6.1 CONCLUSIONS:

A systematic study was undertaken to develop and standardize a novel method of synthesis of Mn-Zn ferrite material. In this method, an attempt was made to combine the mechanical method with the chemical method to obtain the desired ferrite materials. The work was aimed at preparing a series of the ferrite materials with the general formula \( \text{Mn}_{x}\text{Zn}_{1-x}\text{Fe}_2\text{O}_4 \), where \( x = 0.40, 0.50, 0.60, 0.625, 0.65, 0.675, 0.70 \) and 0.80. Further, it was proposed to investigate the structural, the physical, the magnetic and the electrical properties of the ferrite products by using known methods.

In addition to the above, the synthesis of these ferrite materials was also carried out, with the same compositions, by using the wet chemical method. The various properties of the ferrite products, obtained by this route were studied and compared with those obtained by mechano-chemical method.

In both the methods, hydrazinium acetate was used as ligand for obtaining the precursors and a conventional mode of decomposition of the precursor was adopted.
The ferrite products obtained on decomposition of the precursors were found to yield the expected single phase Mn-Zn ferrite materials. This was confirmed from analysis of the compounds by different known methods of characterization like: the Atomic Absorption Spectroscopy (AAS), the Infrared Spectroscopy (IR), the X-ray Diffraction Spectroscopy (XRD) and the thermal analysis.

The relevant data has been presented in Chapter Three. The percentage composition of metals Mn, Zn and Fe in the ferrite products, obtained by the AAS, confirmed the formation of Mn-Zn ferrites by these novel methods. The IR spectra for the samples show typical metal-oxygen absorption bands for the ferrites. The XRD data obtained for the samples are in agreement with the reported d-values for Mn-Zn ferrites.

The particle size calculated by using the Scherer formula indicates the presence of nano sized particles in the oxide products. The calculated average crystallite sizes were in the range of 16.12 nm to 42.20 nm for the mechano-chemical method and 18.14 nm to 67.29 nm for the wet chemical method. The TEM micrographs of the samples further confirmed that these Mn-Zn ferrite materials contained nanometer sized particles. The particles were found to agglomerate due to their fine size.

Calculated lattice constant values for the Mn-Zn ferrite samples, obtained by both mechano-chemical as well as the wet chemical method are in the range (Table 3.8) of 8.4198 Å - 8.4519 Å and 8.4529 Å -
8.4687 A° respectively. These values are in agreement with the reported values for these types of ferrite materials. From the percentage porosity calculated by the known method, the samples prepared by the mechano-chemical and the wet chemical methods are found to have high porosity of 43.14 – 30.23% and 38.57-28.21% respectively (Table 3.12).

The saturation magnetization (Ms) studies for Mn-Zn ferrite samples prepared by the mechano-chemical method show (Table 4.1) the highest value of 50.91 emu/g for $\text{Mn}_{0.625}\text{Zn}_{0.375}\text{Fe}_2\text{O}_4$ among the unsintered samples; which increases to 68.59 emu/g for $\text{Mn}_{0.65}\text{Zn}_{0.35}\text{Fe}_2\text{O}_4$ (sintered at 1300°C) among the sintered samples. While for Mn-Zn ferrite samples prepared by the wet chemical method (Table 4.2), $\text{Mn}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4$ was found to have $M_s=43.66$ emu/g, the highest among unsintered samples. The value increased to 68.36 emu/g for $\text{Mn}_{0.625}\text{Zn}_{0.375}\text{Fe}_2\text{O}_4$, sintered at 1300°C, among sintered samples.

The magnetic permeability of $\text{Mn}_{(x)}\text{Zn}_{(1-x)}\text{Fe}_2\text{O}_4$ samples prepared by the mechano-chemical method were found to increase with the sintering temperature. The values were in the range of 1791 (for $\text{Mn}_{0.5}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4$ sintered at 1000°C), being lowest, and 16183 (for $\text{Mn}_{0.6}\text{Zn}_{0.4}\text{Fe}_2\text{O}_4$ sintered at 1300°C) being highest.

The relative loss factor was found to be low for all the samples sintered across all the temperatures.
The Curie temperature for a few unsintered samples was found to show superparamagnetic behavior with single domain, which gets converted to multi domain on sintering the samples above 1200\(^\circ\)C.

The resistivity variation with temperature for all the samples was found to show the general semi-conductor behaviour. The resistivity decreases with increase in sintering temperature. This is due to the increase in their particle size as can be seen from the SEM micrograph of the sintered samples at various sintering temperatures.

The thermoelectric power (TEP) studies of the samples, obtained by both the methods, indicated that the ferrite samples show n – type semi conductor behaviour for all the compositions. The Seebeck coefficient values of the samples obtained by the mechano-chemical method were in the range \(-186\) uV/K to \(-452\) uV/K. The samples prepared by the wet chemical method were found to show overall higher values of Seebeck coefficient as compared to mechano-chemical method. The values were from \(-285\) uV/k to \(-782\) uV/K.

The dielectric constant values for all the Mn-Zn ferrite samples were found to decrease with frequency and were found to increase with increasing temperature as reported for most of the ferrite samples. The transition dielectric temperature values were above 550\(^\circ\)C for most of the samples.
The dielectric loss factor was found to be very low for the unsintered as well as the sintered samples.

In the present study, high values of $M_s$, the saturation magnetization, for the unsintered ferrite powder samples were observed compared to the most of the reported values for nanoparticle ferrite samples and the hysteresis losses for the samples were found to be lower. The highest value of $M_s$ was obtained for the sample with $x=0.625$ and $x=0.70$ in the case of the samples prepared by the mechano-chemical method and the wet chemical method, respectively.

For the samples synthesized by both the above methods, the Curie temperature values were also found to be higher than the reported values for such oxides. The lattice constant ‘$a$’, the particle size, the saturation magnetization, the Curie temperature and the hysteresis loss were found to depend on the Mn concentration in the Mn-Zn ferrite samples prepared during the studies.

All the above mentioned measurements were also carried out for the bulk samples obtained by sintering of the nanoparticle samples and similar results were obtained.

Further, the study also provided more information on the behavior of the bulk samples. It was also observed that the properties of the samples depended not only on the Mn concentration, but also on the microstructure
of the samples. The microstructure of the samples was found to change with sintering temperatures. Scanning electron micrographs of the samples sintered at elevated temperatures, 1000°C, 1100°C, 1200°C and 1300°C indicated an increase in the grain size as well as in the densification of the samples. The magnetic and the electrical properties study carried out for these samples confirmed that, apart from Mn concentration in the sample, the other factors affecting these properties are the microstructure and the sintering conditions of the sample. The samples exhibit high values for saturation magnetization, low loss as mentioned earlier, and also higher magnetic permeability, higher resistivity and the dielectric constant. The saturation magnetization (68.59 emu/g), the permeability value 16183 (for mechano-chemical method), low relative loss factor ($10^2$ to $10^{-6}$), the room temperature resistivity ($10^8$ ohm-cm), and the Seebeck coefficient with negative thermo-emf indicated n-type semiconductor behaviour of these ferrites prepared by these novel methods. With hopping of electron between Fe$^{2+}$ and Fe$^{3+}$ as the mechanism of conduction in these ferrite materials, the high value of dielectric constant ($10^8$) and the extremely low loss are important parameters for Mn-Zn ferrite samples to exhibit enhanced performance.

Structural and magnetic studies of the Mn-Zn ferrite samples, prepared by employing two simple routes, revealed that the critical Mn proportion for high performance oxide material by these routes is about 0.65.
Thus, through the present study a novel mechano-chemical method of synthesis has been successfully tried and standardized. This new method yields Mn-Zn ferrite materials at lower temperatures thus minimizing the stoichiometric defects which are normally observed in high temperature synthesis. This method yields nanosized ferrite material exhibiting improved performances. Similar observations have been made for the Mn-Zn ferrite material obtained by the novel wet chemical method used for the synthesis. The bulk samples produced by sintering of this nanoparticle Mn-Zn ferrite material, prepared by both the methods, also exhibit enhanced and unique properties.

6.2. SCOPE FOR THE FUTURE WORK:

Research in science and its technological development are important for the future growth, for the benefit of the mankind. As there is always a scope for extension of research, useful work can be done on these materials, for their use in wider areas,

Further study can be undertaken in following areas;

1. The structural properties of the sintered samples for deeper understanding of the same.

2. The surface area measurement and adsorption studies of this ferrite materials for the catalytic applications and also as sensors,

3. Study of magnetite properties at low temperatures and low field by employing VSM and SQUID,
4. The investigation of superparamagnetism in unsintered nano size samples,

5. Study of permeability of the sintered samples prepared by the wet chemical method, and,

6. Any other relevant investigations on neutron diffraction, microwave absorption studies, etc can be carried out for a variety of applications of these materials.

As these materials find use in high technological areas, one can always take up future studies on different properties of these materials.