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SYNTHESIS AND CHARACTERIZATION OF NANOCRYSTALLINE Cu-Zn-Ce MIXED OXIDE BY COMBUSTION ROUTE

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ABSTRACT

The $Cu_xZn_{(1-x)}Ce_3O_5$ nanocrystalline mixed oxide was synthesized by combustion method . In Pyrex glass dish requisite quantities of copper(II) nitrate , zinc nitrate and cerric ammonium nitrate along with urea (as a fuel) were mixed thoroughly. The mixture was heated at temperature 373 K using hot plate. The mixed oxides were obtained in the form of dry powders. This powder was sintered at various temperatures to get the required phase. The phase formation of all samples of the series was conformed by x-ray diffraction data. Further these powders were characterized by SEM technique.

Nanocrystalline size of $Cu_xZn_{(1-x)}Ce_3O_5$ oxide has been observed in SEM analysis. The pellets were formed from these powders. The pellet was then used to measure the resistivity of nanoparticles in the temperature range 27°C–510° C and also dielectric constant at room temperature.

Ketwords: Nanoparticles, XRD, SEM, resistivity, dielectric constant, $Cu_xZn_{(1-x)}Ce_3O_5$ where (x= 0.25,0.5,0.75)

INTRODUCTION

Nanocrystalline materials are expected to be one of the most important materials during the present century. Nanocrystalline oxides are widely used in the development of heterogeneous catalyst, semiconducting materials, magnetic materials, automobile ceramics, gas sensors, microwave devices[1,2] . In present work we have synthesized mixed oxide $Cu_xZn_{(1-x)}Ce_3O_5$ containing rare earth and transitions metals. The above properties of oxides mainly depend on the preparative methods.[4-9] The various conventional methods such as co-precipitation, sol-gel, citrate gel, amorphous complementary have been employed to synthesis mixed oxides. Here we report that preparation of mixed oxide by solution combustion method. Cerric ammonium nitrate, copper nitrate, zinc nitrate used for synthesis of oxide. A.R. grade (Merck) reagents are used. The advantage of this method is that no use of water phase or any other solvent hence pure and dry powder can be obtained.

EXPERIMENTAL

Mixed oxide samples of composition $Cu_xZn_{(1-x)}Ce_3O_5$ were prepared by solution combustion method. All the reagents used in this preparation were of A.R.grade. Cerric ammonium nitrate (Merck 99 %) was used as the main source and copper (II) nitrate trihydrate, zinc nitrate hexahydrate (Merck) both as dopent reagents used for preparation of $Cu_xZn_{(1-x)}Ce_3O_5$. Extra pure urea used as fuel. In a typical combustion synthesis[9-12] the stoichiometric amount of cerric ammonium nitrate, copper nitrate and zinc nitrate weighed by the electronic balance were mixed with 10 gm urea. The mixture of above reagents along with urea stirred in the pyrex dish (300 cm^3) using glass rod. The formation of semisolid then liquid form of mixture was obtained at room temperature. This mixture was then heated in air on hot plate at 100 ^{0} C. during continuous heating the solution boiled with foaming and frothing. Further the mass ignited itself to burn with flame and blackish gray color powder was obtained. Exhaust was on during the firing in

laboratory. The yield of powder as a mixed oxide is about 4 gm. Then the powder sintered at various temperatures (600 ^{0}C, 800 ^{0}C, 1000 ^{0}C) for about 3 hours.

X- ray diffraction of all the samples taken by Philips PW1710 diffractometer. Crystallite size calculated by using Scherrer's equation .The SEM micrograph studies were carried using SEM model JEOL-JSM 6360 Kolhapur. The grain size of sample was calculated using Cottrell's method which gives the relation between the number of intercepts of the grain boundary per unit length (PL) and total number of intercepts (n) as,

$$PL= (n/2\prod r) M \quad [1]$$

Where M is the magnification at which SEM micrograph is scanned. r is radius of circle and n is the number of grains in the circle. Using equation 1 the grain diameter L can be calculated as

$$L= 1 / PL-1 \quad [2]$$

This equation gives average grain diameter .The oxide powder of composition Cu0.5Zn0.5Ce3O5 was mixed with 2 % PVA binder and pressed into pellets of 1.58 cm diameter and 0.12 – 0.25 cm thickness under a pressure of 10 tons.The pellet was heated in air at 600 ^{0}C to remove adhesive.

Flat surfaces of the sample pellet were coated with silver paste for good ohmic contact. The D.C.resistivity measurements of the oxide were performed by means of two- probe method [13]. The resistivity of sample was calculated using the relation

$$\rho_{dc}= R\prod r^2 / t$$

Where R is ohmic resistance of oxide, r is the radius of sample and t is the thickness of sample. The activation energies were computed from the plot of log ρ_{dc} vs. temp. in degree centigrade.

RESULTS AND DISCUSSION

The X- ray diffraction patterns of the oxide samples show single phase cubical structure and only reacted constituents were present in the oxide. A typical spectrum of sample $Cu_{0.5}Zn_{0.5}Ce_3O_5$ is shown in Fig. 1

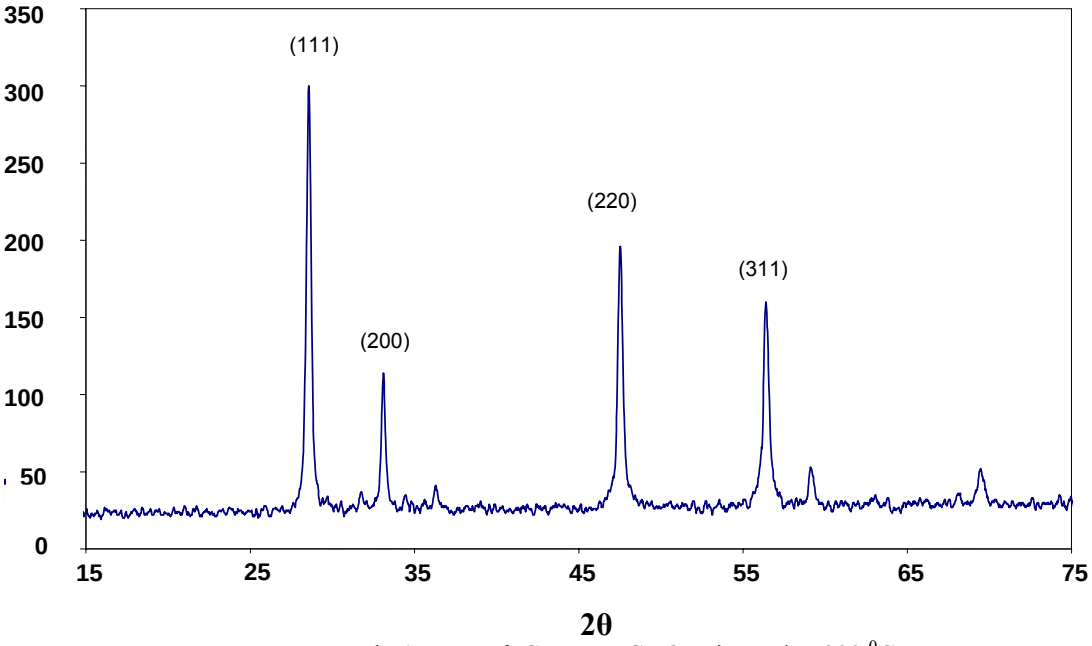


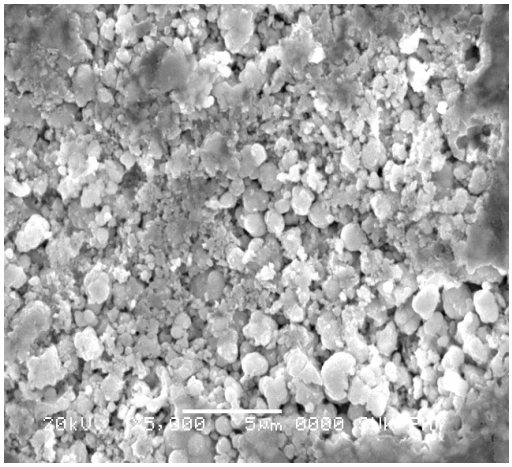
Fig 1-XRD of $Cu_{0.5}Zn_{0.5}Ce_3O_5$ sintered at 800 ^{0}C

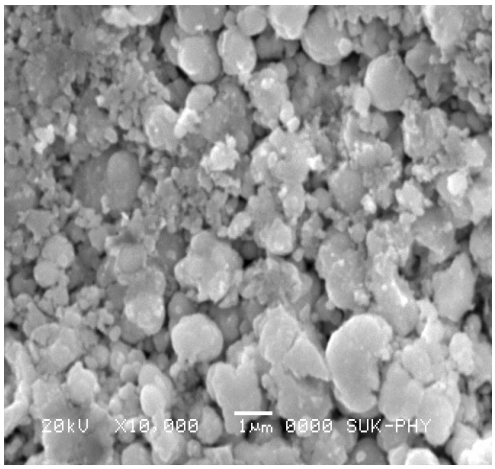
The presence of sharp picks in XRD pattern indicate the formation of polycrystalline oxide. The intense picks at 2θ degrees (28.735, 33.26, 47.685, 56.51) which are convinently be assigned to reflections corresponding to (111),(200),(220) and (311) planes respectively.All the crystallographic parameters and calculated grain size of particle of the oxide are shown in Table- 1.

Table-1

d- values (JCPDS)	d- values (observed)	hkl plane	Grain size
3.124	3.1042	111	20.0 nm
2.706	2.6915	200	
1.91	1.9056	220	
1.63	1.6271	311	

The SEM micrographs were used to observe surface morphology of oxide particle. Figure 2 shows micrograph images for the oxide sample sintered at 800 ^{0}C for 3 hours . Micrograph show polycrystalline compact and uniformly distributed nonporous particles of oxide . The average grain size of nanoparticle is 17 nm.





(b)

Fig.-2 SEM images of $Cu_{0.5}Zn_{0.5}CeO_5$ sintered at 800^0 C. for (a) 5000 magnification (b) 10,000 magnification

The temperature dependant variation of D.C. resistivity for the oxide $Cu_{0.5}Zn_{0.5}Ce_3O_5$ is shown in figure 3. Linear decrease in resistivity with increase in temperature showing semiconducting nature of oxide .The conduction mechanism in oxides is explained on the basis of Verwey de Bohr mechanism. The exchange of electrons takes place between the ions of same element present in more than one valency states and distributed randomly over equivalent crystallographic lattice sites. The decrease in resistivity with temparature is attributed to increase in drift mobility of the charge carriers.

CONCLUSION

Nanocrystalline $Cu_{0.5}Zn_{0.5}Ce_3O_5$ oxide is obtained successfully from their crystalline nitrates by simple solution combustion method at 100 ^{0}C . The average particle size obtained is 17-20 nm. The result indicates that oxide has electrical properties like D.C. resistivity. The oxide material offers inexpensive alternative in many new applications of semiconducting materials .The preparative method is simple, safe and time effective.

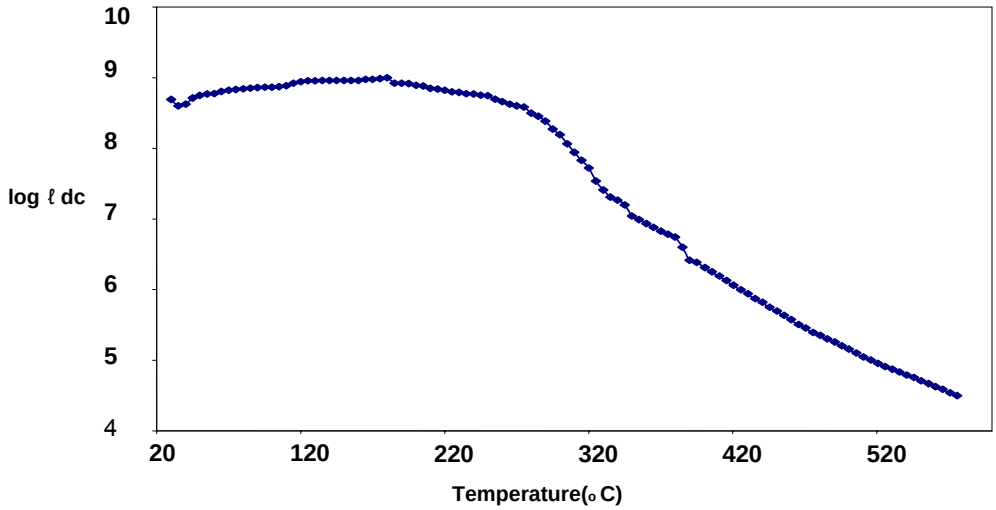


Fig.-3:Variation of dc resistivity of Cu 0.5 Zn 0.5 Ce3 O5 mixed oxide as a function of temperature

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