## **9.1 Conclusions**

In summary, detailed analysis of structural and electro-magnetic properties of  $SrAl_4Fe_8O_{19}$  hexaferrites had been investigated. Firstly, synthesis process of pure  $SrAl_4Fe_8O_{19}$  hexaferrite was being optimized by the sol-gel auto combustion process. Further, cationic substitutions were being explored to tailor the electro-magnetic properties of  $SrAl_4Fe_8O_{19}$  hexaferrite. Substitution of  $Co^{+2}$ ,  $Cr^{+3}$  &  $Sn^{+4}$ ,  $Y^{+3}$ , and  $La^{+3}$  &  $Sm^{+3}$  at  $Fe^{+3}$  site had been investigated in  $SrAl_4Fe_8O_{19}$  hexaferrite. The significant conclusions of this work are as follows:

- SrAl<sub>4</sub>Fe<sub>8</sub>O<sub>19</sub> hexaferrite had been synthesized by three synthesis processes: sol-gel auto combustion, co-precipitation and solid state method. This work demonstrated the effect of synthesis process on the microstructure as well as the electro-magnetic properties of prepared SrAl<sub>4</sub>Fe<sub>8</sub>O<sub>19</sub> hexaferrite. Sol-gel auto combustion process formed single phase strontium hexaferrite with high crystallinity. Rietveld refinement exposed hexagonal crystal regularity with *P63/mmc* space group. SEM micrograph demostrated that the grains were in well-defined hexagonal platelet shape with clear boundaries. Maximum intrinsic coercivity (*iH<sub>c</sub>*) along with (*BH*)<sub>max</sub> up to 18.15 kOe and 6.03 MGOe had been achieved, respectively by the sol-gel auto combustion method. Intrinsic coercivity (*iH<sub>c</sub>*) and small grain size, obtained by the sol-gel auto combustion method.
- Thus, the nominal sol-gel auto combustion method was being optimized to synthesis the substituted SrAl<sub>4</sub>Fe<sub>8</sub>O<sub>19</sub> ferrites in the following sections.
- Cobalt substitution found to increase the density of the prepared  $SrAl_4Fe_{8-x}Co_xO_{19}$  (0.0  $x \leq 1.0$ ) hexaferrite. The grain size of the matrix increased with the increase in

concentration of  $\text{Co}^{+2}$  ions. The magnetic properties of the prepared samples were enhanced by the partial Co inclusions. This enhancement was due to an increase in bulk density. Density raised up the spin rotational involvement, which in turn increased the magnetization. Consequently, maximum *Br* was achieved about 5.01 kG for SrAl<sub>4</sub>Fe<sub>7.2</sub>Co<sub>0.8</sub>O<sub>19</sub>. Whereas, *iH<sub>c</sub>* and *(BH)<sub>max</sub>* were 0.86 kOe and 3.68 MGOe, respectively. Dielectric spectra showed decreasing trend of  $\varepsilon$  with respect to frequency in the studied frequency range. For SrAl<sub>4</sub>Fe<sub>7.2</sub>Co<sub>0.8</sub>O<sub>19</sub>,  $\varepsilon$  and  $\rho$  were observed about 33.76 and 3.06 x10<sup>4</sup> at 1 MHz, respectively.

- Effect of  $Cr^{+3}$  and  $Sn^{+4}$  had been investigated in substituted  $SrAl_4(Cr_{0.5}Sn_{0.5})_xFe_{8-x}O_{19}$ ( $0.0 \le x \le 0.8$ ) ferrites. The Rietveld refinement revealed the formation of a hexagonal structure with *P63/mmc* space group with no secondary phase formation. The unit cell parameters, atomic positions, bond lengths along with bond angles had also been explored by the X-ray diffraction studies. Characteristic bands of  $SrFe_{12}O_{19}$  showed in all the samples. The morphological studies revealed that the average size of grains was decreased with the dopant concentrations and dopants were homogeneously distributed. Maximum remnant magnetization Br,  $iH_c$  and  $(BH)_{max}$  was found to about 6.22 kOe, 13.84 kOe and 7.26 MGOe for  $SrAl_4(Cr_{0.5}Sn_{0.5})_{0.6}Fe_{7.4}O_{19}$ , respectively. Remnant magnetization Br was increased due to the enhancement in super-exchange interaction with magnetic moment of the system with substitution of  $Cr^{+3}$  and  $Sn^{+4}$ . The increased values of magnetic properties stated that it could be favorable candidates for permanent magnets applications.
- $Y^{+3}$  ion substituted  $SrAl_4Y_xFe_{8-x}O_{19}$  ferrite had three active modes 443, 574 and 621 cm<sup>-1</sup>. This indicated the presence of characteristic bands of  $SrFe_{12}O_{19}$ . The XRD study

revealed the phase formation of M-type hexaferrite without any secondary phase formation. Rietveld refinement confirmed that all peaks were associated to the M-type hexagonal structure of *P63/mmc* space group. The fitted patterns were in good matching along the experimental data indicating the better quality of the results. The grain size of the matrix decreased with increase in  $Y^{+3}$  ions. In magnetic properties, it was found that the *iH<sub>c</sub>* was enhanced from 18.15 kOe to 36 kOe by  $Y^{+3}$  inclusions. It was happened due to a reduction in grain size and enhancement in k<sub>eff</sub>. k<sub>eff</sub> was explored from the law of approach method. Maximum Tc was observed for x = 0.2 was 614 °C, which was higher than the pure strontium hexaferrite. This increase in curie temperature might be owing to the increase in k<sub>eff</sub> with  $Y^{+3}$  content in the system. The resistivity increased from 5.43x10<sup>4</sup> to 13.80x10<sup>4</sup>  $\Omega$ -cm with  $Y^{+3}$  substitution.

• The La and Sm substitution formed the secondary phase of SmFeO<sub>3</sub> in SrAl<sub>4</sub>(La<sub>0.5</sub>Sm<sub>0.5</sub>)<sub>1.5</sub>Fe<sub>6.5</sub>O<sub>19</sub> hexaferrite. Because of the larger ionic radius of RE ions, it induced the lattice strain in the structure. Rietveld refinement justified the M-type hexagonal structure with *P63/mmc* space group for SrAl<sub>4</sub>Fe<sub>8</sub>O<sub>19</sub>. Grains were homogeneously distributed and closely packed throughout the matrix in the sintered ferrites. It was desirable for the magnetic properties in Sr ferrite. This substitution of RE found to increase the *Br* and (*BH*)<sub>max</sub> up to 5.18 kG and 6.69 MGOe, respectively for SrAl<sub>4</sub>(La<sub>0.5</sub>Sm<sub>0.5</sub>)<sub>1</sub>Fe<sub>7</sub>O<sub>19</sub>. This was due to the enhancement in grain size and magnetic moment of the system. With substitution, grains were observed to increase up to x = 1.0 , i.e., SrAl<sub>4</sub>(La<sub>0.5</sub>Sm<sub>0.5</sub>)<sub>1</sub>Fe<sub>7</sub>O<sub>19</sub> composition. The  $\varepsilon$  was observed to increase whereas,  $\rho$  was decreased about 25.95 and 2.11x10<sup>4</sup> Ohm-cm, respectively (at 1 MHz) with substitution.

• Further, table 9.1 shows the comparison of the best proposed substituted  $SrAl_4Fe_8O_{19}$  hexaferrites with the commercial strontium hexaferrite.

Table 9.1 Comparison of various parameters of prepared SrAl<sub>4</sub>Fe<sub>8</sub>O<sub>19</sub> hexaferrite [Cochardt

Properties	Commercial	SrAl <sub>4</sub> Co <sub>0.8</sub> F	$SrAl_4(Cr_{0.5}Sn_{0.5})_{0.6}$	SrAl <sub>4</sub> Fe <sub>7.8</sub> Y <sub>0.2</sub>	SrAl <sub>4</sub> Fe <sub>7</sub> (La <sub>0.5</sub> Sm
	SrM	e <sub>7.2</sub> O <sub>19</sub>	Fe <sub>7.4</sub> O <sub>19</sub>	O <sub>19</sub>	0.5)1O19
Br (kG)	2.65	5.01	6.22	2.11	5.18
$iH_c$ (kOe)	3.5	0.86	13.25	36	6.17
(BH) <sub>max</sub>	4	3.68	7.26	3.63	6.69
(MGOe)					
Curie temp	456	203	357	614	219
( C)					

(1963), Kostishyn *et al.* (2016)]

To the best of our knowledge, the best magnetic property was achieved in  $SrAl_4(Cr_{0.5}Sn_{0.5})_{0.6}Fe_{7.4}O_{19}$  hexaferrite.

## 9.2 Future scope

With the deficiency of the availability of rare earth elements, the opportunity opens up to develop the less or rare earth free permanent magnets. Permanent magnetic materials are extensively used in magnetic resonance imaging, information recording, magnetic visualization, rotating machine, transducer and optical filter etc. SrAl<sub>4</sub>Fe<sub>8</sub>O<sub>19</sub> hexaferrites shows the suitability of these demands and deliberates to the new advancement in permanent magnetic materials. As far as material application is concerned, following work objective can be taken for further advancements and growth of the permanent magnets:

- Understanding the physics behind the improvement in remanence magnetization (*Br*), which is tricky compared to improve in coercivity.
- To explore the mechanism of sintering for different sintering aids
- Sintering kinetics can be studied to understand the effect of microstructure. It has been observed that the microstructure plays a key role to effect the electro-magnetic properties of the materials.
- Fabrication and characterization of thin film using modified Sr ferrites for antenna applications.
- To investigate the properties at high frequency (at GHz) range.