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## CHAPTER - II

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## PREPARATION AND CHARACTERIZATION

### INTRODUCTION

The process leading to the formation of ferrites from their component oxides belong to the class of solid state reactions in which the reaction between two solid components results in the formation of a third solid component. Generally, the components here are divalent metal oxides reacting with ferric oxide to form ferrite.

The investigation of solid state reactions leading to ferrite formation began as early as 1909 by Hilpert<sup>1</sup> who recognised the importance of these magnetic materials. Hedvall<sup>2</sup>, Tammann<sup>3</sup>, Jandar<sup>4</sup> and others contributed much to the understanding of ferritic reactions. Though ferrite technology was advanced greatly in the thirties by group of scientists from Japan<sup>5</sup> and U.S.A.<sup>6</sup>, the modern theory of preparation of spinel ferrite was first introduced by Wagner<sup>7</sup> and further developed by Schmalzried<sup>8</sup> and Hauffe<sup>9</sup>. The mechanism of solid state reactions in the formation of spinel ferrites involves the diffusion of divalent metal oxide and ferric oxide into each other.

The ceramic method is most widely used to prepare the

ferrites with desired chemical composition, shape and microstructure. This method is relatively simpler and cheap as compared to hot pressing and other techniques.

The properties of ferrites are consequences of their internal structures. They are very much sensitive to preparation condition and chemical structure and highly influenced by heat treatment, crystal structure and cation distribution. The grain size, porosity, density and orientation of grains are some of the other factors which play very important role in deciding their properties. The properties also depend upon the purity of starting compound, stoichiometry, physico-chemical history and chemical inhomogeneity etc.

In this chapter different methods of ferrite preparation are discussed, with a slightly more emphasis on ceramic technique. The details on the development of microstructure such as size, shape and orientation of grain, porosity etc are also included.

## 2.1 METHODS OF PREPARATION

The following are the general methods of preparing ferrite compositions.

### 2.1.a Ceramic Method

This method is most extensively used method in the production of ferrites commercially. In this method high purity divalent metal oxides having desired particle size and ferric oxide (  $\text{Fe}_2\text{O}_3$  ) are taken and weighed accurately to the requirement of molar composition. Initially, the powders are subjected to the fine mixing by wet milling process in water medium. This is carried out in a ball milling machine for a few hours. This machine consists of a chamber containing steel balls and a vibrator. The machine chamber is filled with powder of raw-material, water and then they are subjected to rapid mechanical vibrations. Later on the mixture is calcined at an elevated temperature for the formation of powder. This powder is dried and pressed into suitable shape and then finally sintered at high temperature. The method is simpler, straightforward and gives good results.

### 2.1.b Decomposition Method

Instead of using oxides as starting materials, one may start with salts such as carbonates, nitrates and oxalates. These salts are milled in the requisite proportions and then preheated in air to produce oxides by thermal decomposition.

The oxides so prepared undergo solid state reaction more readily<sup>10</sup>. The other details of this method, though similar to the oxide process, still rely upon mechanical processes used to produce uniform dispersion of the constituents.

#### 2.1.c General Procedure and Flow Chart

The general procedure of ferrite preparation constitutes the following basic steps.

2.1.c (i) Intimate mixing of starting materials

2.1.c (ii) Presintering or calcination

2.1.c (iii) Crushing the presintered powder and compacting  
to final shape

2.1.c (iv) Final sintering to get the desired product

The various stages in the ferrite preparation are represented by the flow chart (Fig.2.1)

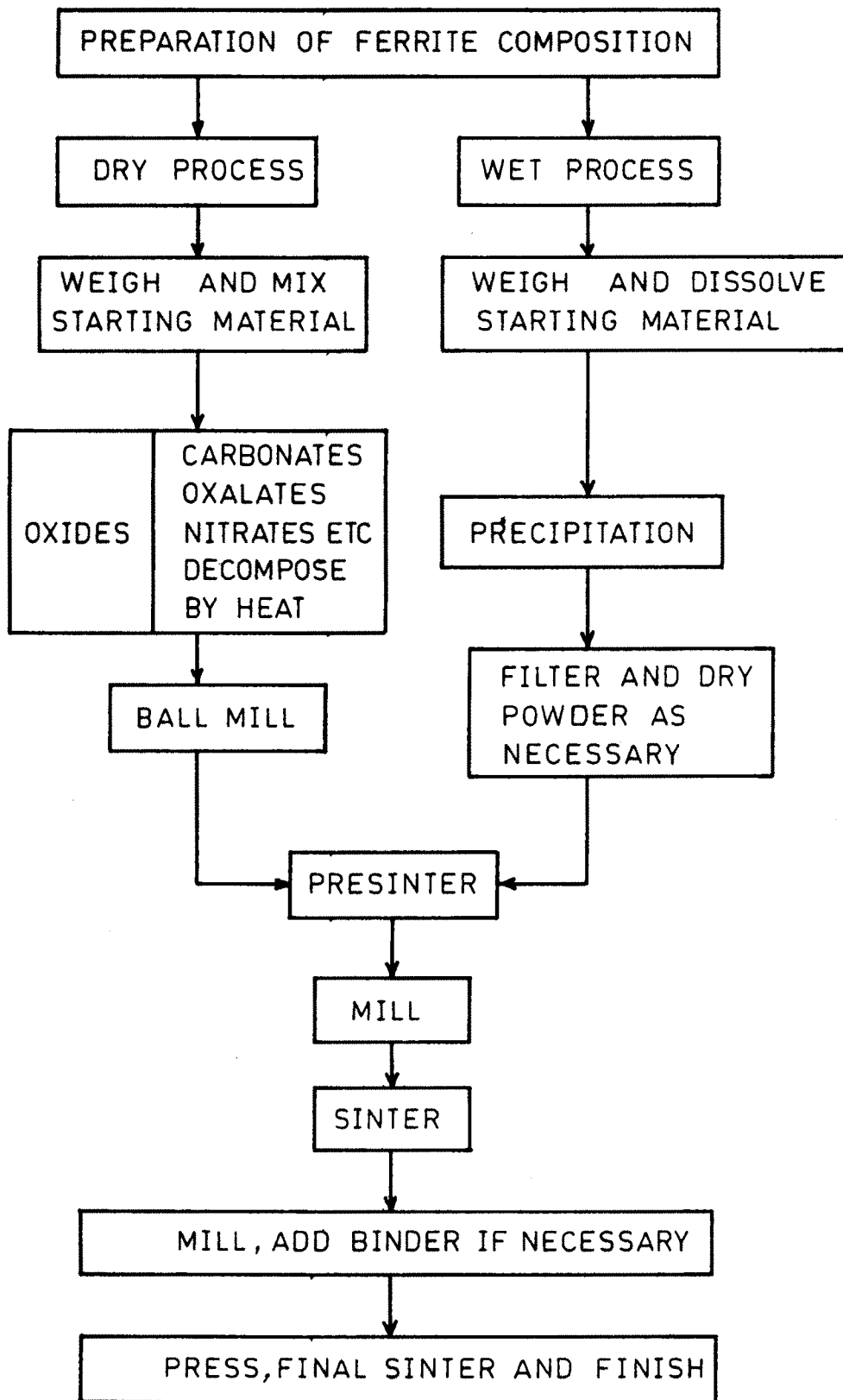


FIG.2.1 FLOW CHART OF STAGES IN THE FERRITE PREPARATION .

#### 2.1.d Chemical co-precipitation method

This method involves the process of mixing the solutions of soluble salts of divalent metals such as  $ZnCl_2$ ,  $MgSO_4$ ,  $Co(NO_3)_2$  etc and pure ferric chloride solution in a desired molar proportion. As the cations are mixed on the molecular dimensions, the method is more effective than others.

#### 2.1.d (i) Hydroxide Precipitation

This method is proposed to avoid the lengthy milling process in dry mixing. In this, attempts have been made to precipitate simultaneously, the required hydroxides from a solution intimately mixed so that the precipitate contains the required metals in the requisite proportions. Economos<sup>11</sup> was the pioneer to establish this method for the fabrication of ferrites. This method was also used by Wolf and Rodrigue<sup>12</sup> for the preparation of YIG. Some of the difficulties faced by this method are

- (i) the chemical process must be understood quantitatively to ensure the simultaneous precipitation of hydroxides. The method loses its importance if simultaneity is not achieved.

(ii) the precipitates may form in such a way that filtration becomes difficult.

(iii) it is very difficult to remove the impurities due to the absorption of sodium ions by the precipitate.

However Sato and his co-workers<sup>13,14</sup> prepared ultrafine spinel ferrites by this method and studied their properties.

#### 2.1.d (ii) Oxalate Precipitation

Precipitation of metallic oxides is preferable on account of similarity in crystal structure of metal oxalates, using ammonium oxalate which does not leave any trace of residue after ignition. Therefore precipitation tends to produce mixed crystals containing metallic cations in the proportion in which they were present in the solution. Hence the mixing in the correct ratios can be achieved on a molecular scale. Mixed crystals do not form uniformly if precipitation occurs at widely different rates. However careful calcining of precipitate at a particular temperature yields a ferrite with an average particle size less than 1 micron. This method was used by LinBo and ZhaoLeyi<sup>15</sup> to prepare Mn-Zn ferrite.



### 2.1.d (iii) Chelate Precipitation

The organo metallic compounds are called by a common name chelates. Some of these compounds are unstable and insoluble in water. However they can be co-precipitated and reduced to ferrites after calcining. Busev and others<sup>16</sup> suggested a method of mixing ferrite powders by co-precipitation of compounds with B-hydroxy-quinoline, followed by roasting. They reported that the method gives the ferrite powders of constant composition and permit regulations of dimensions of the precipitated particles.

## 2.2 SINTERING

Sintering is the most common technological process of integrating and manufacturing solid materials. It involves the process of growth of interfaces between adjacent particles, spheroidization and elimination of pores in a powder compact. It is carried out in two stages.

### 2.2.a Presintering

The purpose of presintering is to decompose higher oxides and carbonates, that reduce with evolution of gas in the final sintering process. It assists in homogenization of material and reduces the variations in the composition of

raw-materials. Also it is useful to control the shrinkage of the material during final sintering. The raw-materials partly react to form final products and the reactivity depends upon the reactivity of the components and presintering temperature<sup>17</sup>. The most critical region of temperature is 60°C to 350°C within which most of the moisture and organics get volatilised and burnt out.

The solid-state reaction is a diffusion process whose rate depends upon the temperature. The correct partial pressure of oxygen in the furnace has to be maintained during the firing in order to retain the stoichiometry and valence states. Otherwise these will be destroyed during the cooling period.

The effect of presintering on microstructure and initial permeability is observed in Mn-Zn ferrites<sup>18</sup>.

### 2.2.b Final Sintering

The final microstructure develops during this process. Under proportionate cations, the suitable microstructure can be achieved with appropriate oxygen content. The microstructure is greatly influenced by sintering temperature and time, sintering atmosphere and cooling rate. The content of

$\text{Fe}_2\text{O}_3$  depends upon the degree of oxidation and reduction of ferrites and which in turn depends upon sintering atmosphere. The cation diffusion is explained on the basis of cation vacancies. The microstructure and magnetic properties are largely affected by sintering temperature and time<sup>19</sup>.

Sintering consists of heating the compact to a temperature at which the mobility is sufficient to permit the decrease of free energy associated with grain boundaries. During sintering, densification and grain growth occur simultaneously<sup>20,21</sup> giving rise to variety of microstructure. The sintering reactivity, amount of energy available for sintering should be sufficiently high for the process to proceed. This means that the particle size of the powder must be small.

### 2.2.c Hot Pressing

The term hot pressing, as a fabrication procedure, describes the enhancement of densification by simultaneous application of external pressure with temperature<sup>22</sup>. The application of pressure serves to augment the normal driving force for densification.

The main factors affecting the hot pressing process are time, temperature, pressure, particle characteristic and environment. When carried out as a function of particle size of the starting material, keeping other factors constant, it is observed that material with finer particle size densifies more rapidly than that with the coarser particles. The concentration of  $\text{Fe}^{2+}$  ions, density, Curie point and degree of inversion increase with the rise in hot pressing pressure.

The densification by hot pressing has proved to be an important method of ceramic pressing. This technique has a significant control over the microstructure<sup>23</sup>. The selection of hot pressing is desirable for very stringent microstructure control in addition to requirement of high density.

### 2.3 ACTUAL PREPARATION OF FERRITE SAMPLES

#### 2.3.a Preparation

As oxide method is simplest and straightforward, it is selected in the present case. Ferric Oxide ( $\text{Fe}_2\text{O}_3$ ), copper oxide ( $\text{CuO}$ ) and cobalt oxide ( $\text{CoO}$ ), (all analar grade) were taken as raw-materials and weighed accurately by using microbalance to the required molar composition.

The gram molecular weights of these oxides are as follows

(1)	$\text{Fe}_2\text{O}_3$	.....	159.6942 grams
(2)	$\text{CuO}$	.....	79.5394 "
(3)	$\text{CoO}$	.....	74.9324 "

Three compositions of copper-cobalt ferrites with the general formula  $\text{Cu}_x\text{Co}_{1-x}\text{Fe}_2\text{O}_4$  were prepared

where  $x = 0, 0.4$  and  $1.0$ .

The composition weights of the powders for the preparation of about 10 grams of the final product are as under

Serial Number	Composition	Weight of $\text{CuO}$ grams	Weight of $\text{CoO}$ grams	Weight of $\text{Fe}_2\text{O}_3$ grams
1	$\text{CoFe}_2\text{O}_4$	0.0000	3.193715	6.806285
2	$\text{Cu}_{0.4}\text{Co}_{0.6}\text{Fe}_2\text{O}_4$	1.345461	1.901295	6.753244
3	$\text{CuFe}_2\text{O}_4$	3.324787	0.0000	6.675213

The compositional weights of powders were mixed physically and blended in agatemorter in acetone medium. All the three samples were presintered at about  $800^\circ\text{C}$  for 20 hours, keeping them in separate platinum crucibles. The presintered powders were subjected to hard milling process in acetone medium for

six hours. The dried powders were sieved to eliminate the large sized particles.

### 2.3.b Pellet Formation

To prepare a pellet, a small quantity of powder (roughly 1 gram) was ground in acetone by adding few drops of binder (poly-vinylacetate). The powder was subjected to a pressure of about 5 tonnes per square inch for three minutes, by keeping it in a die of 1 cm in diameter. The pellets were subjected to final sintering by keeping them on platinum sheets. The final sintering was carried out in a silicon carbide furnace, at desired constant temperature value. Calibrated chromel-Alumel thermocouple was used to measure the temperature of furnace. The final sintering was carried out according to the following procedure, so as to complete the solid state reaction.<sup>24</sup>

Sample	Sintering time in hours	Sintering temperature in °C
Co Fe <sub>2</sub> O <sub>4</sub>	20	800
"	20	900
"	30	900
Cu <sub>0.4</sub> Co <sub>0.6</sub> Fe <sub>2</sub> O <sub>4</sub>	14	800
"	20	900
"	30	900
Cu Fe <sub>2</sub> O <sub>4</sub>	20	800
"	20	900
"	30	900

The pellets were furnace cooled at the rate of about 80°C/hour. Lastly, the pellets were removed from the furnace and polished.

#### 2.4 CHARACTERIZATION BY X-RAY DIFFRACTION

The discovery of X-ray diffraction in 1912 is one of the great masterpieces of physics. Since X-rays can penetrate solids, the diffraction of X-rays provides information about the internal structure of crystals. X-ray diffraction plays an important role in ferrite research. It is used to confirm the spinel structure of ferrites and completion of solid state reaction during their preparation.

##### 2.4.a X-ray Diffraction

The simplest explanation of the observed diffraction pattern that results from the passage of X-rays through the crystal was first given by Bragg<sup>25</sup>. The diffraction maxima occur when the Bragg's law is satisfied viz.,

$$2d \sin\theta = n\lambda \quad \text{--- --- --- --- ---} \quad 2.1$$

where n = order of diffraction

$\lambda$  = wavelength of monochromatic X-rays

d = Interplanar distance

there are three principal methods of crystal structure analysis

by X-ray diffraction viz.

- (i) Laue method
- (ii) Rotating crystal method
- (iii) Powder method or Debye-Scherrer method

the powder method has been utilised in the present case.

#### 2.4.b Powder Method

The powder method was developed by Debye and Scherrer for the determination of crystal structure of finely powdered crystalline materials. In this method the finely powdered specimen is placed in a thin walled capillary, held on a movable mount at the centre of a cylindrical camera. For an arbitrary orientation of the sample with respect to incident monochromatic X-rays, some set of crystal planes satisfy the Bragg's condition. The reflected rays lie on the surface of a cone having apex angle  $2\theta$ . Other sets of planes having different spacings give rise to additional cones of different semivertical angles. These cones intercept the photographic film in concentric rings or arc of circles with a sharp circular spot at the centre due to undeflected beam. To ensure that all possible sets of planes face the incident monochromatic X-rays, the specimen is rotated slowly during exposure. In X-ray diffractometer, counter is mounted instead



of a film. The counter gives the graphical record proportional to intensity of diffracted X-rays.

The X-ray diffraction patterns were obtained from Regional Sophisticated Instrumentation Centre, University of Nagpur. All samples were in the form of fine powders and Fe  $K\alpha$  -radiations of wavelength  $1.93604 \text{ \AA}$  was used. The diffractograms were taken within the range of  $2\theta$  between  $10^\circ$  and  $85^\circ$ . For cubic system, the interplanar distance is given by

$$d = \frac{a}{(h^2+k^2+l^2)^{\frac{1}{2}}} \cdot n$$

and for tetragonal system

$$\frac{1}{d} = \left[ \frac{h^2+k^2}{a^2} + \frac{l^2}{c^2} \right]^{\frac{1}{2}} \cdot \frac{1}{n} \quad \text{--- -- -- -- --} \quad 2.2$$

where  $a$  = lattice parameter,

$c$  = lattice parameter

$h, k, l$  = miller indices of the reflecting planes

$n$  = order of the diffraction.

From equation (2.1) and (2.2), the lattice parameter can be given by an equation (for cubic system)

$$a^2 = \frac{\lambda^2 (h^2+k^2+l^2)}{4 \sin^2 \theta} \quad \text{--- -- -- -- --} \quad 2.3$$

It is possible to calculate the lattice parameter provided

we know  $\lambda$ ,  $\sin^2 \theta$  and the (hkl) values.

#### 2.4.c Indexing and Calculation of Lattice Parameter

In spinel ferrite, the XRD peak corresponding to (311) plane is most prominent and can be easily identified from the X-ray diffraction pattern. The corresponding value of  $2\theta$  is noted and then  $\theta$ ,  $\sin\theta$  and  $\sin^2\theta$  are determined. Substituting the values of  $\sin^2\theta$ , h, k, l and  $\lambda$  in equation (2.3), lattice parameter is calculated. For e.g. consider the case of cobalt ferrite  $\text{Co Fe}_2\text{O}_4$

In this, the (311) peak has  $2\theta = 45.0^\circ$ . - - - -  
(Fig.2.2)

$$\therefore \theta = 22.5^\circ, \sin\theta = 0.3827 \text{ and } \sin^2\theta = 0.1465;$$

$$= 1.93604 \text{ \AA}^\circ \text{ substituting these values in equation(2.3)}$$

$$a^2 = \frac{1.93604^2 (9+1+1)}{4 \times 0.1465}$$

$$= \frac{3.740 \times 11}{0.5860}$$

$$= \frac{41.140}{0.5860}$$

$$= 70.20$$

$$\therefore a = 8.3890 \text{ \AA}^\circ$$

Once the value of 'a' is known, the indices for other

planes can be readily decided by the following method.

Consider the second peak with  $2\theta = 23.11^\circ$  (Fig.2.2)

$$\therefore \theta = 11.555^\circ ; \sin\theta = 0.1997 ; \sin^2\theta = 0.03988$$

from equation (2.3)

$$\begin{aligned}(h^2+k^2+l^2) &= \frac{4a^2 \sin\theta}{\lambda^2} \\ &= \frac{4 \times 8.3543^2 \times 0.1997^2}{1.93604^2} \\ &= 2.9879 \approx 3\end{aligned}$$

$$\therefore (hkl) = (111)$$

Similarly all other indices may be decided.

For a tetragonal system, the diffraction formula is

$$\sin^2\theta = \frac{\lambda^2}{4} \left[ \frac{h^2+k^2}{a^2} + \frac{l^2}{c^2} \right] \quad \dots (2.4)$$

In order to calculate the lattice parameter 'a' we take the (400) index plane and to calculate the lattice parameter 'c' we take the (004) index plane. The ratio of  $\frac{c}{a}$  is calculated and it is always greater than one. The indexing is done by matching the value of 'd' for a particular plane with the observed standard value of 'd'. If they match well, the indexing for a particular plane is supposed to be correct.

Calculation of lattice parameter 'a' for a tetragonal ferrite

for a peak with  $2\theta = 55.95^\circ$  --- Fig (2.4)

$\theta = 27.975^\circ$ ,  $\sin\theta = 0.4691$   $\sin^2\theta = 0.2200$ .

$(hkl) = (400)$  ;  $\lambda = 1.93604 \text{ \AA}$

Since  $h = 4$ ,  $k = 0$  and  $l = 0$ , from equation (2.4) we get

$$\sin^2\theta = \frac{h^2 \lambda^2}{4a^2}$$

$$\therefore a = \frac{h\lambda}{2\sin\theta} \quad \text{--- (2.5)}$$

substituting the values of  $\lambda$ ,  $\sin\theta$  and  $h$  we get

$$\begin{aligned} a &= \frac{4 \times 1.93604}{2 \times 0.4691} = \frac{2 \times 1.93604}{0.4691} \\ &= 8.2543 \text{ \AA} \end{aligned}$$

Calculation of lattice parameter 'c' for a tetragonal ferrite

for a peak with  $2\theta = 52.63^\circ$  --- (Fig.2.4)

$\theta = 26.315^\circ$ ,  $\sin\theta = 0.4433$  ;  $\sin^2\theta = 0.1965$

$(hkl) = (004)$

since  $h = 0$ ,  $k = 0$  and  $l = 4$ , from equation (2.4) we get

$$c = \frac{\lambda l}{2\sin\theta} \quad \text{--- (2.6)}$$

substituting the values of  $\lambda$ ,  $l$  and  $\sin\theta$  we get

$$\begin{aligned} c &= \frac{1.93604 \times 4}{2 \times 0.4433} \\ &= 8.7347 \text{ \AA} \end{aligned}$$

DIFFRACTION PATTERN OF  $\text{CoFe}_2\text{O}_4$  SAMPLE

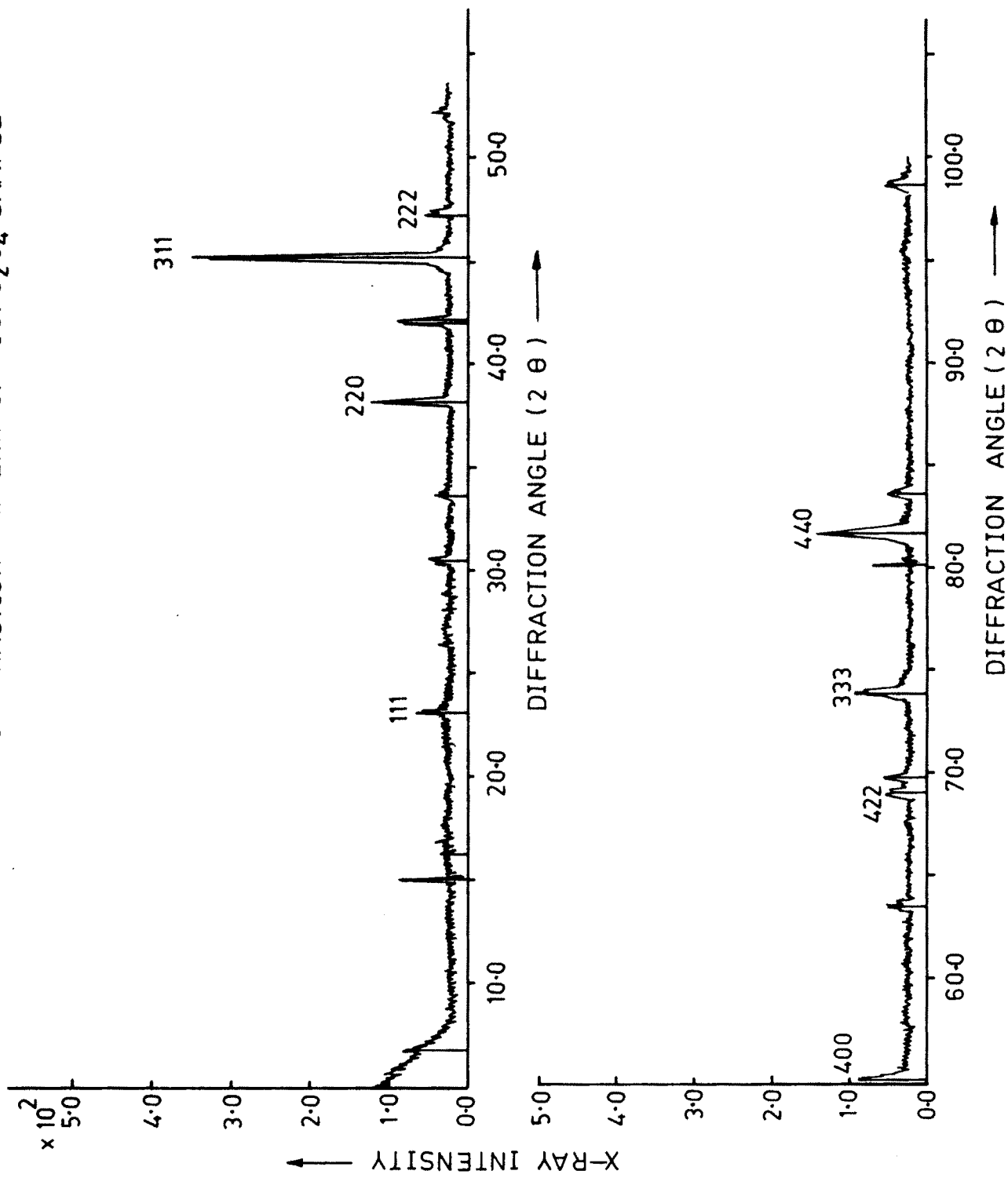


Fig. 2.2

DIFFRACTION PATTERN OF  $\text{Cu}_{0.4}\text{CO}_{0.6}\text{Fe}_2\text{O}_4$  SAMPLE

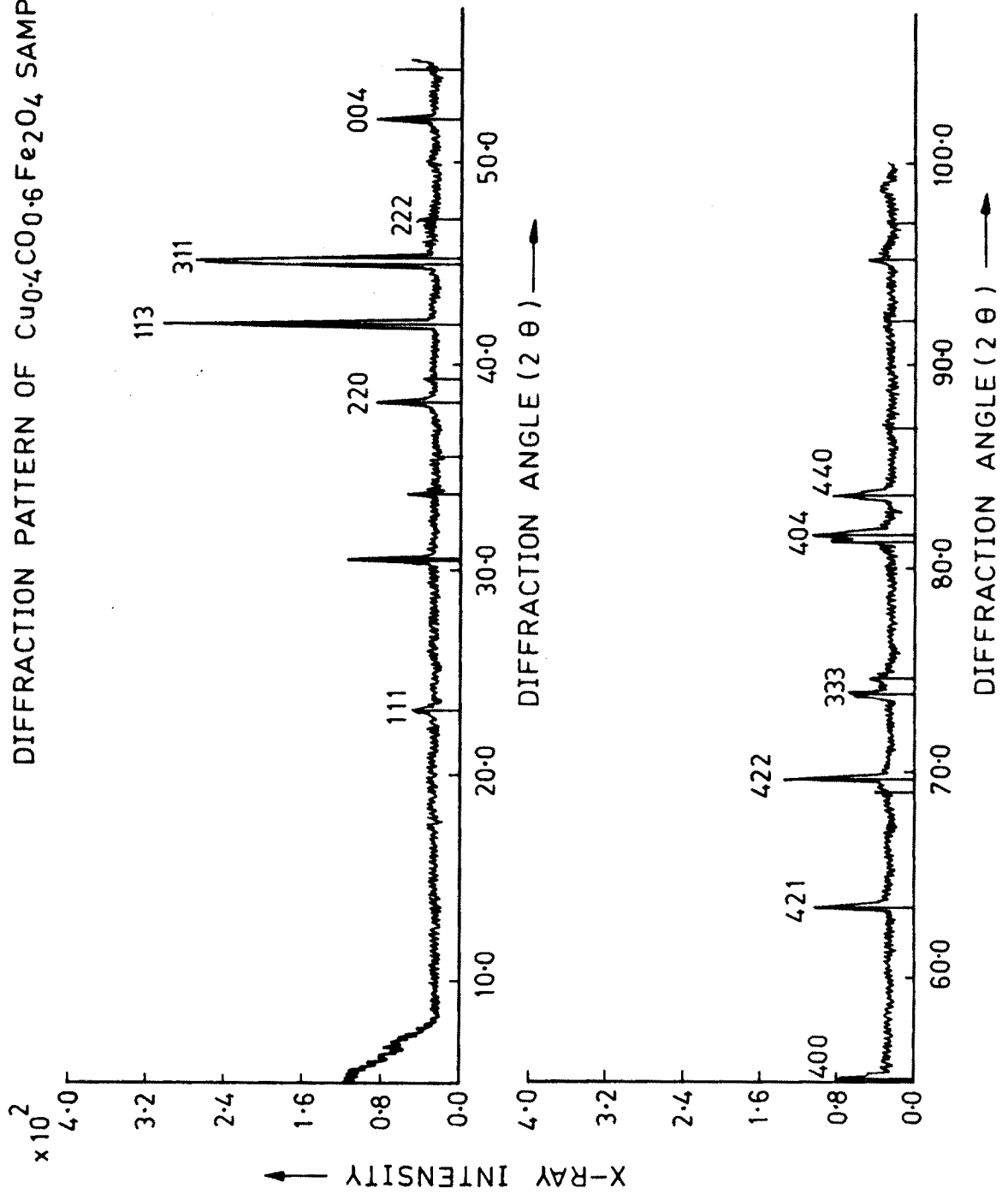


Fig. 2.3

DIFFRACTION PATTERN OF  $\text{CuFe}_2\text{O}_4$  SAMPLE

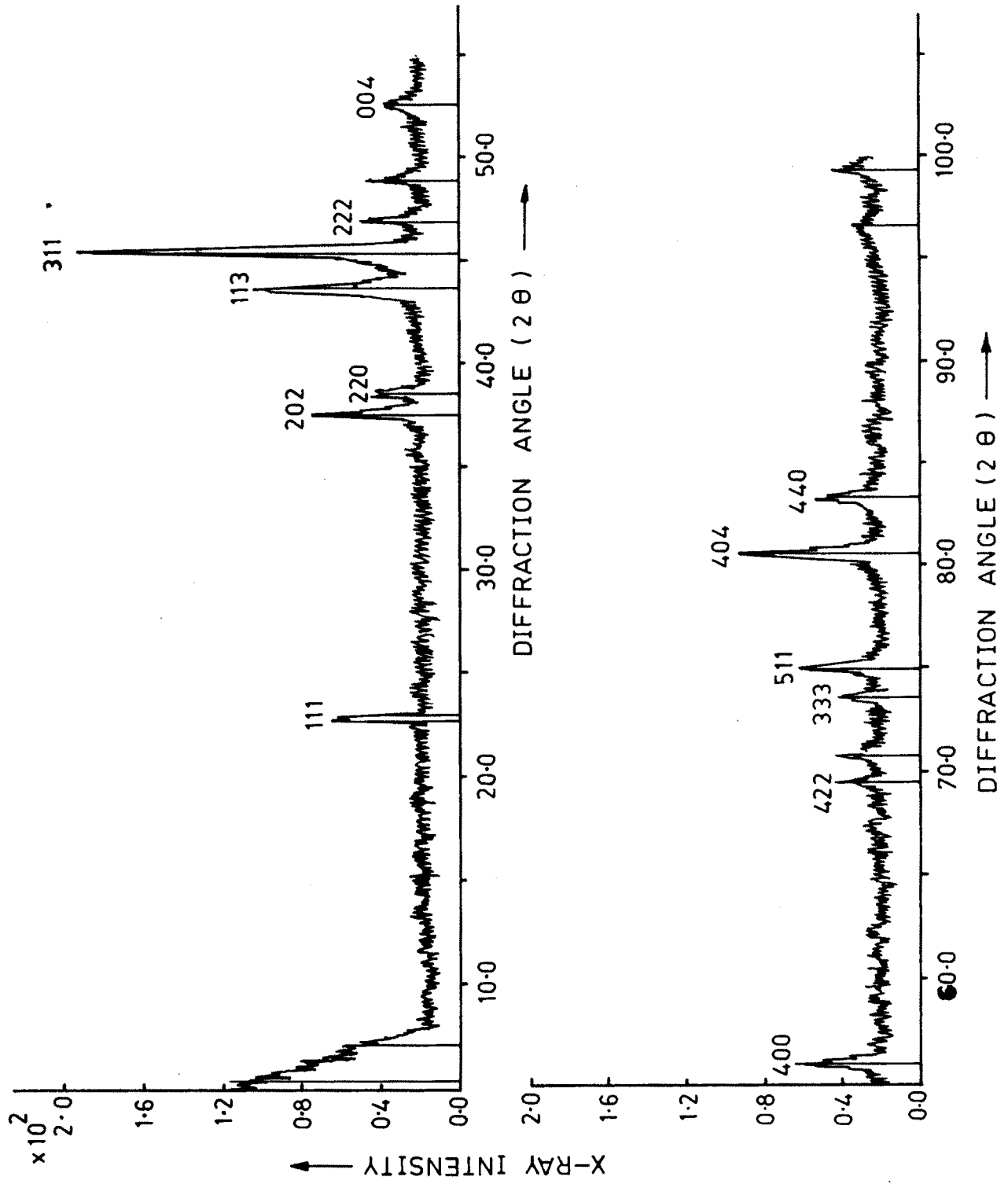


Fig. 2.4

For this particular composition  $\frac{c}{a}$  ratio comes out to be 1.058. The miller indices for various planes are given in tables 2.1, 2.2 and 2.3 for cobalt ferrite, copper-cobalt ferrite and copper-ferrite respectively.





TABLE 2.1

Sample: Co Fe<sub>2</sub>O<sub>4</sub>.      a = 8.3890 Å

$2\theta^\circ$	$\theta^\circ$	$\sin\theta$	hkl	observed d Å	calculated d Å
23.11	11.555	0.2003	111	4.8326	4.8326
38.22	19.11	0.3274	220	2.9592	2.9585
45.00	22.50	0.3827	311	2.5290	2.5189
47.20	23.60	0.4003	222	2.4187	2.4182
55.10	27.55	0.4625	400	2.0965	2.1570
69.05	34.525	0.5666	422	1.7088	1.7085
73.86	36.93	0.6009	333	1.6110	1.6063
81.70	40.85	0.6541	440	1.4800	1.4799

TABLE 2.2

Sample:  $\text{Cu}_{0.4}\text{Co}_{0.6}\text{Fe}_2\text{O}_4$   $a = 8.3744 \text{ \AA}$ ;  $c = 8.8203 \text{ \AA}$ ;  $\frac{c}{a} = 1.0533$

$2\theta^\circ$	$\theta^\circ$	$\text{Sin}\theta$	hkl	observed d $\text{ \AA}$	calculated d $\text{ \AA}$
23.14	11.57	0.2006	111	4.8265	4.9163
30.48	15.24	0.2629	210	3.6826	3.7451
33.69	11.845	0.2898	211	3.3405	3.4473
38.17	19.085	0.3270	220	2.9606	2.9608
42.07	21.035	0.3590	113	2.6969	2.6363
45.33	22.665	0.3853	311	2.5121	2.5336
47.25	23.625	0.4008	222	2.4155	2.4582
52.08	26.04	0.4390	004	2.2051	2.2051
55.08	27.54	0.4624	400	2.0936	2.0936
63.47	31.735	0.5260	421	1.8404	1.8318
69.73	34.865	0.5716	422	1.6934	1.7236
73.85	36.925	0.6008	333	1.6113	1.6388
74.60	37.30	0.6060	511	1.5974	1.6146
81.31	40.655	0.6515	404	1.4858	1.5183
81.63	40.815	0.6536	440	1.4810	1.4804

TABLE 2.3

Sample:  $\text{CuFe}_2\text{O}_4$ .  $a = 8.2543 \text{ \AA}$ ;  $c = 8.7347 \text{ \AA}$ ;  $\frac{c}{a} = 1.058$

$2\theta^\circ$	$\theta^\circ$	$\text{Sin}\theta$	hkl	observed d $\text{\AA}$	calculated d $\text{\AA}$
22.75	11.375	0.1972	111	4.8529	4.9080
37.59	18.795	0.3222	202	2.9996	3.0044
38.60	19.30	0.3305	220	2.9183	2.9290
43.59	21.795	0.3713	113	2.6054	2.6071
45.41	22.705	0.3860	311	2.5010	2.5078
46.91	23.455	0.3980	222	2.4265	2.4320
52.63	26.315	0.4433	004	2.1837	2.1837
55.95	22.975	0.4691	400	2.0636	2.0636
69.53	34.765	0.5702	422	1.7001	1.6977
73.62	36.81	0.5992	333	1.6176	1.6155
74.98	37.49	0.6083	511	1.5917	1.5914
80.62	40.31	0.6469	404	1.4999	1.4964
83.36	41.68	0.6650	440	1.4592	1.4557

## 2.5 RESULTS AND DISCUSSION

The X-ray diffraction patterns of ferrite samples, obtained by using iron filtered  $k - \alpha$  radiation are shown in Figs. 2.2, 2.3 and 2.4. For all spinel ferrites, (311) line appears to be most intense one. The interplanar distances 'd' are calculated for each line with respect to the line of maximum intensity. The calculated and observed values of 'd' spacings are tabulated in the tables 2.1, 2.2 and 2.3 along with lattice parameters. From the tables it is observed that calculated values and observed values of 'd' are in good agreement. The indexing of diffraction maxima is done by the method discussed earlier. They tally well with those expected for spinel structure<sup>26,27</sup>. Hence formation of spinel structure is confirmed.

The reflections observed in cubic structure are (111), (220), (311), (222), (400), (422), (333) and (440). While reflections observed for tetragonal structure are (111), (210), (211), (220), (113), (311), (222), (004), (400), (421), (422), (333), (511), (404) and (440).

The variation of lattice constant with cobalt concentration for copper-cobalt ferrite is shown in table 2.4.

TABLE 2.4

Sample	Lattice Constants in $\text{\AA}^\circ$	X-ray density in gram/c.c
$\text{CuFe}_2\text{O}_4$	8.2543	
	tetragonal	5.6510
	8.7347	
$\text{Cu}_{0.4}\text{Co}_{0.6}\text{Fe}_2\text{O}_4$	8.3744	
	tetragonal	5.3488
	8.8203	
$\text{CoFe}_2\text{O}_4$	8.3890 cubic	5.2900

From the table it can be seen that the lattice constants are varying linearly with cobalt concentration. The linear behaviour of lattice constant with concentration is expected from the crystal structure, because atomic radius of cobalt is more compared to that of copper. As cobalt concentration increases, cobalt ions will occupy the B sites, replacing  $\text{Fe}^{3+}$  ions. These in turn replace  $\text{Cu}^{2+}$  ions in the A sites. This results in the increase of the total length of the cube edge.

Though the X-ray diffraction studies are not enough to reveal the presence of small amounts of oxides, the absence of any extra line confirms the single phase nature of the ferrites.

For cubic structure, the cyclic sum of the miller indices  $(h+k)$ ,  $(k+l)$  and  $(l+h)$  is even integer. This is observed in case of cobalt ferrite, indicating that it is having cubic nature.

Thus, in addition to the confirmation of the spinel structure, the X-ray diffractometer study throws some light on the variation of lattice parameter with concentration of cobalt.

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