different weight ratio (marked C2 to C12, Table I). Samples were fired in the range 1275 to 1450 °C.

All samples were analysed by XRD. C1 and C13 samples had better than 96% and 99% of their respective theoretical densities. Other samples were also estimated to have similar densities. Measurements of properties like saturation magnetisation (4 π Ms), Curie temperature (T_c), dc resistivity (ρ_{dc}) initial permeability (μ_i), 3dB resonance linewidth (ΔH) at X-band and hysteresis loop parameters were carried out using standard test methods. Some of the samples were also examined by CAMECA Electron Probe Micro Analyser (EPMA) for observing microstructural distribution of the phases present.

3. RESULTS AND DISCUSSION

From the XRD analysis of the samples it is observed that as the content of YIG in MF is increased the diffraction lines corresponding to YIG start appearing in the patterns of samples from C5 onwards. The samples C10 to C12 show only the lines of the garnet phase. However, C5 to C9 contain some extra lines which were identified as belonging to perovskite yttrium-orthoferrite (YFeO₃) phase. These lines are quite strong in the samples C6 to C8. It is reported[3] that the free energy

of formation of YFeO_3 is at least one order of magnitude lower than that of garnet and YFeO_3 is found as an intermediate product in this process of preparation of YIG [4]. It is observed here that when the MF content is substantial both orthoferrite and garnet phases get formed in the samples. In YIG rich samples (C10 to C12) perovskite phase is not observed, may be due to its small content. At the sintering temperatures employed here complete conversion of YFeO_3 to YIG phase was expected.

Table I. Properties of the samples

wt. ratio (YIG:MF)	C1 0:100	C2 1:99	C3 2:98	C4 5:95	C5 10:90	C6 25:75	C7 50:50	C8 75:25	C9 90:10	C10 95:5	C11 98:2	C12 99:1	C13 100:0
$4\pi\text{Ms}$ (Gauss)	2320	2300	2200	2190	2125	1940	1570	1720	1850	1850	1910	1830	1790
T_c ($^{\circ}\text{C}$)	330	330	335	337	335	355	> 350	> 350	> 350	278	280	278	280
ΔH (Oe)	300	320	-	300	400	640	-	-	470	60	50	50	50
ρ_{dc} ($\Omega\text{-cm}$)	1×10^8	2×10^8	1×10^8	7×10^7	2×10^7	2×10^6	4×10^3	2×10^2	2×10^0	$>10^{-11}$	$>10^{-11}$	$>10^{-11}$	$>10^{-11}$
μ_i (at 1kHz)	21	18	18	17	20	22	18	118	124	140	113	-	110

Fig. 1. Elemental scans for sample C6. Regions appearing in white contrast depict Y in (a) and Mg in (b).

From table I it is noted that the samples near the ends of the series largely follow the behaviour of parent members C1 and C13. Depending upon the relative concentrations the small content of the second phase is embedded in the major phase in the form of small grains. This is inferred from the microphotographs taken on samples using EPMA technique. The elemental maps of yttrium and magnesium is shown in Fig.1 for the sample C6. The Mg and Y regions representing MF and YIG, respectively, are complementary to each other. The grain size is estimated to be below 5 μm . In the micrographs YFeO_3 is not distinguishable from the YIG as both appear as Y containing area.

Fig. 2. BH loop tracings

The other properties show a notable departure from parent materials for samples with > 5 wt % of the other phase. $4\pi\text{Ms}$ values become lower because of the contribution from YFeO_3 , which has a low $4\pi\text{Ms}$ value. Increase in ΔH for intermediate samples is expected due to presence of the other phases. The ρ_{dc} of samples near the ends of the series are close to the values of the parents, but the C7 and C8 samples have quite low values. The formation of some Fe_3O_4 cannot be ruled out which could have led to this low resistivity values. The high T_c values observed for these samples could also be a consequence of this. The μ_i value exhibits an abrupt change after C7. The hysteresis loop behaviour varies a lot from a highly rectangular one for MF ($R \sim 0.9$ and $H_c \sim 2.0$ Oe) to a narrow loop for YIG ($R \sim 0.65$ and $H_c \sim 0.50$ Oe). Fig. 2 shows the tracings for some typical samples.

REFERENCES

[1] Boomgoard JVD, Terrel D R, Born RAJ and Giller HFJI, J. Mater. Sci., 9(1974)1705.
[2] Suryanarayana SV, Bull. Mater. Sci., 17(1994)1259.
[3] Piekarczyk W, Weppner W and Rabenau A, Mater. Res. Bull., 13(1978)1077.
[4] Thachenko EV, Shapovalov A G and Pozdnikov IP, J. Appl. Chem.(USSR), 52(1979)1088.