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# Effect of γ- irradiation on Dielectric, AC conductivity and IR absorption in the $Sr_{1-x}La_xFe_{12}O_{19}$ ferrites

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**Abstract :** A series of samples of $Sr_{1-x}La_xFe_{12}O_{19}$ ( x = 0.0, 0.10, 0.20 and 0.30 ) were prepared by employing the ceramic technique. The samples so prepared were irradiated by $Co^{60}$ gamma source for 30 hours. The infrared absorption spectrum of the samples were analysed. The gamma radiation induced modifications on electrical behavior and Infrared absorption spectrum of $Sr_{1-x}La_xFe_{12}O_{19}$ ferrites have been studied. The dielectric constant ($\varepsilon'$), Dielectric loss tangent ( tanδ ) and AC conductivity ($\sigma_{AC}$) as a function of frequency from 20 Hz to 1 MHz were measured at room temperature for un–irradiated and irradiated samples. The observations reveal that the conductivity of the irradiated samples increases with increase of frequency whereas the dielectric constant shows a decrease with increase of frequency .

**Keywords:** Hexagonal ferrites, Gamma radiation, AC conductivity, Dielectric Constant ($\varepsilon'$), and Dielectric loss ( tanδ ).

## Introduction

The hexagonal ferrites have been extensively studied during the past years due to their technological interest as traditional permanent magnets, microwave device materials and magnetic recording media [1,2 ]. The ferrites have an interesting and complex-magnetoplumbite type structure in which the iron ions are coordinated tetrahedrally ($FeO_4$), trigonal bipyramidally ($Fe_2O_5$), and octahedrally ($FeO_6$) by oxygen ions. Many researcher have studied the magnetic properties of each Ba-ferrite and Sr-ferrite prepared by various techniques such as chemical coprecipitation method [3], the glass crystallization [4], the salt melt method [5], the sol-gel method [6] and ceramic process [7]. Most of the research has emphasized the modification of magnetic properties by the substitution of $Fe^{3+}$ with 3d ions such as $Co^{2+}Ti^{4+}$, $Co^{2+}Sn^{4+}$, $Cr^{3+}$ and $Al^{3+}$ [8,9]. Magnetic properties of La substituted in the system $Sr_{1-x}La_xFe_{12}O_{19}$ has also been reported [10,11]. However a very little information is available on effect of gamma irradiation on AC conductivity and dielectric properties of Sr-La ferrites in the literature.

Interaction of radiant energy with matter, especially γ-radiation, is an extremely important problem from the viewpoint on the materials. The electrical properties of the materials may change and new electrical phenomenon may be developed. Also, profound changes take place in structure of the materials with result of changing mechanical strength, optical properties, etc. A broad development of nuclear engineering and use of radioactive isotopes in the past years have put forward more often problem of the action of various kinds of ionizing radiation of materials [12]. Elementary charged particles and electromagnetic radiation with high energy interact with an electronic shell or the atomic nuclei of the substance passing through them . These interaction results in elastic and inelastic scattering of atoms as well as the initiation of nuclear reaction and also disturbance in the structure of matter, the so called radiation damage. For this reason, much attention has been given to study the effect of γ-irradiation on electrical properties of the samples under investigation in the present work.

## Experimental Details

A series of $Sr_{1-x}La_xFe_{12}O_{19}$ samples with different substitution ratios were prepared by a standard ceramic processing technique [13-16]. High purity materials $SrCO_3$, $La_2O_3$ and $Fe_2O_3$ procured from LOBA CHEMIE PVT. LTD. Mumbai (India) were used as starting raw materials and were mixed together in the appropriate composition, calculated from the following chemical reaction:

$$(1-x)\ SrCO3 + 6\ Fe_2O_3 + (x/2)\ La_2O_3 \rightarrow Sr_{1-x}La_xFe_{12}O_{19} + (1-x)CO_2 + (x/4)O_2 \quad (1)$$

Where, x varies from 0.0 to 0.3 with an increment of 0.1 . The powders so obtained were mixed in distilled water and milled for 6 hours in an agate motor and pestle fitted with an electric grinder rotating at a speed of 100 RPM. After grinding the powders were dried in hot air. The powders thus dried were calcined at $1200^0$C for six hours in a resistance furnace at a controlled heating and cooling rate ($5^0$C/min.). The calcined powders were further wet- milled in distilled water for 8 hours to obtain very fine and uniform size powders. After milling the powders were again dried and 5% polyvinyl alcohol was added to it, which acts as binder. These materials were then granulated through sieves of 60-80 mesh B.S.S (250-180 µm approximately). The granules were cold pressed in a die at a compressed pressure of 75 KN to produce pellets of 14.5 mm diameter and 3.0 mm to 4.0 mm thickness. These pellets were finally sintered at $1250^0$C for 2 hours. The samples were cooled in the furnace itself to room temperature. The phase analysis of these sintered pellets was carried out by X- Ray diffractometer (Rigaku DMAX IIIC). AC conductivity and dielectric measurements of all ferrites samples were measured (HP4284A) with standard two-probe technique [17]. The AC conductivity was calculated by using the formula :

$$\sigma_{AC} = \frac{Gt}{A} \qquad (2)$$

Where t and A are thickness and cross-sectional area of pellets and G is the conductance. The value of dielectric constant ($\varepsilon'$) of the ferrite samples can be calculated by using the following formula

$$\varepsilon' = \frac{C_p t}{\varepsilon_0 A} \qquad (3)$$

Where $C_p$ is the capacitance of samples in pf, t the thickness of the samples in cm, A the cross-sectional area of the samples in sq.cm and $\varepsilon_0$ the permittivity in free space having value $8.854 \times 10^{-2}$ pf/cm.

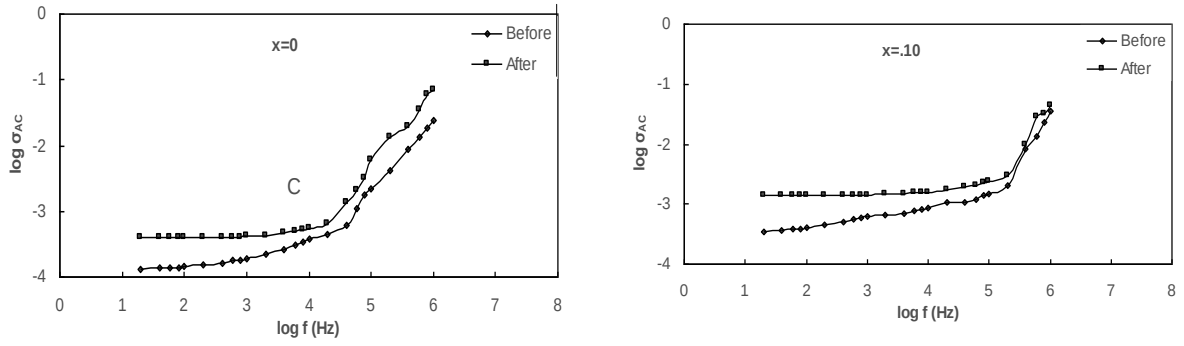
The Fourier transform-infrared (FT-IR) Spectra were recorded at room temperature for all the samples in the transmission mode using Fourier transform spectrometer in range 4000-400 $cm^{-1}$.

## Result and discussion

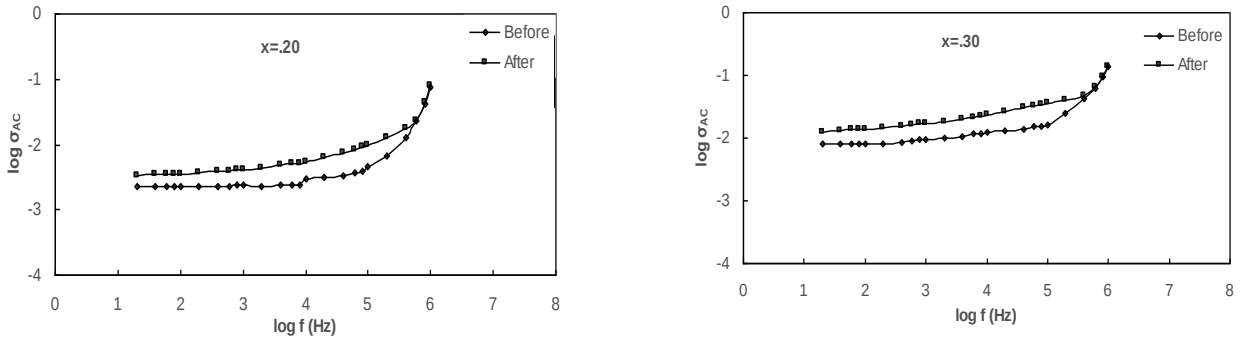
### Effect of γ- irradiation on AC conductivity

The variation of AC conductivity for un-irradiated and γ -ray irradiated Sr-La ferrites as a function of frequency from 20 Hz to 1MHz has been measured. Fig.1. shows the AC conductivity as a function of frequency un – irradiated and irradiated Sr1-xLaxFe12O19 (x = 0.0, 0.10, 0.20, 0.30) materials.

Observations reveal that for all the compositions the AC conductivity for un- irradiated and irradiated samples first increases gradually with the increase of frequency followed by a rapid increase.




(a) (b)




(c) (d)

**Fig. 1. (a-d) Variation in AC conductivity of $Sr_{1-x}La_xFe_{12}O_{19}$ with frequency before and after irradiation**.

The frequency dependence of $\sigma_{AC}$ has been explained with the help of Maxwell-Wagner two-layer model [18,19]. According to this model the hopping frequency of electron between $Fe^{2+}$ and $Fe^{3+}$ ions at the meta interface is less at lower frequencies. As the frequency of applied field is increased , the conduction mechanism becomes more active by promoting the hopping of charge carriers between $Fe^{2+}$ and $Fe^{3+}$ ions, thereby increasing the hopping frequency. They observed a gradual increase in conductivity with frequency but at higher frequencies, the frequency of hopping between the ions could not follow the applied field frequency but lagged behind it. It has been reported that frequency dependence of AC conductivity is large in ferromagnetics [20]. As the frequency increases, the mean displacement of the charge carriers is reduced. Similar results and reasons have been reported by B. Kaur et. al. [21] The real part of AC conductivity can be expressed generally by the following relation [12].

$$\sigma' = \sigma_{DC} + \sigma_{AC} \qquad (4)$$

Where $\sigma_{AC}$ is electric conductivity due to the free charges and $\sigma_{AC}$ is the electric conduction due to bound charges and can be expressed in terms of angular frequency $\omega$ by the relation

$$\sigma_{AC} = A\,\omega^{S} \qquad (5)$$

Where A is the parameter having units of conductivity and s is dimensionless parameter . The parameter s is temperature and compositional dependent and $\omega = 2\pi f$ is the angular frequency .

**Effect of γ- irradiation on the dielectric constant**

The dielectric properties of ferrites are dependent upon several factors namely chemical composition , method of preparation, grain size etc. The Study of dielectric constant is very useful in the study of phase transition in the material occurring.

The variation of dielectric constant ($\varepsilon'$) and dielectric loss tangent ($\tan\delta$) of irradiated and un-irradiated Strontium hexaferrites was carried out in the range $20 - 200^0$C under the application of applied a.c field in the frequency range of $10^3 - 10^7$Hz. The dielectric constant shows an increases at a particular frequency for both irradiated and un-irradiated crystals with the increase in temperature. For temperature $200^0$C , the crystal did not show any response at lower frequency of 60 KHz for un-irradiated and at a frequency of 70 KHz for irradiated crystals . The dielectric constant decreases as the frequency is increased for different temperatures and becomes saturated at higher frequencies above 10 KHz there ist for a small increase in dielectric constant at initial frequencies at a temperature of $150^0$C for un-irradiated and at $100^0$C of irradiated crystals . This behaviour of dielectric constant with frequency may be attributed due to the following reasons [22,23, 24] as has also been argued by Dogra et al. [22] for other ferrites :

**(a)** On irradiation, $Sr_{1-x}$ $La_xFe_{12}O_{19}$ crystals show decrease in the value of dielectric constant. It is well known that the maximum polarization occurs only if maximum numbers of ferrous ions are involved in the exchange phenomenon. The observed decrease in the value of dielectric constant in a composition where Fe content of ferrous site to a much lower value that may be available for polarization.

**(b)** On irradiation, the flow of space charge carriers is obstructed by the defect s generated by the irradiation. Thus the build up of space charge polarization is impeded. The possibilities may be related to space charge polarization at the metal interface as reported by Costantini et al.[25].

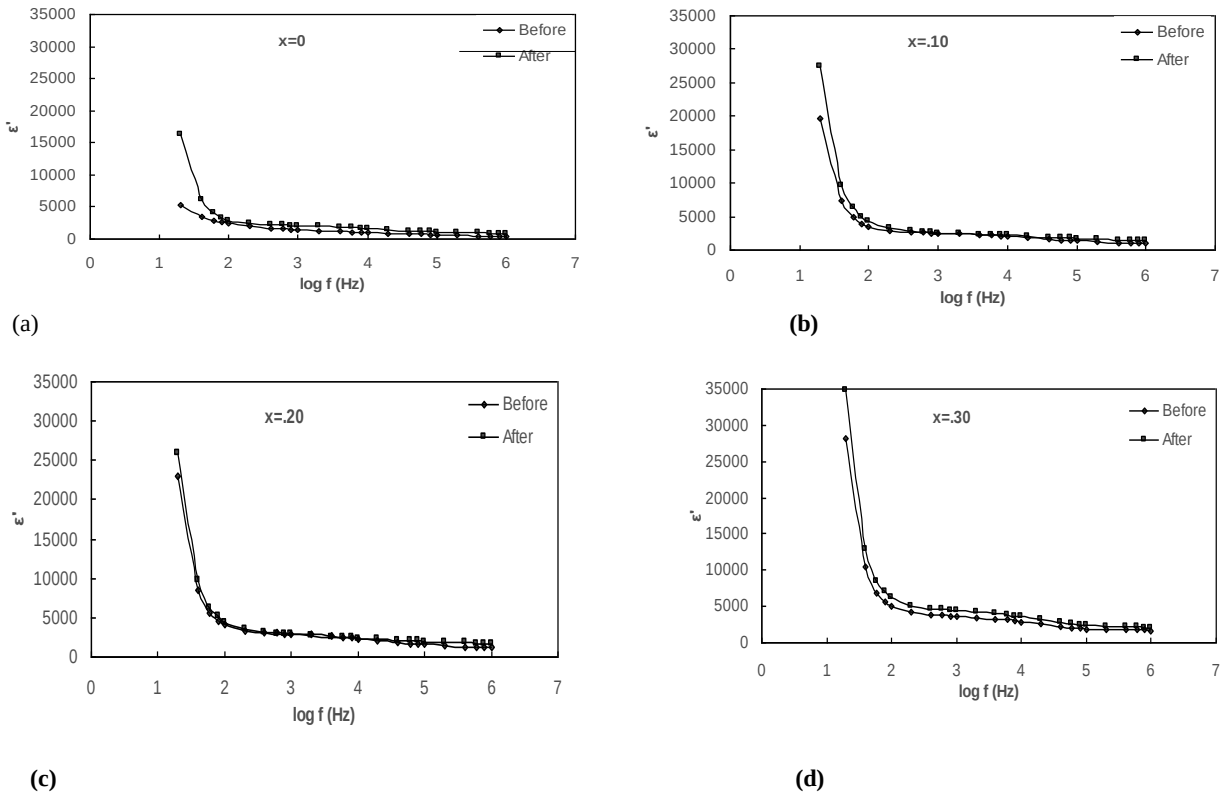



**Fig. 2.(a-d) Variation in dielectric constant ($\varepsilon'$) of $Sr_{1-x}La_xFe_{12}O_{19}$ with frequency before and after irradiation.**

**Effect of γ- irradiation on Dielectric Loss**

It is well known that dielectric of a material depends upon the polarization that arises from the induced electrical dipoles with the material . With the increase of temperature , the ionic distance increases , which in turn weakens the ionic and electronic contribution . The decrease in the value of dielectric constant and attaining saturation with increase in frequency is due to decrease in ionic polarization as beyond a certain frequency of the externally applied filed, the electronic exchange can not follow alternating field .

If the crystal is ferroelectric , the dipoles do not generally line up across the whole crystal but form domains . This minimizes the energy and represents a compromise between domain wall energy and external electrostatic energy. On the application of field, the polarization increases , not by an increase in the sizes of the individual dipoles but by the migration domains boundaries in such a way that favorably oriented domains grow and vice- versa . It has been observe that at lower frequency dielectric loss decreases with increase in frequency at almost all temperatures , at frequency above 100 KHz the dielectric loss increases and shows sharp resonance peaks at around of 2 MHz . The loss factor is the ratio of imaginary ($\varepsilon''$) and the real ($\varepsilon'$) parts of the dielectric constant . Therefore peaks in the loss factor can be attributed to the Maxwell- Wagner Interfacial polarization. When an external alternating electric field is applied with same frequency as that of the natural frequency of oscillating ion, maximum energy is transferred to the oscillating ions due to which the power losses shoots up,

this results in the resonance[26-27] . B. Kaur et al. [28] has also presents the similar results for irradiation effect on dielectric behaviour of M-Type Hexaferrites.

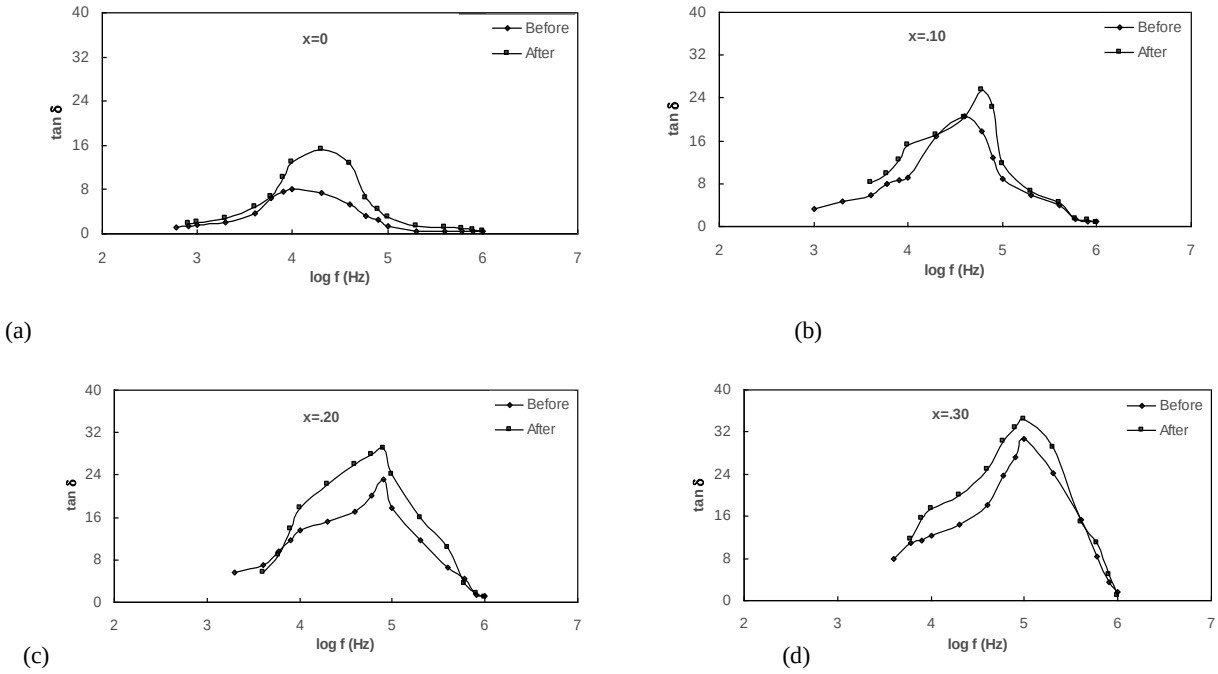



**Fig.3 (a-d) Variation of dielectric loss tangent (tanδ)of $Sr_{1-x}La_xFe_{12}O_{19}$ with frequency before and after irradiation**

**Effect of γ- irradiation on IR Spectra**

Infrared absorption frequency for Ferrites before and after γ- irradiation and the results are shown in Table -1. and fig.(4). It has been observed that some absorption bands disappear after irradiation . This may be due to shifting of some ions of small size to interstitial positions in the crystal lattice by γ- rays resulting in the disappearance of some absorbed bands after gamma irradiation.

Further, some ferric ions exposed to γ-irradiation changed to ferrous ions. These ferrous ions (r = 0.78 nm) have large size than $Fe^{3+}$ ions (r = 0.64 nm) leading to the shift of some of the absorbed bands. The IR absorption in $Co_{0.6}Zn_{0.4}Mn_xFe_{2-x}O_4$ system before and after γ-irradiation by Hamada [29] has shown the same results.

**Table : Frequency bands of different ratios.**

| Frequency bands ($cm^{-1}$) | Before | After |
|---|---|---|
| **(a) x=0** | | |
| $v_1$ | 470.60 | ------ |
| $v_2$ | 549.67 | 549.67 |
| $v_3$ | 595.90 | 601.75 |
| $v_4$ | 906.48 | ----- |
| $v_5$ | 1193.85 | 1139.85 |
| $v_6$ | 1217.00 | ---- |
| $v_7$ | 1265.22 | ---- |
| $v_8$ | 1373.22 | 1367 |
| $v_9$ | 1610.45 | 1610.45 |
| $v_{10}$ | 1990.40 | ---- |

| | | |
|---|---|---|
| **(b) x=0.10** | | |
| $v_1$ | 435.88 | ---- |
| $v_2$ | 549.67 | 549.67 |
| $v_3$ | 599.82 | 594.82 |
| $v_4$ | 1259.43 | ---- |
| $v_5$ | 1288.36 | ---- |
| $v_6$ | 1313.43 | 1319.22 |
| $v_7$ | 1614.31 | 1608.59 |
| $v_8$ | 1716.53 | ----- |
| $v_9$ | 1990.40 | ---- |
| **(c) x=0.20** | | |
| $v_1$ | 549.67 | 549.67 |
| $v_2$ | 599.82 | 599.82 |
| $v_3$ | ---- | 613.32 |
| $v_4$ | 1218.93 | ---- |
| $v_5$ | 1251.72 | 1236.43 |
| $v_6$ | 1389.37 | 1367.44 |
| $v_7$ | 1616.24 | 608.52 |
| $v_8$ | 1749.32 | ---- |
| $v_9$ | 1992.33 | ---- |
| **(d) x=0.30** | | |
| $v_1$ | 547.75 | 547.75 |
| $v_2$ | 594.03 | 592.11 |
| $v_3$ | 1255.57 | ---- |
| $v_4$ | 1288.3 | ---- |
| $v_5$ | 1313.43 | 1317.29 |
| $v_6$ | 1367.44 | 1367.44 |
| $v_7$ | 1614.31 | 1610.45 |
| $v_8$ | 1714.60 | 1716.53 |

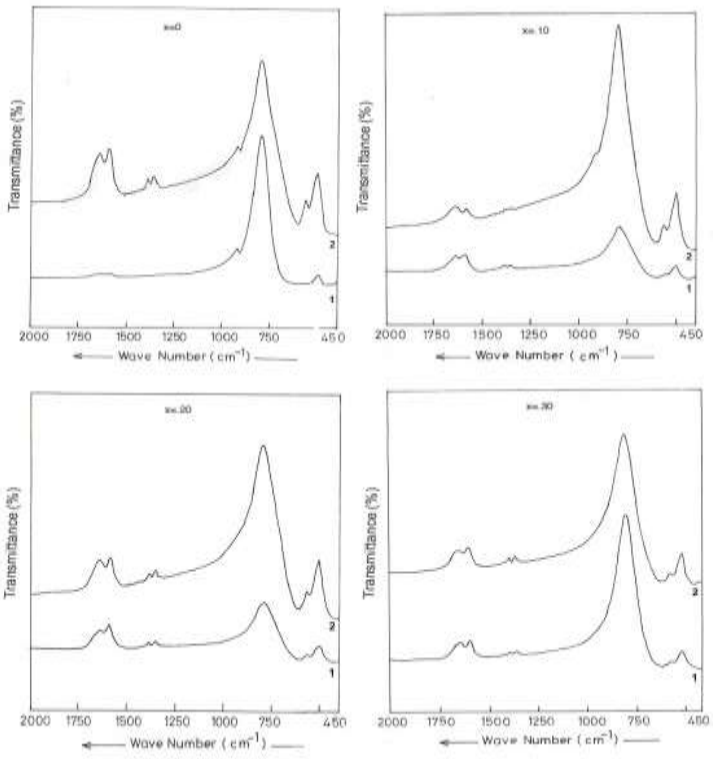



**Fig.4. Variation of IR Spectra of $Sr_{1-x}La_xFe_{12}O_{19}$ with frequency before and after irradiation.**

## Conclusion

The value of dielectric constant ($\varepsilon'$), dielectric loss tangent (tanδ) and AC conductivity of $Sr_{1-x}La_xFe_{12}O_{19}$ are strongly dependent on the frequency, applied voltage and dose rate, which further increases after irradiation. Also, value of $\varepsilon'$ and $\varepsilon''$ increases with increasing dose rate, but decrease with increasing frequencies. The AC conductivity $\sigma_{AC}$ increases with increasing dose rate and frequency . These changes of $\varepsilon'$ , $\varepsilon''$ , tanδ and $\sigma_{AC}$ values are different at different frequencies and radiation (γ-ray). It is also considered that the interface state density increase with increasing amount of irradiation on dose.

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