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Effect of Fe_2O_3 on impedance properties of CuO in $_{1-x}(CuO)$- $_{x}(Fe_2O_3)$ Ceramic Composites

Rajnish[1], Sunita Dahiya[2], Sunil Kumar[3*], Naveen Kumar[4]

[1,2]Department of Physics, Baba Mastnath University, Rohtak, Haryana, India
[3]Department of Physics, RR BAWA DEV College, Batala (143505), Punjab, India
[4]Department of Physics, Panjab University Chandigarh, Chandigarh, India
*Corresponding Author E-mail: sunil_nano@yahoo.com

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ABSTRACT

$_{1-x}(CuO)$-$_{x}(Fe_2O_3)$ ceramic composites (x = 0.05, 0.10, 0.15 & 0.20) have been successfully prepared using ball mixing approach and reported for temperature dependent impedance characteristics (Resistive Response), dielectric relaxation as well asfor contribution of grains and grain boundaries in electric properties. Z' vs. Frequency at different temperature shows effect of increase of temperature on resistive behavior of prepared ceramic composites. The shift in maxima as temperature increases in Z" vs. Frequency confirms presence of temperature dependent dielectric relaxation. The semi circles in Z' vs. Z" show contribution of grains and grain boundaries in electric properties.

Keywords: **Composites material; Dielectric Relaxation, Nyquist Plots, Barrier Properties.**

INTRODUCTION

The importance of a material in electronic industries as key components has been described in terms of value of dielectric constant. The dielectric constant (k) which demonstrates ability of a material to store charge while examining its dielectric characteristics. Low-k valued materials have been used in electrical insulation and high-speed integrated circuits whereas high-k materials employed as gate dielectrics in MOS transistors, memory cells, capacitors, super capacitors, etc. Dielectric materials also be important candidates in semiconducting industry used to develop cutting-edge microelectronics stream. Due to their exceptional mechanical and dielectric qualities, metal oxide dielectric materials are regarded as essential components for a variety of thin-film electronic applications.

Due to the scientific and technological importance, metal oxides holds great position in semiconducting and microelectronic industry due totheir tunable band gap. Among such type of metal oxides semiconductors, transition metals oxides are prime candidates in field of semiconductors as well as microelectronic applications due to their unique, diverse, and rich physical as well as chemical properties. Transition metal oxides have been used electrode materials in various components of electronic industries in past few years. Semiconductor nanocrystal materials with narrow band gap attracts researcher from various sectors of industry including semiconducting applications, Dye Solar Cell, Catalysts etc.

Copper oxide (CuO) is a basic, most stable transition metal oxide based magnetic semiconductor which most commonly used in electronic industry because of its quantum confinement effect, tunable band gap ~1.2 to ~2.1 eV. CuO is a p-type semiconductor can also be used optoelectronic devices, sensors, catalysis, lithium-ion batteries, high critical temperature superconductors, super-capacitors, magnetic storage media, solar energy conversion, smart windows, optical limiters, thin film transistors, field emission emitters, and biomedicine. CuO has a space group of C2/c and crystallizes in a monoclinic crystal structure [1-13].

In this paper, effect of Fe_2O_3 on impedance properties and contribution of grain and grain boundaries from nyquist plots $_{1-x}(CuO)$-$_x(Fe_2O_3)$ ceramic composites (x = 0.05, 0.10, 0.15 & 0.20) have been reported.

Experimental Methodology & Characterization

High energy milling technique has been used to synthesize $_{1-x}CuO$-$_xFe_2O_3$ (x = 0.05, 0.10, 0.15, & 0.20) ceramic composites. High Energy ball milling machine used for this synthesis process. In this process, CuO and Fe_2O_3 of high pure quality have been taken from Sigma Aldrich weighed and mixed in stoichiometric ratio mentioned in introduction section as well as in starting of this section and ball milled using milling machine and round shaped zirconia ball in a bottle.

The milling process carried out in presence of water taken as non-reacting solvent. After completion of milling process, powder taken out from bottles, let for dry. The dried powder mixed with polymer binder (Most commonly used Polyvinyl Alcohol) and pressed in disc shaped tablets. These disc shaped tablets sintered at 900 °C for densification. The impedance characteristics of sintered pallets have been studied. For these measurement, electrode (Circular Shaped circle) of conducting material (Either Silver or Gold conducting paint) has been used.

Characterization Methodology Used: Complex Impedance Spectroscopy:

Impedance spectroscopy (Z' vs. Frequency, Z" vs. Frequency & Z" vs.Z') at different temperature has been used to study effect of temperature on resistive behavior, dielectric relaxation as well as contribution of grain and grain boundaries on electrical properties of prepared ceramic samples. For this, Z' & Z" has been calculated using relation given below from experimentally collected data in form of Z & θ vs. Frequency at selected temperature. For this, LCR meter commonly known as impedance analyzer Keysight Technologies (E4990A) interfaced with computer and furnace has been used.

$$Complex\ Impedance, Z^* = Z' - jZ''$$

$$Z' = |Z| \cos\theta$$

$$Z'' = |Z| \sin\theta$$

Nyquist Plots: Z" vs. Z'

RESULTS & DISCUSSION

Impedance Spectroscopy:

Figures 1: shows variation of Z' vs. Frequency (Hz) in given temperature range varies from 323K-548K of ceramic composites of $_{1-x}CuO$-$_xFe_2O_3$ (x = 0.05, 0.10, 0.15, & 0.20) in entire frequency range from 100 Hz-10^6Hz. It has been clearly visualized from graphs of Z' vs. Frequency (Hz) that value of Z' decreases with increasing both temperature as well as frequency evident for decrease in resistive properties of prepared ceramic composites. The decrease in resistive response may also resulted due to formation of oxygen vacancies or defects as temperature increases during measurement. The hopping of Fe ion into multiple valence states (Fe^{2+}& Fe^{3+}) with increasing may be another aspects of decrease in resistive response because this hopping results due to oxygen vacancies.

The resistive response also decreases with increasing Fe_2O_3 oxide in CuO-Fe_2O_3 because of conductive behavior of FeO. This decrease of resistive behavior also reveals that prepared ceramic composites exhibit negative temperature coefficient of resistance (NTCR) [13]. This decrease in value of Z' with increasing temperature directly proclaim the reduction in resistive properties followed by increasing conductive behavior due to increasing concentration of ferrite (Fe_2O_3) in prepared ceramic composites. The temperature dependent conductivity (σ_{ac}) behavior directly support decreased in resistive behavior or barrier as temperature increases for CuO-Fe_2O_3[14-15].

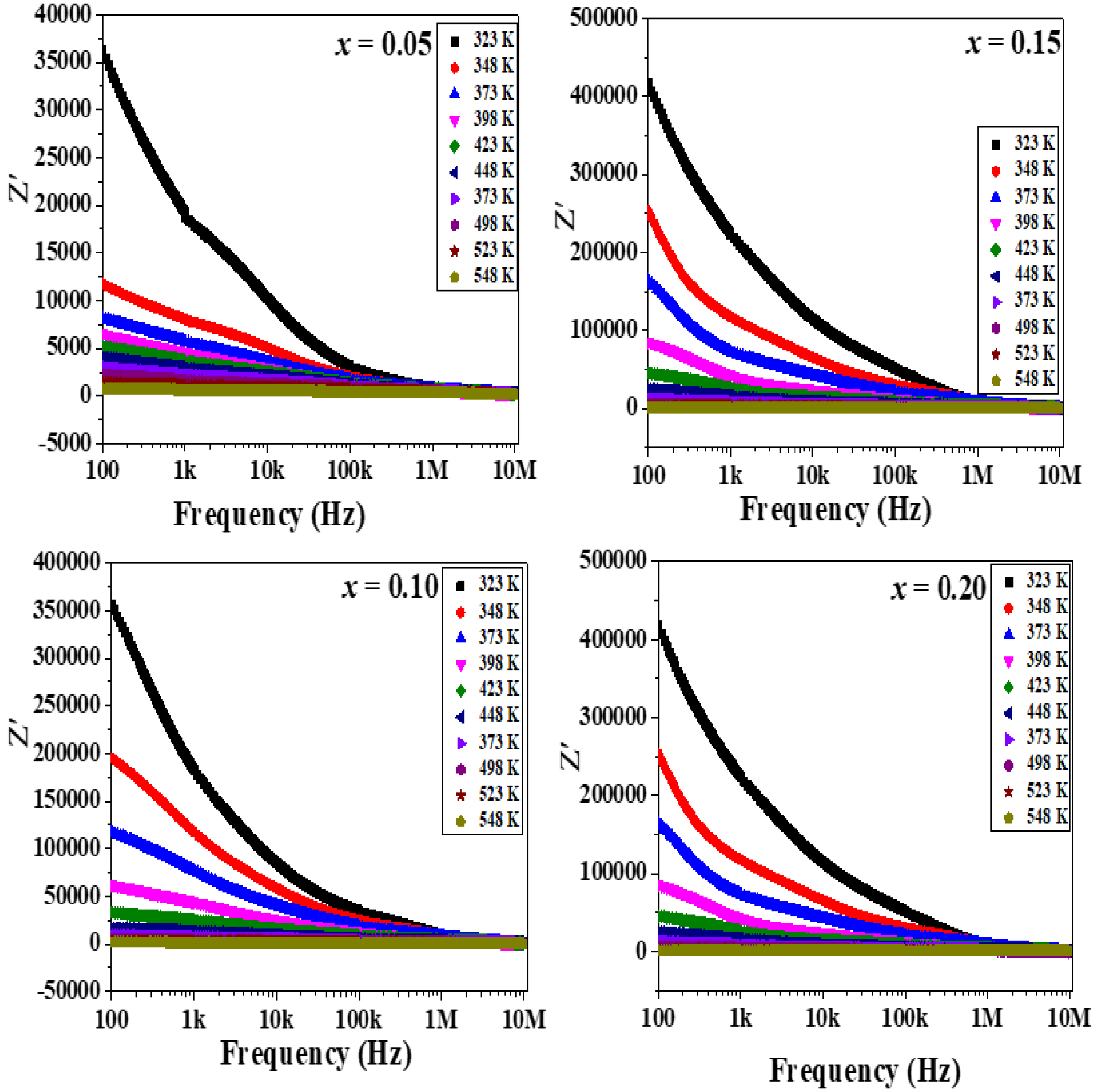

Figure 1: $Z^{\prime}$ vs. frequency temperature range 323K-548K of ceramic composites of $_{1-x}$CuO-$_x$Fe$_2$O$_3$ (x = 0.05, 0.10, 0.15, & 0.20)

Figures 2: shows variation of $Z^{\prime\prime}$ vs. Frequency (Hz) in given temperature range varies from 323K-548K of ceramic composites of $_{1-x}$CuO-$_x$Fe$_2$O$_3$ (x = 0.05, 0.10, 0.15, & 0.20) in entire frequency range from 100 Hz-10^6Hz. The analysis of $Z^{\prime\prime}$vs. frequency as well as temperature is a conclusive component in impedance analysis of any ceramic oxides used to study resistive response of prepared samples. It has been clearly delineated from graphs that $Z^{\prime\prime}$first increases as frequency increases reaches upto certain maximum value and then starts decreases with further increase in frequency. The maximum value of $Z^{\prime\prime}$termed as maxima and this maxima corresponds to dielectric relaxation. It has also be depicted from graphs that as temperature increases, peaks of $Z^{\prime\prime}$vs. frequency gets broadening and after particular temperature, peaks merged in higher frequency region. This broadening and merging of peaks with increasing temperature may results due to vacancies or defects that have been generated at higher temperature, elimination of space charges as well as immobile charges at low temperature [13-18].

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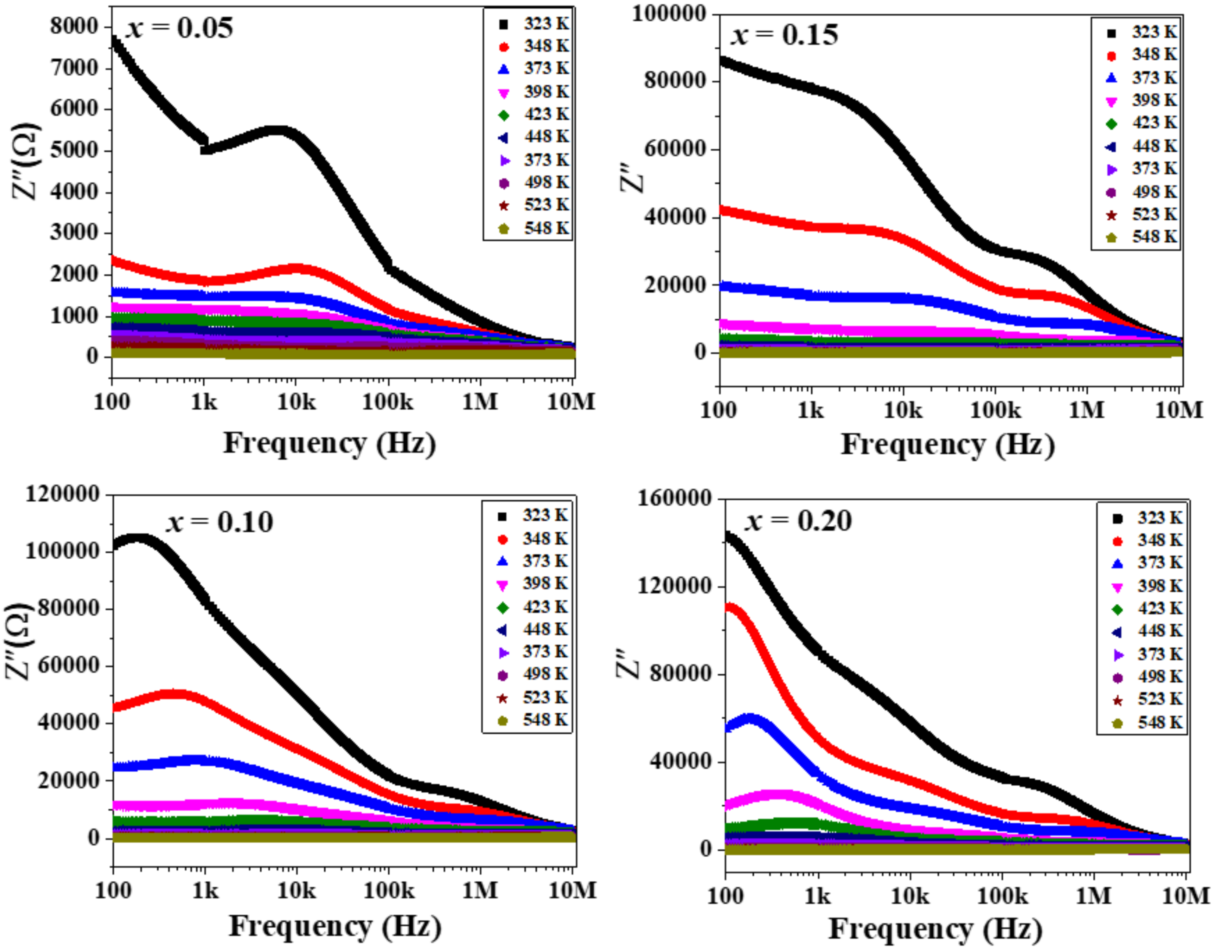

Figure 2: Z'' vs. frequency temperature range 323K-548K of ceramic composites of $_{1-x}$CuO-$_x$$Fe_2O_3$ (x = 0.05, 0.10, 0.15, & 0.20)

Figures 3 shows variation of Z'' vs. Z' in given temperature range varies from 323K-548K of ceramic composites of $_{1-x}$CuO-$_x$$Fe_2O_3$ (x = 0.05, 0.10, 0.15, & 0.20) in entire frequency range from 100 Hz-10^6Hz. The graph consists of semicircles express their importance in analyzing role of grain and grain boundaries in electrical properties of prepared ceramic composites. Since grain boundaries have been considered as region having low permittivity in comparison of with grains. Therefore grains and grain boundaries have important role in electrical properties of any prepared ceramic samples. The semi-circle variation corresponds to grain and grain boundary resistance (R_g&R_{gb}) as well as grain and grain boundary capacitance (C_g&C_{gb}). The cole-cole plots (Also termed as Nyquist plots) have been used to study variation of grain and grain boundary resistance (R_g&R_{gb}) as well as grain and grain boundary capacitance (C_g&C_{gb}) with both increasing temperature as well as composition of dopant. For this, cole-cole plots (Also termed as Nyquist plots) have been fitted using software known as Zview with suitable circuit of Resistance (R) – Capacitance (C) to calculate resistance and capacitance of grain and grain boundariesdepends upon no. of semicircles appears in experimental data [19]. The most common circuits that have been used to analyze either parallel combination of parallel combination of R_g and CPE_gcorresponds to grain circuit whereas R_{gb} and CPE_{gb}represents grain boundary circuitin parallel combination. The deviation of from ideal Debye behavior has been studied from phase element can be represented as CPE determines. The impedance of CPE is given by $Z_{CPE} = 1/(j\omega)^{\beta}\, CPE$, where $\beta \leq 1$. The equation for the equivalent circuit can be represented by $Z^*(\omega) = Z' + jZ''$:

$$Z' = \frac{R_g}{1+(\omega_g R_g C_g)^2} + \frac{R_{gb}}{1+(\omega_{gb} R_{gb} C_{gb})^2} \quad (3)$$

$$Z'' = \frac{\omega_g R_g^2 C_g}{1+(\omega_g R_g C_g)^2} + \frac{\omega_{gb} R_{gb}^2 C_{gb}}{1+(\omega_{gb} R_{gb} C_{gb})^2} \quad (4)$$

where (R_g, C_g, ω_g) & (R_{gb}, C_{gb}, ω_{gb}) corresponds to their usual meaning such as resistance, capacitance and frequency at the peaks of the semicircles for grain and grain boundaries respectively. It has been clearly visualized from graphs that as temperature increases, semicircles gets merged whereas as concentration of Fe_2O_3 increases, single semicircle get converted into double semicircles. It has also depicted from graphs that as frequency increases, circle starts merging and disappear after certain frequency stamped for trifling contribution of grains in dielectric properties of prepared ceramic composites as well as curtailment in resistive properties. This curtailment in resistive properties starts as radius of semicircle gets smaller with increasing temperature. The shrinking of resistivity (Resistive/Barrier Properties) of grain boundary results for presence of transportation of conduction due to temperature. The centers of all circles (semicircles) in graphs below Z' axis is a direct evidence for presence of statistically distributed relaxation phenomena that stamped for non-Debye relaxations. This non-Debye behavior may be due to oxygen vacancies, defects, orientation of grains and grain boundaries etc.

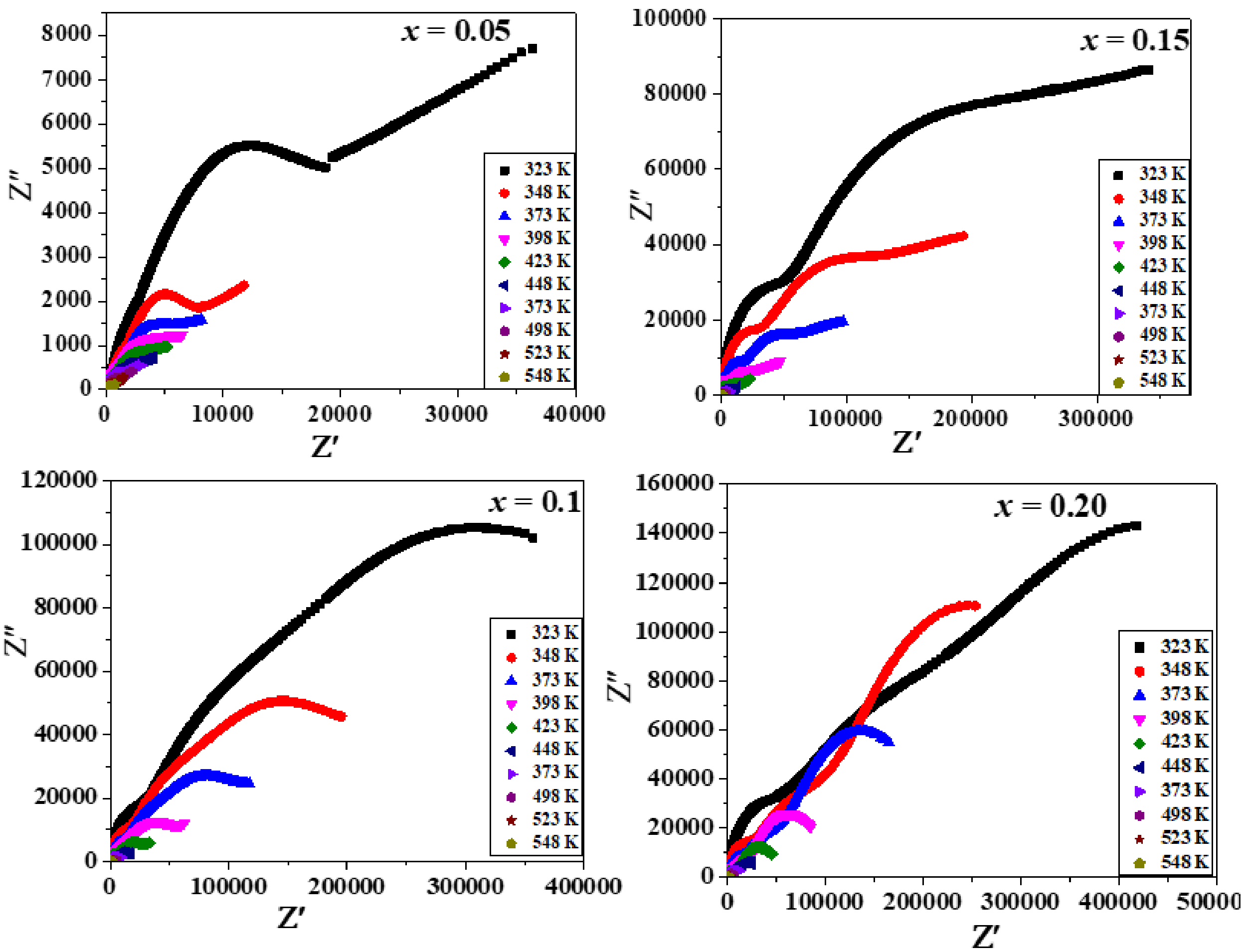

Figure 3: $Z^{\prime}$ vs. frequency temperature range 323K-548K of ceramic composites of $_{1-x}$CuO-$_x$$Fe_2O_3$ (x = 0.05, 0.10, 0.15, & 0.20)

CONCLUSION

Ceramic composites of $_{1-x}$(CuO)-$_x$(Fe_2O_3) have been successfully synthesized using high energy milling process and explored for temperature dependent impedance properties as well as contribution of grain and grain boundaries in electric properties. The decrease in value of Z' as temperature increases stamped for decrease in resistive behavior with simultaneous increase in conductivity. The shift in maxima as temperature increases in both Z" vs. frequency reveals presence of temperature dependent dielectric relaxation. The graphs shows that as temperature increases, peaks of $Z^{\prime\prime}$ vs. frequency gets broadening and after particular temperature, peaks merged in higher frequency region. This broadening and merging of peaks with increasing temperature may results due to vacancies or defects that have been generated at higher temperature, elimination of space charges as well as immobile charges at low temperature The semi circles in Z' vs. Z" show contribution of grains and grain boundaries in electric properties. The graphs that as frequency increases, circle starts merging and disappear after certain frequency stamped for trifling contribution of grains in dielectric properties of prepared ceramic composites as well as curtailment in resistive properties.

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