
CHAPTER - II

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PREPERATION OF FERRITES AND CHARACTERISATION BY X RAY DIFFRACTION

INTRODUCTION

The term ferrite is used to mean magnetic oxides containing iron oxide as one of the components. The ferrites are of greatest technological importance. The technological importance of ferrites lies primarily in their high resistivity. In magnetic materials a low resistivity gives rise to eddy currents losses (1). To obtain a high resistivity together with useful magnetic properties requires a thorough knowledge of chemical composition, ceramic structures, and impurities has formed the basis of intensive physical research on ferrites which has not only given rise to a better understanding of various properties of these materials, but has also contributed greatly to the better understanding of many basic magnetic phenomena.

The microstructure required to give a ceramic material a particular property can often be closely approximated by applying new understanding of the sintering process, new method of preparing the starting material (e.g. Co precipitation, freeze drying, spary drying) and new sintering process Viz. hot pressing (2). The relation between microstructure and

S E C T I O N - I

PREPERATION OF FERRITES

2.1 PREPERATION OF FERRITES

The ferrites are magnetic oxide materials and hence cannot be moulded in desired shapes by melting, as in the case of metals. The normal method of ferrite preparation is thus a ceramic method. The starting materials are mostly oxides or compounds (carbonates, oxalates or sulphates) which are easily convertible into oxides by thermal decomposition. The ferrites are prepared by the following steps.

- i) Preparation of the ferrite containing required metal ion preparation by intimate mixing of the starting materials.
- ii) Presintering or calcination,
- iii) Crushing the presintered powder and compacting to its final shape.
- iv) Final sintering to get the desired final product.

There are four different methods of preparation of ferrite materials. They are as follows,

1. Oxide method.
2. Decomposition method.
3. Hydroxide precipitation.
4. Oxalate precipitation.

2.1.1 OXIDE METHOD

This method is the easiest to use with little chemical knowledge, which is also used in the commercial production of ferrites. High purity oxide materials are taken and weighed accurately to the requirement of molar composition. At first, the powders are subjected to fine mixing by wet milling for a long period in a water or acetone medium. This is carried out in ball milling machine for a few hours. The machine consists of a small chamber containing steel balls and vibrator. The machine chamber is filled with powder of raw materials, water and then they are subjected to rapid mechanical vibrations. The filtered and dried mixture is calcinated at an elevated temperature for the formation of powder. Finally, it is dried and pressed into suitable shape and then sintered.

2.1.2 DECOMPOSITION METHOD

In the decomposition method starting materials are the salts such as carbonates, nitrates and oxalates instead of oxides. These compounds are mixed in the required proportion which may be then preheated in air to produce the oxide by thermal decomposition. The oxides which are prepared by thermal decomposition are more rapid to undergo solid state reaction(3). Further details of this method are similar to oxide method.

Flow chart for typical production process.

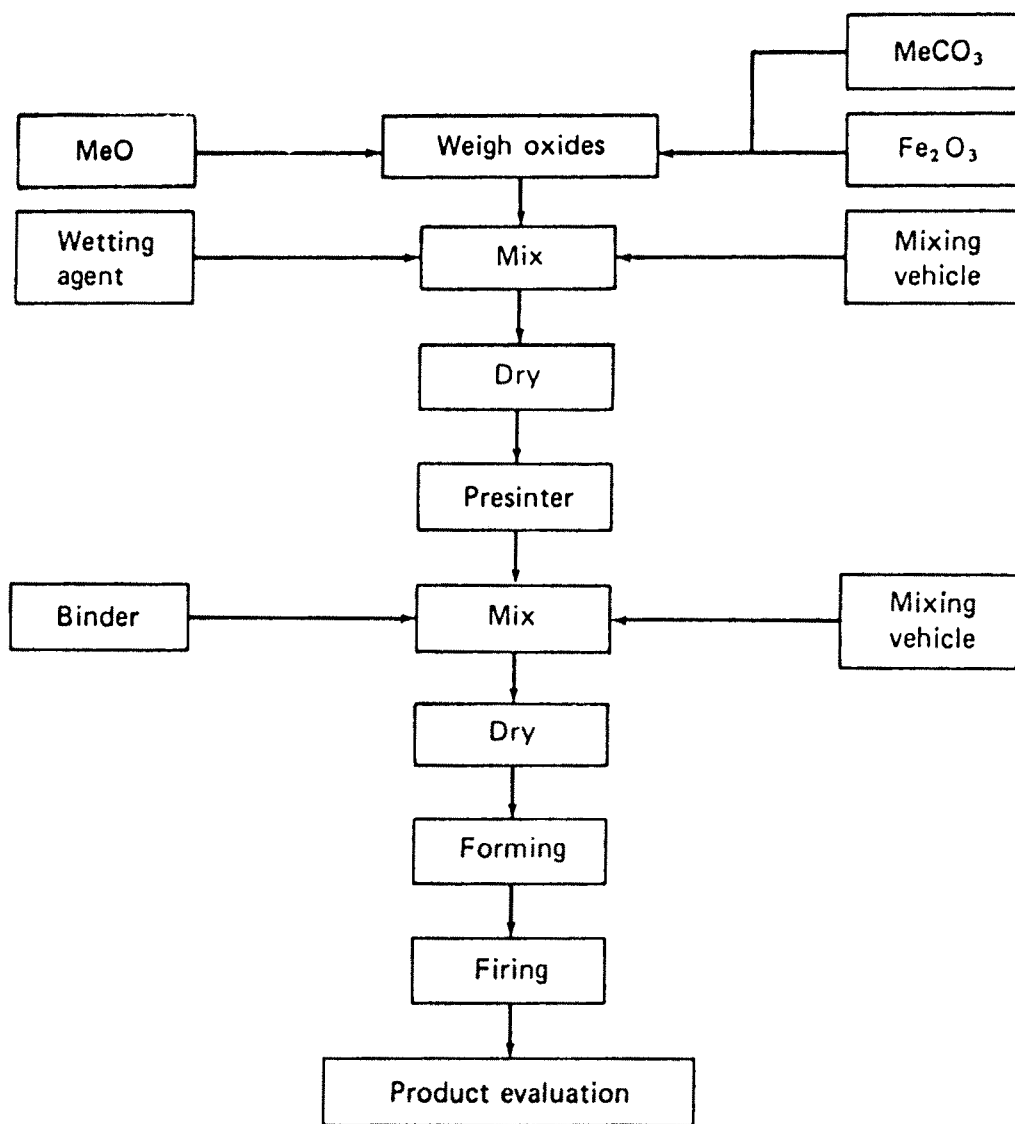


Fig. 2.1

2.1.3 HYDROXIDE PRECIPITATION METHOD

The hydroxides are precipitated simultaneously from a solution to avoid lengthy milling procedures, so that the precipitate contains the required metal ion in the correct proportions. Economus(4) established this method for ferrite preparation. Recently Robbins(5) prepared manganese zinc ferrite by chemical co precipitation with hydroxide and carbonate. The hydroxide precipitation method is also applied for the preparation of yttrium iron garnet by Wolf Rodrigues(6). The qualitative understanding of chemical process involved is essential, because the simultaneous precipitation of the hydroxides must be complete. Sato et al (7,8) prepared ultrafine spinel ferrites by this method. The Cu Ni ferrite have been prepared by Ghani in Japan using precipitation method(9).

2.1.4 OXALATE PRECIPITATION METHOD

Due to some reasons, precipitation of metallic oxalates is necessary. The precipitation can be carried out by using ammonium oxalate which does not leave any residue after heating. Due to similar crystal structure of most oxalates they can be readily precipitated after adding ammonium oxalate to their salt solutions. Here also the mixing in correct ratio can be affected upto molecular scale. However, the precipitation rate does not differ widely. After careful heating, the oxalate

gets thermally decomposed which gives water and carbon dioxide. After careful calcination at temperature of precipitation, it yields ferrites with particle size less than 15. microns, Lin Bo Zhas (10) prepared Mn-Zn ferrite by this method.

2.2 PRESINTERING OF POWDERS

Presintering is a slow solid state reaction in which reactants partly react to form final product. The amount of reaction depends on the reactivity of the components and on the presintering temperature(11). The purpose of presintering is also to decompose higher oxides and carbonates which reduce with the evolution of gas in the final sintering process. Secondly, it assists homogenising of the material and to reduce the variations in the composition of raw material. Lastly, it is necessary to control the shrinkage of the material which occurs during final sintering. The effect of the presintering process on microstructure and initial permeability is observed in Mn-Zn ferrites (12).

2.3 FINAL SINTERING

The microstructure and electromagnetic properties of memory elements prepared from thermally stable ferrites depend upon the sintering conditions of the ferrites (13)

The final microstructure develop during sintering process. Assuming the cations to be present in correct proportions, the object is to achieve a suitable microstructure together with correct oxygen content and the distribution of cations. The microstructure and magnetic properties of a ferrite may be affected by sintering temperature and sintering time (14).

2.4 HOT PRESSING

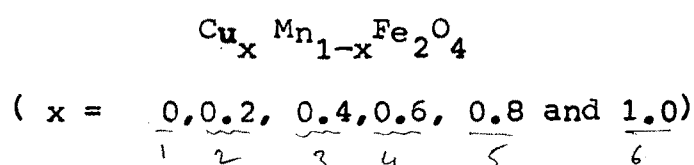
The hot pressing process entails the simultaneous application of temperature and pressure (15). The powder is enclosed in a flexible container of rubber or plastic which may be evacuated and is compacted by immersing the container in a oil bath to which the pressure is applied. The effect is to give much more uniform and higher density than conventional methods. Hot pressing is the only means of achieving a dense ferrite material at sufficiently low temperatures without grain growth. The microstructure also depends on sintering conditions during hot pressing (16). The low temperature is essential for ferrites in which one of the volatile components evaporates under normal sintering conditions.

A continuous hot pressing apparatus is described by Oudemans and Gruinties (17). Theoretically, hot pressing has been much less investigated with regard to densification mechanism. It is believed that both plastic flow and Nabarro Herring creep mechanism play a role (18)

2.5 SAMPLE PREPERATION

Though organometallic technique is very recent, it is not yet well developed. Also chemical coprecipitation and the rate of precipitation cannot be controlled. There is a possibility of picking up of alkali ions in the precipitate leading to contamination. On the other hand oxide method is the simplest and oxides in their purest form are readily available. It is observed that the selection of raw material affect the preparation of ferrites (19). Therefore the oxides in their purest form are generally chosen. We have prepared the ferrites containing divalent copper and manganese ions by standard ceramic technique.

The chemical formula for the ferrites is



①	=	0% Cu, 100% Mn
②	=	20% Cu, 80% Mn
③	=	40% " 60% "
④	=	60% " 40% "
⑤	=	80% " 20% "
⑥	=	100% " 0% "

2.5.1 STARTING MATERIALS

The iron oxide (Fe_2O_3) copper oxide (CuO) and manganese carbonate (MnCO_3) are used as raw materials. The CuO which is in the wire form is mechanically powdered in the steel agate mortar and then sieved under the standard mesh of 53 microns.

2.5.2 WEIGHING OF RAW MATERIALS

To prepare desired samples from the $\text{Cu}_x \text{Mn}_{1-x} \text{Fe}_2 \text{O}_4$ series, molecular weights of oxides and atomic weights of elements are necessary. They are as given in table 2.1

TABLE 2.1MOLECULAR AND ATOMIC WEIGHTS OF OXIDES AND ELEMENTS

Oxide/Element	Molecular weight/ Atomic weight (grams)
CuO	79.5394
MnCo ₃	114.947
Fe ₂ O ₃	159.6922
Cu	63.54
Mn	54.938
C	12.011
O	15.9994

Requisite oxides were weighed on the single pan microbalance according to their molar percentage. The necessary calculations were done by considering relevant chemical formula of the sample. Six compositions of copper manganese ferrite series are prepared in the present case.

The compositional weights of the powders are given in table 2.2

TABLE 2.2

THE COMPOSITIONAL WEIGHTS OF THE POWDERS

Sr. No.	Composition	Weight of CuO (grams)	Weight of MnCO ₃ (grams)	Weight of Fe ₂ O ₃ (grams)
1.	MnFe ₂ O ₄	-	6.2781	8.7219
2.	Cu _{0.2} Mn _{0.8} Fe ₂ O ₄	0.89184	5.1554	8.9528
3.	Cu _{0.4} Mn _{0.6} Fe ₂ O ₄	1.8322	3.9717	9.1962
4.	Cu _{0.6} Mn _{0.4} Fe ₂ O ₄	2.8251	2.7218	9.4532
5.	Cu _{0.8} Mn _{0.2} Fe ₂ O ₄	3.8751	1.4001	9.7250
6.	CuFe ₂ O ₄	4.9871	-	10.0128

Initially the powders were mechanically ground ⁱⁿ an agate mortar in acetone medium. The dried powders after grinding were filled in the separate clean platinum crucibles and kept in the furnace for presintering at 700°C to 800°C for 12 hours. They were allowed to furnace cool. Presintered powders were then subjected to milling in a ball machine in water medium for six hours. The powders were dried again and subjected to final sintering. All powders were filled in separate crucibles and were placed into the furnace. The global furnace used for final sintering is fabricated in the workshop of the physics department, Shivaji University, Kolhapur. The current flowing through the ~~silica~~ rods could be regulated so as to maintain the temperature of the furnace to desired constant value. The calibrated chromel alumel thermocouple was used for the accurate measurement of temperature. This final sintering is carried out at 1000°C for 40 hours. The samples are ^{w. cool} cooled with constant rate of fall of temperature of 80°C hours (20). The powders, after sintering were sieved with standard mesh of 53 microns. Due to sieving, the particles of very large size are eliminated.

For the preparation of a pellet the powder of about 1 gm was ground with acetone and few drops of poly-vinyl acetate, which acts as a binder, in agate mortar. This powder was then introduced into a die of 1 cm dia. and the pressure of the order of 10 tonnes/inch² was applied using hydraulic press,

for the duration of 5 minutes for each pellet. Four sets of pellets were prepared, out of which two sets are sintered at 800°C and two are sintered at 900°C for 20 hours. After sintering the pellets were polished.

2.6 ASPECT OF SINTERING AND GRAIN GROWTH

- A) Initial stage : Atomic diffusion in the beginning can quantitatively account for the initial rate of neck growth between pairs of neighbouring spheres. Grain growth is however, inhibited.
- B) Intermediate stage : When the density is at approximate 60% of its theoretical value, grain growth sets in. The structure still has a completely continuous pore phase. The density increases with the logarithm of time. The densification rate decreases slightly later on due to an increase in grain size. Lattice vacancies get discharged at the grain boundaries. At about 95% of theoretical density the pore phase becomes discontinuous (21) ending the intermediate stage.
- C) Final stage : If discontinuous grain growth occurs, a large number of closed pores get trapped inside the grains and since they are isolated from the grain boundaries cannot shrink further. The sintering is practically stopped. If discontinuous grain growth can be avoided, the last few percent of porosity are eliminated by discharging vacancies at grain boundaries. The compact then sinters to a high density, and in some cases theoretical x ray density has been achieved.

S E C T I O N - I ICHARACTERISATION BY X RAY DIFFRACTIONINTRODUCTION

Usually, to study the crystal structure of materials a well established technique of x ray diffraction is used. For the confirmation of the crystal structure and to determine the lattice parameter x ray diffraction studies of the copper manganese ferrite have been carried out. The diffraction of x ray provides information about internal structure of crystals. The simplest explanation of the observed diffraction pattern that results from the passage of x rays through the crystal was first given by Bragg(22). The diffraction maxima occur when the Bragg's law is satisfied.

2.7 CONDITION FOR X RAY DIFFRACTION

According to Bragg, diffraction is possible only when the wave length of x rays is comparable with interplaner distances (23). If the wave length of an incident monochromatic x rays is known, it is possible to find the lattice parameter by applying the Bragg's law.

Bragg's law can be written as

$$2d \sin \theta = n\lambda \quad (2.1)$$

where n = order of diffraction.

λ = wavelength of monochromatic x rays.

d = interplaner distance.

The interplaner distance for a cubic system is given by the formula,

$$d = \frac{a}{\sqrt{h^2 + k^2 + l^2}} n \quad \dots (2.2)$$

where,

n = order of diffraction

a = lattice parameter.

h, k, l = are the miller indices.

From the equation (2.1) and (2.2) the lattice parameter is given as

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

The above form of the equation is convenient to calculate the lattice parameter in the cubic system.

2.8 DIFFRACTION METHOD

There are three different x ray diffraction methods in use.

- i) Laue method.
- ii) Rotating crystal method
- iii) Rowder method.

Only the powder method is dealt with somewhat more detail here.

2.8

POWDER METHOD

Out of the diffraction methods powder method is suitable for x ray analysis of powder samples. This method of x ray diffraction was first developed by P. Debye and P. Scherrer (24) in 1916 and independently by A. W. Hull (25) in 1917. This method can be used properly to get the knowledge of structural information about the sample under investigation. In this method the specimen holder rotates about the axis of the camera in a beam of x rays, where the film is placed on the inside cylindrical surface of Debye Scherrer Camera.

A small amount of smoothly ground powder can be coated on the surface of a fine glass fibre with a glue or petroleum jelly. The specimen is then mounted in its holder by proper alignment. Since the fine grains of the powder are randomly oriented, the reciprocal lattice vectors of all the crystallites point in all the directions. The incident monochromatic radiation finds some crystal planes which may satisfy the Bragg's diffraction condition to produce maxima. The diffracted radiation is received by the cylindrical film. In the x ray diffractometer direct graphical record is taken. The peak heights in the records are directly proportional to the intensities of diffracted radiations. Though the powder method

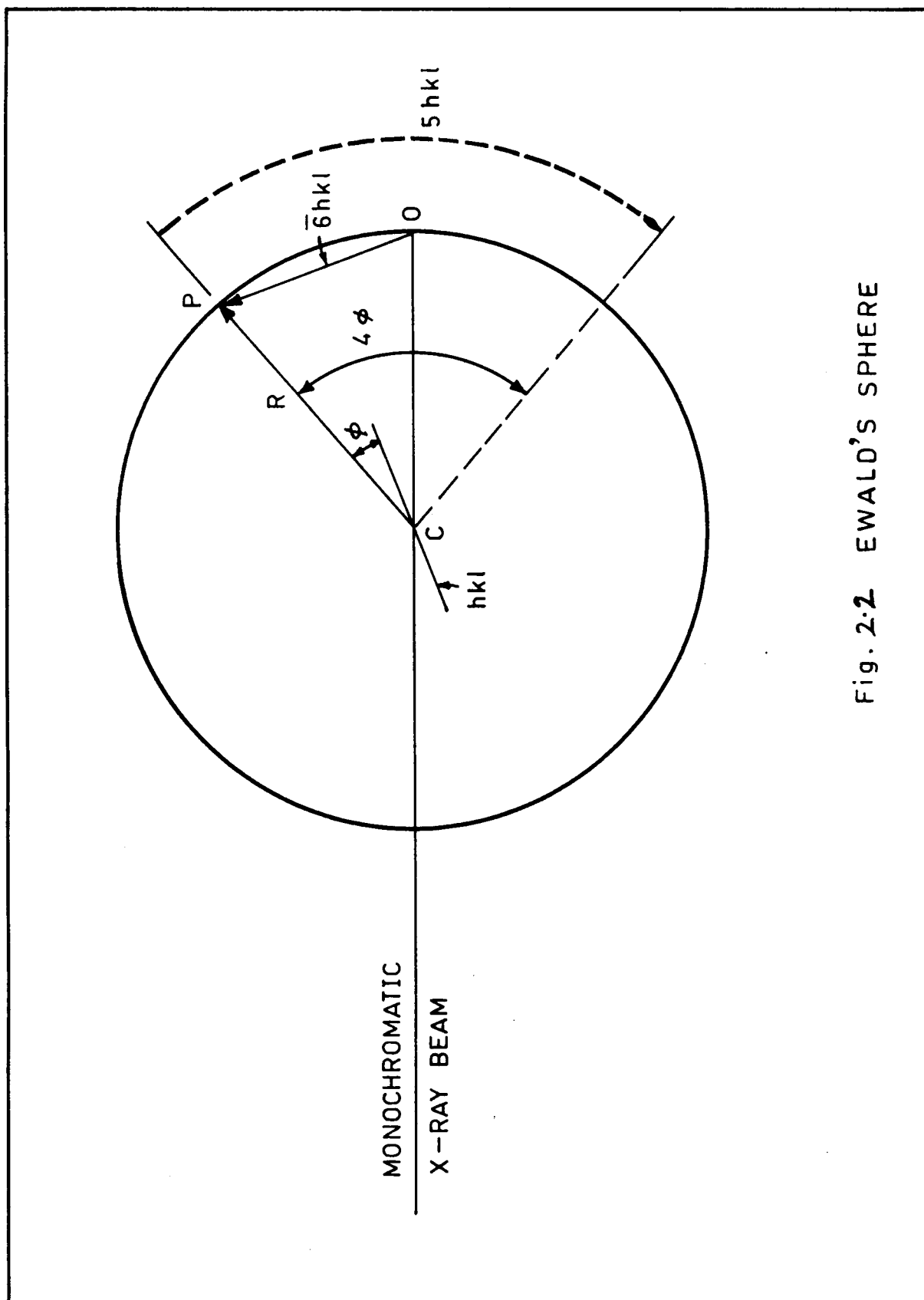


Fig. 2.2 EWALD'S SPHERE

is suitable for x ray analysis, this method does not give a quantitative measure of the intensity, as is given by the diffractometer method.

2.9 EXPERIMENTAL

The x ray diffraction pattern of ferrite system $\text{Cu}_x\text{Mn}_{1-x}\text{Fe}_2\text{O}_4$ with the different values of $x = 0.0, 0.2, 0.4, 0.6, 0.8$ and 1.0 were taken at the applied voltage of 30 KV and the current 15 mA. The facility was made available to us by the Mineralogical Institute, University of Mysore. The fine powder of the sample were used and ^{57}Fe k_α radiation having wavelength 1.93604\AA was used. The range of 2θ angles within which the diffractograms were taken was restricted from 10 to 85 degrees. The rate of angular displacement of the goniometer was adjusted to 1 deg/min.

2.10 INDEXING AND CALCULATION OF LATTICE PARAMETERS

With the help of XRD data it is possible to index the planes and calculate the lattice parameters. The procedure is as follows. Consider the case of copper-manganese ferrite, for which $2\theta = 45.7^\circ$ for (3 1 1) plane (Fig.2.3b).

$$\therefore \theta = 22.85^\circ, \sin\theta = 0.3883,$$

$$\sin^2\theta = 0.1501$$

$$\lambda = 1.93604 \text{\AA}, h = 3, k = 1, l = 1.$$

Substituting these values in equation 2.3 we get.

$$\begin{aligned}
 a^2 &= \frac{\lambda^2}{4\sin^2\theta} (h^2 + k^2 + l^2) \\
 &= \frac{(1.93604)^2 (9 + 1 + 1)}{4(0.1501)} \\
 &= \frac{(3.7482508) \times (11)}{0.6004}
 \end{aligned}$$

$$\therefore a = 8.2868655 \text{ \AA}$$

Once the value of lattice parameter is known the indices for planes can be easily obtained by adopting the following method.

Consider the case of other with $2\theta = 23.15^\circ$ (Fig. 2.3b)

$$\therefore \theta = 11.57^\circ \quad \sin\theta = 0.2006$$

and

$$\begin{aligned}
 \sin^2\theta &= 0.04024 \\
 (h^2 + k^2 + l^2) &= \frac{4a^2\sin^2\theta}{\lambda^2} \\
 &= \frac{4(8.2868655)^2(0.04024)}{(1.93604)^2} \\
 &= \frac{11.053468}{3.7482508} \\
 &= 2.948967 \approx 3
 \end{aligned}$$

$$\therefore (h, k, l) = (1 \ 1 \ 1)$$

Similarly other indices may be decided,

In case of ~~t~~ tetragonal ferrite we have to use the formula.

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

In tetragonal ferrites to calculate 'a' we take (4,0,0) as h,k,l values with its corresponding θ value from the cubic diffractogram and for 'c' we use (0,0,4) as h,k,l values with its corresponding θ value. If the 'd' value for the particular plane matches with the observed standard 'd' value indexing for particular plane will be correct.

- I) Calculation of lattice parameter 'a' for a peak with $2\theta = 54.8^\circ$ in (Fig. 2.3a), $\theta = 27.4^\circ$, $\sin \theta = 0.4602$.

$$\sin^2 \theta = 0.2278; h = 4, k = 0, l = 0$$

and

$$\lambda = 1.93604 \text{ \AA}$$

From equation (2.4) if we substitute the values we get,

$$\sin^2 \theta = \frac{\lambda^2 h^2}{4a^2} \quad (\text{for } k = 0 \text{ and } l = 0)$$

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

$$a = \frac{1.93604 \times 4}{2 \times 0.4602}$$

$$\therefore a = 8.4139069 \text{ \AA}$$

II) Calculation of lattice parameter C. In order to calculate 'C' consider the (0,0,4) peak.

For which $2\theta = 52.4^\circ$ $\theta = 26.2^\circ$

$\sin \theta = 0.4415$, Since $h = 0, k = 0, l = 4$

from equation (2,4) we get.

$$\sin^2 \theta = \frac{\lambda^2 l^2}{4c^2}$$

$$c = \frac{\lambda l}{2 \sin \theta}$$

Substituting for λ, l , and $\sin \theta$

$$c = \frac{1.93604 \times 4}{2 \times 0.4415}$$

$$c = 8.7702831 \text{ \AA}$$

For copper ferrite the ratio $c/a = 1.0423556$

TABLE 2.3

PHYSICAL DENSITY AND X RAY DENSITY DATA FOR SAMPLES SINTERED
FOR 20 HOURS

Composition	Sintering Temperature (°C)	Physical density (gms/cm ³)	X ray density (gms/cm ³)
CuFe_2O_4	800	4.8849	5.446
$\text{Cu}_{0.6}\text{Mn}_{0.4}\text{Fe}_2\text{O}_4$	800	4.1299	5.5559
$\text{Cu}_{0.4}\text{Mn}_{0.6}\text{Fe}_2\text{O}_4$	800	3.7145	5.519
CuFe_2O_4	900	4.8765	5.446
$\text{Cu}_{0.6}\text{Mn}_{0.4}\text{Fe}_2\text{O}_4$	900	4.8437	5.5559
$\text{Cu}_{0.4}\text{Mn}_{0.6}\text{Fe}_2\text{O}_4$	900	4.3777	5.519

TABLE 2.4Sample CuFe_2O_4 , Sintered at 950°C for 20 hours.

$a = 8.4139069 \text{ \AA}$

$c = 8.7702831 \text{ \AA}$

$c/a = 1.0423556$

$\lambda = 1.93604 \text{ \AA}$

Structure Tetragonal.

Obs. No.	2θ (deg.)	θ (deg.)	$\sin \theta$	(h,k,l)	Observed d (Å)	Calculated d (Å)
1.	22.7	11.35	0.1968	1 1 1	4.9136	4.9183
2.	37.2	18.6	0.3189	1 0 2	3.0317	3.0326
3.	38.0	19.0	0.3256	2 0 0	2.9702	2.9716
4.	42.2	21.65	0.3689	1 1 3	2.6210	2.6210
5.	44.6	22.3	0.3795	3 1 1	2.5484	2.5527
6.	46.3	23.15	0.3931	2 2 2	2.4597	2.4592
7.	52.4	26.2	0.4415	0 0 4	2.1902	2.2902
8.	54.8	27.4	0.4602	4 0 0	2.1013	2.1013
9.	60.0	30.0	0.5000	3 3 1	1.9340	1.9323
10.	68.0	34.0	0.5592	4 2 2	1.7293	1.7272
11.	72.3	36.15	0.5899	3 3 3	1.6392	1.6393
12.	79.2	39.6	0.6374	4 0 4	1.5117	1.5163
13	81.2	40.6	0.6508	4 4 0	1.4859	1.4858

TABLE 2.5

Sample $\text{Cu}_{0.6}\text{Mn}_{0.4}\text{Fe}_2\text{O}_4$, Sintered at 950°C for 20 hours

$$a = 8.2968655 \text{ \AA} \quad \lambda = 1.93604 \text{ \AA}$$

Structure cubic.

Obs. No.	2θ (deg.)	θ (deg.)	$\sin\theta$	(h,k,l)	Observed d(\AA)	Calculated d(\AA)
1.	23.15	11.57	0.2006	1 1 1	4.8090	4.8213
2.	38.6	19.3	0.3305	2 2 0	2.9297	2.9257
3.	45.7	22.85	0.3883	3 1 1	2.4851	2.4902
4.	47.8	23.9	0.4051	2 2 2	2.3868	2.3868
5.	55.9	27.95	0.4681	0 0 4	2.0665	2.0632
6.	70.2	35.1	0.5750	4 2 2	1.6817	1.6817
7.	75.15	37.57	0.6097	3 3 3	1.5849	1.5859
8	83.2	41.6	0.6639	4 0 0	1.4565	1.4564

TABLE 2.6

Sample $\text{Cu}_{0.4}\text{Mn}_{0.6}\text{Fe}_2\text{O}_3$, Sintered at 950°C

for 20 hours.

$a = 8.2868655 \text{ \AA}$

Structure cubic

$\lambda = 1.93604 \text{ \AA}$

Obs No.	2θ (deg)	θ (deg)	$\sin\theta$	(h,k,l)	Observed d(Å)	Calculated d(Å)
1.	23.2	11.6	0.2011	1 1 1	4.8090	4.8090
2.	38.6	19.3	0.3305	2 2 0	2.9257	2.9257
3.	45.7	22.85	0.3883	3 1 1	2.3963	2.4902
4.	50.4	25.2	0.4258	3 2 0	2.4532	2.2710
5.	56.0	28.0	0.4695	0 0 4	2.0598	2.0597
6.	70.2	35.1	0.5750	4 2 2	1.6817	1.6817
7.	75.2	37.6	0.6101	3 3 3	1.5449	1.5848
8.	83.3	41.65	0.6646	4 0 0	1.4565	1.4550

2.11 RESULTS AND DISCUSSIONS

The diffractograms for the copper manganese ferrites are shown in figs. 2, 3a, 2.3b and 2.3c. Tables 2.4, 2.5 and 2.6 represent the data on observed and calculated d values and the miller indices of diffracting planes.

The diffraction maxima are indexed in the light of the spinel structure (26). The reflections observed are (111), (220), (311), (222), (400), (422), (333), (511) and (440) for cubic structure and (111), (202), (220), (113), (311), (222), (004), (400), (422), (333), (511), (404) and (440) for tetragonal structure (27). These corresponds to allowed reflections for spinel structure. The d values are in good agreement with each other. Therefore, the formation of spinel structure is confirmed. Non appearance of any other unwanted peak shows the purity of the compound formed. The x-ray and physical density data is contained in table 2.3. The x ray density values shows that it is nearly independent of the manganese content, in the system. This may be because the Mn^{2+} is both lighter and smaller in size compared with copper and this may make the density independent of the manganese content. The actual physical densities that we have measured can be seen to differ a little and are smaller than the corresponding x ray densities. This may be because of small porosity prevalent in the pressed pellets, and sintered / As is well known this affects the electrical

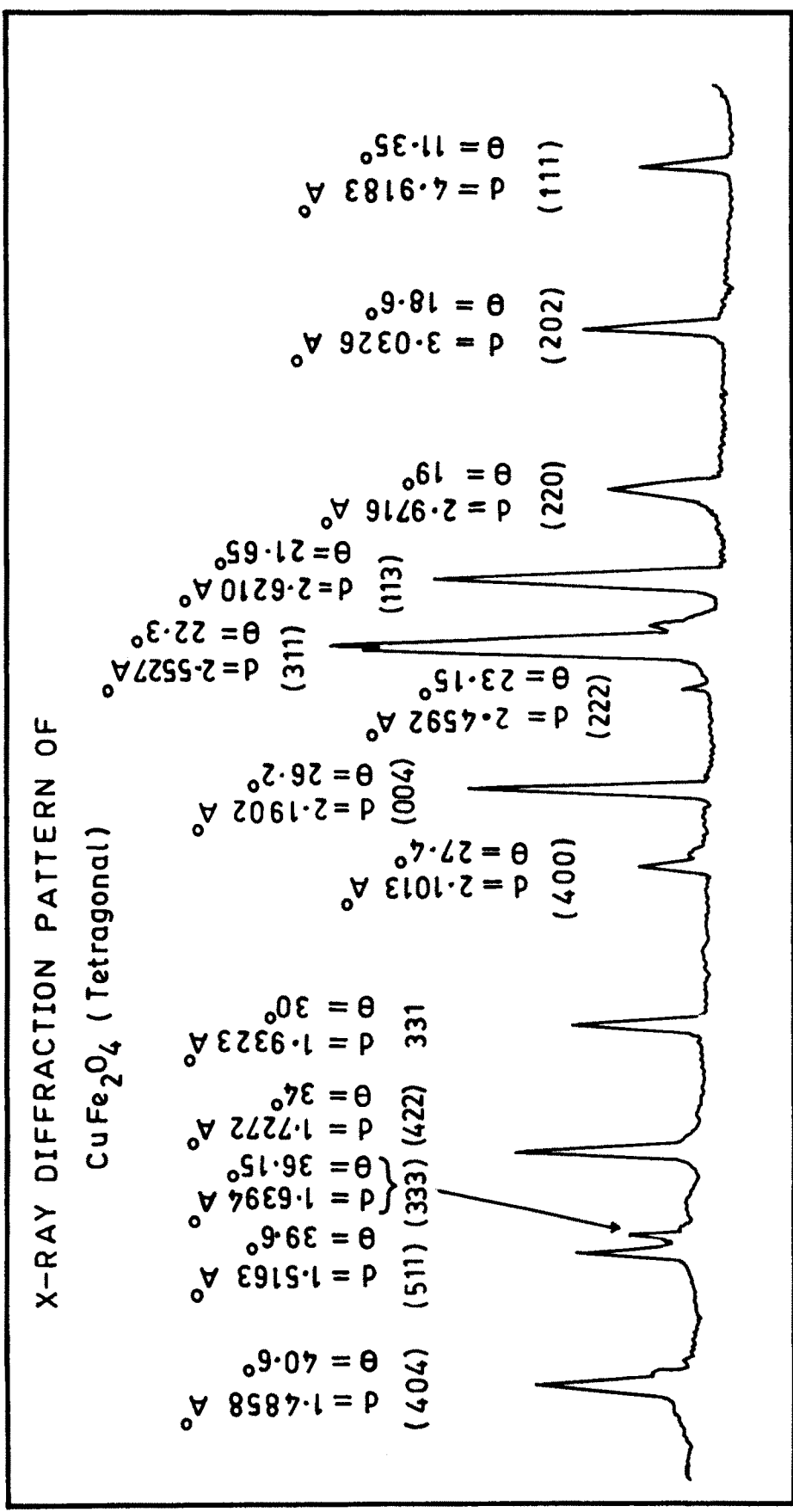


Fig. 2.3(a)

INTENSITY

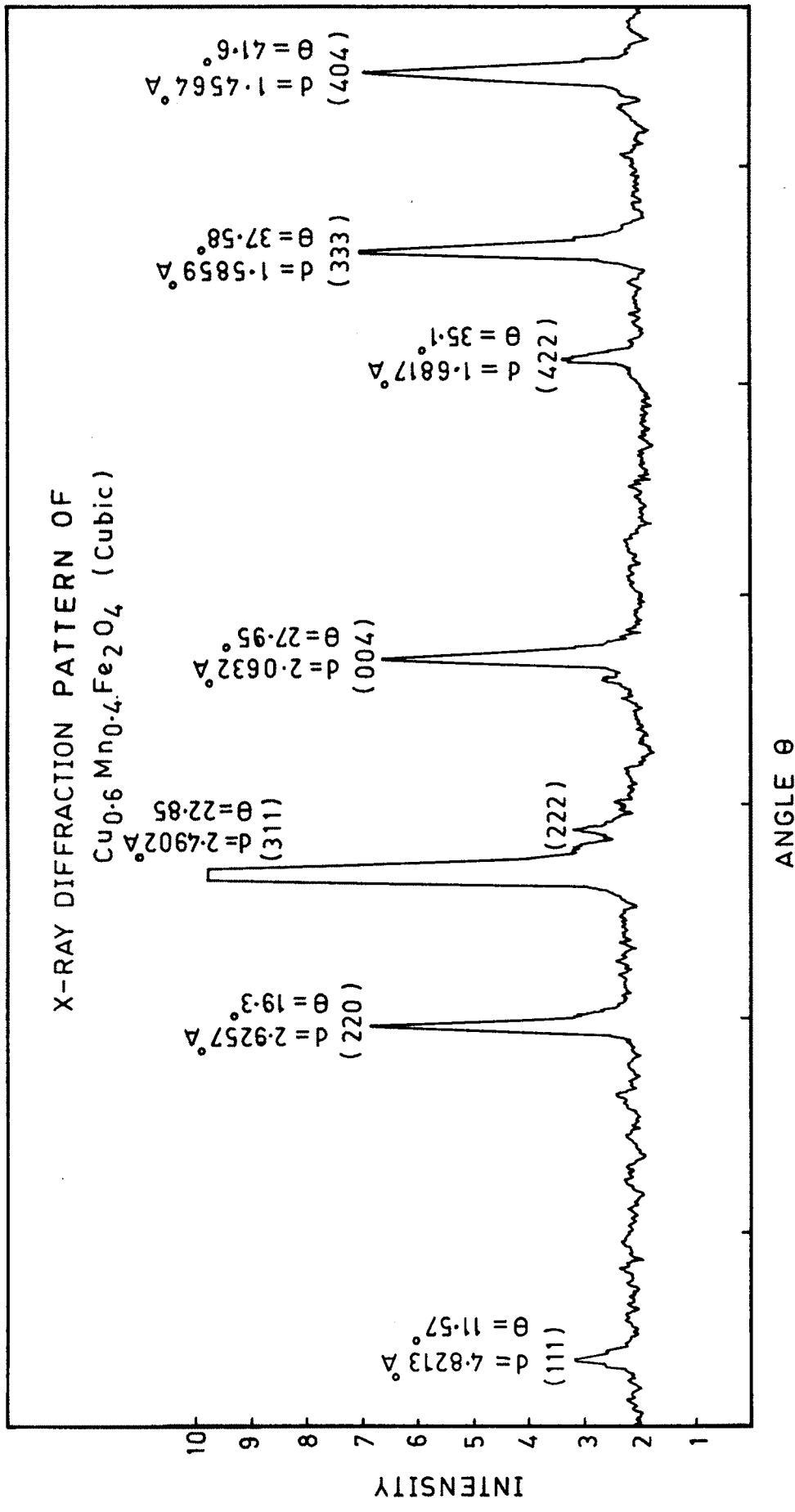
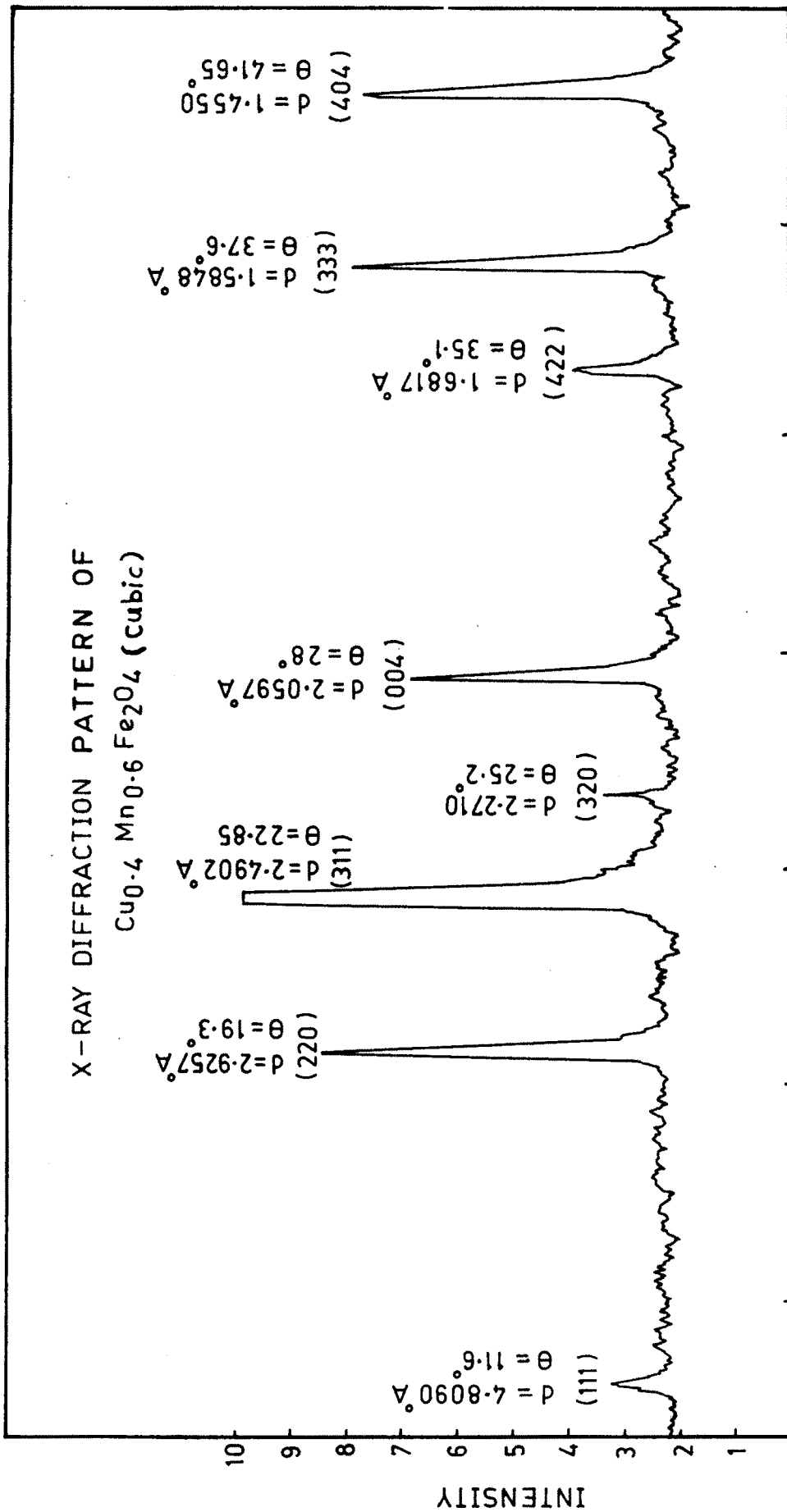


Fig. 23 b



ANGLE θ

Fig. 2.3 (C)

and magnetic properties in general and therefore, given consideration in the further chapters.

From our studies the tetragonality of copper ferrites comes out to be $c/a = 1.0423556$. Oninishi et al (28) have shown that the tetragonality of copper ferrites varies between 1.06 to 1 as the function of Cu ions increases from 0 to 0.25. The sample CuFe_2O_4 is tetragonal while for $\text{Cu}_{0.6}\text{Mn}_{0.4}\text{Fe}_2\text{O}_4$ and $\text{Cu}_{0.4}\text{Mn}_{0.6}\text{Fe}_2\text{O}_4$ are cubic.

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