

# Effect on the Properties of NiO-GDC20 Nano-composite Anodes with the Variation of NiO for Solid Oxide Fuel Cell

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**Abstract:** In the present research work, cermet anode composites were synthesized for intermediate temperature operating solid oxide fuel cells. The anode composites synthesized as X (NiO) +1-X (Ce<sub>0.8</sub>Gd<sub>0.2</sub>O<sub>2-δ</sub>) where X = 30, 40, 45, 50 and 55 wt. %. For cermet anode materials, nickel oxide (NiO) and 20mol% gadolinium doped ceria, GDC20 (Ce<sub>0.8</sub>Gd<sub>0.2</sub>O<sub>2-δ</sub>) nano powders were mechanically mixed and calcined from 600 to 750 °C. Further, these composite powders were pelletized and sintered at 1300°C. The structure, phase and structural parameters of anode samples were carried out by XRD. Micro-hardness of the samples were measured by Vickers micro-hardness test and observed that microhardness of anode composites with the variation of NiO varying from 8.29Gpa to 12.02 Gpa. Electrical properties of cermet anode characterized using four probe dc- conductivity measurements and impedance spectroscopy analyser. The dc conductivity and their activation energies of the anode samples were computed from 500 - 700 °C. The activation energies varying from 0.52 to 0.76 eV and dc conductivity is attained maximum at x = 45 wt. % i.e. 9.96X10<sup>-2</sup> S/cm at 700°C. The complex impedance data and ac conductivities of grain and grain boundary were calculated. The activation energies of A.C conductivity have observed that decrease with increase in temperature and frequencies.

**Keywords:** Activation energy, cermet anode, conductivity, Micro-hardness, SOFC.

## I. INTRODUCTION

Solid oxide fuel cells (SOFC) convert chemical fuel to electrical energy via an electro chemical reaction and these are zero-pollution electric power generators (S.C. Singhal, et al, 2000). Over the past decade, a lot of efforts have been devoted for making of high efficiency SOFC. Nanostructured components have been attracting much in recent years due to

their enhanced properties compared with bulk materials for use in advanced energy conversion and storage applications such as solid oxide fuel cell (SOFC). For fabrication of Solid oxide fuel cell required solid electrolyte, composite electrodes such as anode and cathode. Many of the investigations on SOFC, reveals that composite electrodes are essentially physical mixtures of two or more solid phases for mixed conductivity that is ionic and electronic conductivity is vital in batteries and SOFC (Zhongli Wu, et al, 1996). In SOFC, electricity is generated due to the reaction between a fuel supply (hydrogen fuel at anode) and an oxidizing agent (cathode).

In recent years, a lot of efforts have been devoted to lower the operating temperature to the intermediate range i.e. 600–800°C of SOFC, because of high operating temperatures led to numerous problems related to compatibility of materials that result in higher costs, thermal mismatch between the cell components, degradation problems and slow start-up (L. Almar, et al, 2013).

The anode supported SOFC's are vital candidate to operate at Low or Intermediate temperatures (LT/IT) SOFC's i.e. from 500°C to 700°C, subsequently improve the performance, reliability, and long operational life of the SOFC than high temperature SOFC (L. Almar, et al, 2013; Chourashiya, M.G et al, 2010). Regarding the anode, the most commonly used anode at high temperatures is Nickel- Yttrium stabilized zirconia (Ni-YSZ) and is usually substituted by an alternative metal/oxide cermet to get excellent catalytic performance together with a good thermochemical compatibility with a ceria-based electrolyte makes ceria cermets are a typical choice anode for IT- SOFCs i.e. T < 800°C (S. Zha, et al, 2004). For an anodic supported SOFC, an anode must have electronically as well as ionically conducting, good catalytic activity or high electrochemical for fuel oxidation, proper porosity at the microstructure in reduced environment at operating temperature to supply fuel and remove reaction products (Zhonglin Wu, et al,

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1996; L. Almar, et al, 2013; L. Almar, et al, 2013; Chourashiya, M.G et al, 2010; Changsheng Ding, et al, 2009; S. Zha, et al, 2004).

In the present research, cermet anode composites were prepared using NiO (metallic) and 20mol% gadolinium doped ceria (ceramic) i.e.  $Ce_{0.8}Gd_{0.2}O_{2-\delta}$ , for ideal thermal expansion match with the  $Ce_{0.8}Gd_{0.2}O_{2-\delta}$ (GDC20) electrolyte at operating temperatures. This composite, increase the length of triple phase boundary (TPB) for improves electrode catalytic reactions and thereby fuel cell performance ( S. Zha, et al, 2004 ; W.Z. Zhu, et al, 2003; S. Zha, et al, 2004 ; Sun-dong Kim , et al, 2006; Macedo Daniel, et al, 2012).

In the NiO-GDC20 cermet anode composite, the metal content nickel oxide (NiO) for catalytic reactions that is oxygen activation, besides that provides good electrical conductivity and ceramic content 20mol% gadolinium (GDC20) perform as the matrix to support the catalyst (NiO) and prohibit the agglomeration of Ni metal content at the operating temperatures (Chourashiya, M.G et al, 2010; Changsheng Ding, et al, 2009; S. Zha, et al, 2004; W.Z. Zhu, et al, 2003). The conductivity of the anode composite material is the contribution of the ionic (GDC20 phase) and electronic conductivity (metal nickel Phase). The anode performance is mainly depend on its composition, microstructure and fabrication process in compliance with that find the better composition cermet anode material (Sun-dong Kim, et al, 2006).

However it has been never reported the wt% variation of NiO with GDC20 in NiO-GDC20 nanocomposite. In this work, cermet anode materials prepared by using the nano powders of NiO and GDC20 for good microstructure. The NiO and GDC20 are mixed and processed by mechanically mixing method.

## II. EXPERIMENTAL

The synthesis of NiO and Gadolinium doped ceria (GDC20) nano powders has been reported in detail (M.Narsimha Reddy, et al, 2016). The anode composites were prepared using 30nm and 32nm crystallite size nanopowders of NiO and GDC20 respectively, as synthesized nanopowders were mixed as X (NiO) +1-X ( $Ce_{0.8}Gd_{0.2}O_{2-\delta}$ ) where X = 30,40, 45, 50, 55 and 60 wt.%. The mixed powders were ground in agate mortar for 3hrs and calcined at the temperature of 750°C for 4hrs and these samples were pressed into cylindrical pellets with the diameter of 10mm and thickness of 1.5- 2.5mm using hydraulic press machine under pressure of 3 tons/cm<sup>2</sup>. The pellets were sintered at the temperature of 1300°C for 4hrs and in the sintering process, up to 800°C used the heating rate of 5°C /min and from 800 to1300°C used the heating rate reduced to 2°C /min.

Anode samples prepared with X(NiO)+(1-X) $Ce_{0.8}Gd_{0.2}O_{2-\delta}$  where X = 30,40,45,50,55 and 60wt.%, compositions are coded as Anode A1, A2, A3, A4, A5 and A6 ( herein after called ) respectively. The crystal structure, lattice parameters and crystallite size of the anode samples were determined from the X-ray diffraction (Phillips-3710 powder XRD) using CuK $\alpha$ 1 radiation ( $\lambda = 1.5406 \text{ \AA}$ ) over the range 20-80° of 2 $\theta$  with the

scanning rate 5°C/min. The relative densities of anode samples A1, A2, A3, A4,A5 and A6 are 89.8%, 92%, 93.6%, 94% ,93% and 92% respectively at sintering temperature 1300°C, calculated using Archimedes method. The samples purity and surface morphology of the samples were studied by using SEM with EDAX (ZEISS).

Micro-hardness of the anode composites were characterized by Vickers micro-hardness test to find the chemical stability of the anode composites. Vickers hardness test consist of pyramid form diamond indenter with the square base and an angle of 136° between the opposite faces subjected to load on test specimen dwell time 10 seconds. Micro-hardness measured with three diamond indentations on the surface of the pellets at 500 grams load.

For electrical properties, pellets were coated with platinum paste on both the surfaces using dc sputtering unit, Q150TS, Quorum, UK. The electrical properties of the samples viz., dc-conductivity and ac-conductivity were characterized using dc four probe method and Impedance spectroscopy respectively. The impedance spectroscopy is a powerful technique to characterize electrical properties of materials. This technique is based on analyzing the ac response of a system to a sinusoidal perturbation and subsequent calculation of impedance as a function of the frequency of the perturbation. Complex impedance analysis is useful to evaluate and separate the contributions of various processes such as electrode effects, grain effects and the interfaces i.e grain boundaries and the overall electrical properties in the frequency domain due to electrode reactions at the electrode-sample interface and migration of ions through the grains and across the grain boundaries in anode composite materials. Activation energies ( $E_a$ ) were estimated from the temperature range 500°C-700°C, which is the expected intermediate operating temperature of SOFC's.

## III. RESULTS AND DISCUSSION

### A. X-ray diffraction

Crystal structure and phase composition of cermet anode samples were confirmed using X-ray diffraction (XRD). The X-ray diffraction patterns of all anode composites have shown in Fig.1, the peaks compared and indexed accordingly with JCPDS file no# 01-075-0162 and JCPDS file no.# 01-78-0643 for for GDC20 and NiO respectively, in NiO –GDC20 composites . XRD patterns of all NiO –GDC20 composites reveals that multiphase with GDC has fluorite cubic structure (Fm-3m group) and NiO also has cubic structure (Fm-3m group) (Chavan, A.U, et al, 2012; R.V Wandekar, et al, 2006).

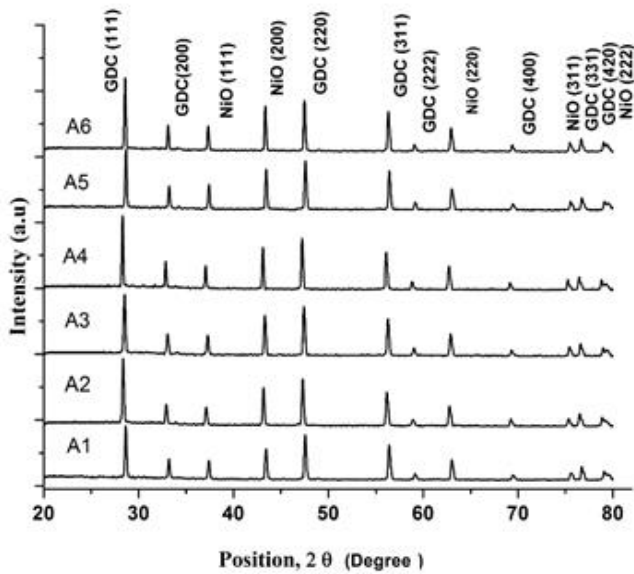


Fig. 1. XRD patterns of anode compositions A1, A2, A3, A4, A5 and A6.

From the XRD data, lattice parameters of NiO and GDC20 phase were calculated in the anode composites shown in Fig.1 and observed that lattice parameters are independent of ‘X’ and are  $4.178 \pm 0.005 \text{ \AA}$  and  $5.426 \pm 0.008 \text{ \AA}$  respectively for NiO and GDC20 at the  $1300^\circ\text{C}$  (Chavan, A.U, et al, 2012 ).

The avg. crystallite size (D) of the anode samples were estimated by the Scherrer’s formula,

$$\text{i.e. } D = \frac{0.9\lambda}{\beta \cos\theta} \rightarrow (1)$$

Where  $\lambda$  - Radiation wavelength  $\text{Cu K}\alpha = 0.15406 \text{ nm}$ ,  
 $\theta$  - Diffraction angle and  
 $\beta$  - Full width half maximum (FWHM).

The calculated, average crystallite size of NiO and GDC20 phase have shown in Table I. The crystallite sizes of NiO and GDC increased a few nm with the variation of NiO content up to  $X = 50 \text{ wt}\%$  and slightly decreasing trend have shown above  $X = 50 \text{ wt. \%}$  at  $1300^\circ\text{C}$ .

Table I. Avg. Crystallite size of anode compositions

Anode samples	Crystallite size, D ( in nm)	
	NiO	GDC20
A1	40.85	47.42
A2	42.28	47.73
A3	46.07	49.86
A4	50.69	52.68
A5	44.51	48.40
A6	41.26	48.05

B. Microstructure analysis

The SEM micrographs of the anode samples A3 and A4 are shown in Fig.2 and Fig.3, sintered at  $1300^\circ\text{C}$ . SEM images of all NiO-GDC20 nano composites shows that well connected NiO and GDC20 and uniformly distributed with small porosity, which is desirable for anodic reactions and electronic conduction. From SEM, observed that anode compositions has spherical grains and no agglomeration. The SEM with EDAX shown in Fig 4. and Fig.5, given that no other elements present except mixed elements in the anode compositions of A3 and A4, which reveals that samples are pure and support the XRD data.

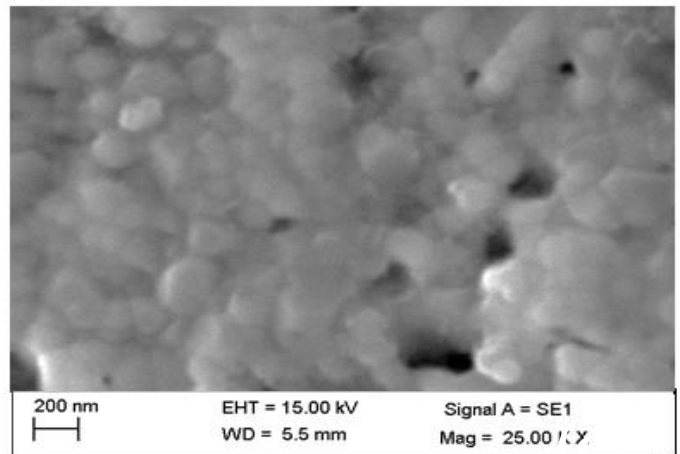


Fig. 2. SEM image of anode composition A3 (X=45wt %)

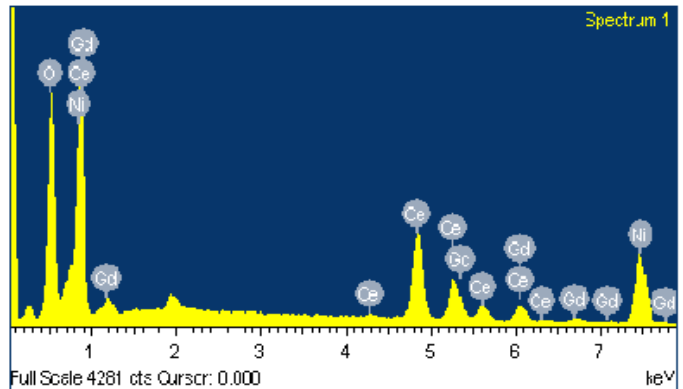


Fig. 3. EDAX of anode composition A3 (X=45wt %)

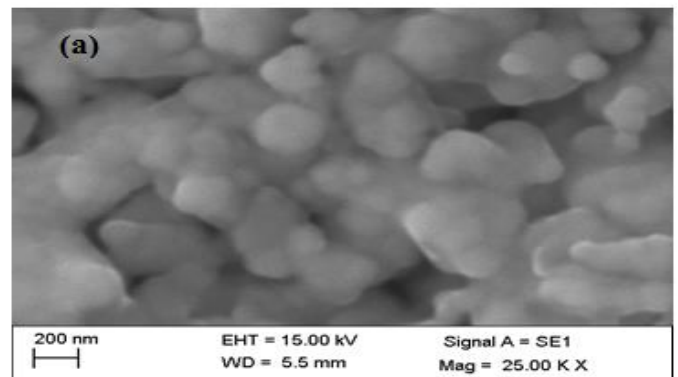


Fig. 4. SEM image of anode composition A4 (X= 50wt %)

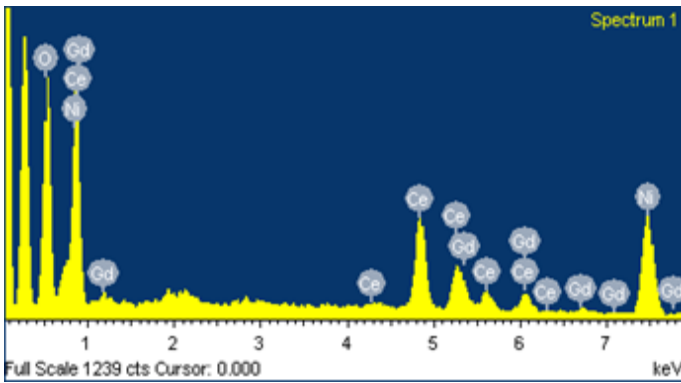


Fig. 5. EDAX of anode composition A4 (X= 50wt %)

C. Micro hardness

Vickers micro- hardness test is a best test for measure chemical stability or strength of material of composites in terms of micro-hardness. It is essential to know the micro-hardness of cermet anodes for choose good anode composites for chemical stability at microstructure under reduced environment for long operational life of SOFC's and also for withstand mechanical stress by cells use in the cell stack for power generation.

The average micro-hardness was obtained from the three indentations on the surface of anode material with the 500 grams load, dwelling time 10 seconds shown in Fig.6. Three indentations results three Vickers hardness numbers (VHN) and taken the average VHN and converted into GPa.

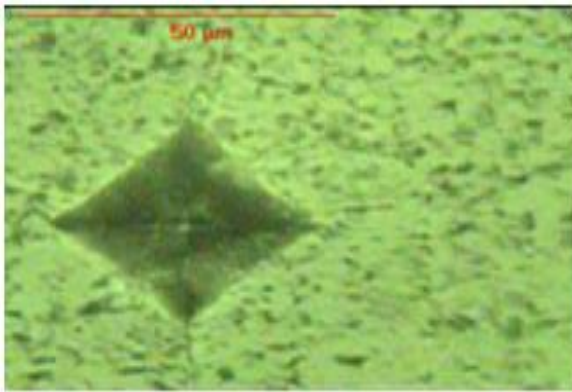


Fig.6. Diamond indentation on NiO-GDC20 compositions

Vickers Micro hardness number at each load is given by

$$H_v = \frac{1.8544 P}{d^2} \quad (2)$$

Where P is the applied load (Kg- f) and d is the diagonal length of the indentation in milli metres. Micro hardness of all pelletized anode composites sintered at 1300oC were measured and shown in Table II.

Table II. Avg. micro hardness of anode compositions

Anode samples	VHN	Hardness (GPa)
A1	845.4 ± 3	8.29 ± 0.2
A2	956.5 ± 3	9.38 ± 0.3
A3	1147.6 ± 4	11.25±0.4
A4	1225.4± 2	12.02±0.4
A5	1010.2 ± 5	9.90± 0.5
A6	948.3 ± 6	9.30± 0.6

In the anode compositions ,hardness has been increased with the increasing NiO concentration up to X = 50 wt.% , whereas gradually decreasing the hardness above X = 50 wt.% and got maximum average micro-hardness is at X = 50 wt.%, which has 12.02±0.4 GPa at 500g load.

D. Electrical properties

1) DC conductivity measurements

The dc electrical conduction is a thermally activated process, dc electrical conductivities of all cermet anode composite pellets sintered at 1300°C were carried out using four probe dc-electrical conductivity measurement in air in the temperature range 500°C -700°C, which is the probable operating temperature range of the NiO –GDC20 composites of anode supported SOFC's.

The variations of ln(σ<sub>dc</sub>) vs. 1000/T for anode composites A2, A3, A4 and A5 are shown in Fig.7.

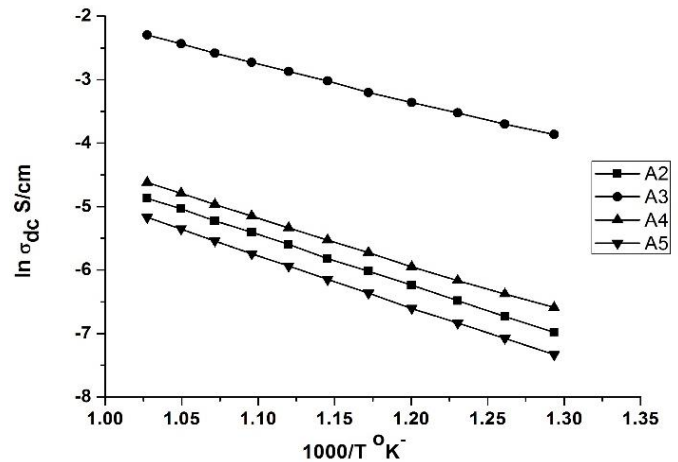


Fig.7 Arrhenius plot of dc conductivity of anode composites A2, A3, A4 and A5

The dc conductivities of all NiO –GDC20 composites were linearly fit into the Arrhenius relation and estimated the activation energies (E<sub>a</sub>) of the anode composites are given by

$$\sigma_{dc} = \sigma_o \exp\left(\frac{-E_a}{K_B T}\right) \quad (3)$$

Where σ<sub>o</sub> is the pre-exponential factor, E<sub>a</sub> is the activation energy, T is the absolute temperature and K is the Boltzmann

constant.

The dc conductivities of anode samples at 600 and 700°C and calculated activation energies from the temperature range 500-700°C are tabulated in the table III.

Table III. DC conductivities and activation energies

Anode sample	DC conductivity (S/cm) at temperature		Activation energy( $E_a$ ) in ev
	600°C	700°C	
A2	0.0030	0.0076	0.68
A3	0.0488	0.0996	0.52
A4	0.0040	0.0098	0.64
A5	0.0022	0.0057	0.70
A6	0.00083	0.0019	0.76

The maximum in conductivity or minimum in activation energy of the anode samples are caused due to the interaction of the gadolinium and cerium ions and the oxygen vacancy or point defects. Hence, the electrical conduction with temperature in the cermet anode is controlled by the charge-carrying defects. For NiO concentrations below  $X=45\text{wt.}\%$ , the conductivity is predominantly ionic and above  $45\text{wt.}\%$ , it is predominantly electrical (typical of metals). The conductivity almost saturates above  $45\text{wt.}\%$  of NiO in NiO-GDC20 cermets, this shows the percolation limit (Jae-Ha Myung, et al, 2012). As the requirement of the anode for SOFC is that, it should be electronically as well as ionically conducting. In the present study, dc-conductivity of composite A3 attains maximum i.e.  $9.96 \times 10^{-2} \text{ S/cm}$  at 700°C. Anode materials have low NiO concentration, NiO-GDC20 connectivity is poor and NiO is distinctly distributed throughout the composite, as a result ionic conductivity is dominated over the electronic. It is also expected that proper composition of anode would increase the NiO-GDC20 contacts and results the leading conduction path along the interface between NiO and GDC, so that conductivities in the composites increased at  $x=45\text{wt.}\%$  and  $50\text{wt.}\%$ .

#### E. Impedance measurements

The impedance spectroscopy is a non-destructive highly sophisticated experimental technique for the electrical characterization and microstructural properties of solid ionic materials or cermet's.

In the present paper, anode composites A3, A4 electrical properties only discussed extensively using complex impedance data in the frequency range 1Hz -5MHz and temperature range 500-700°C. According to micro hardness and dc conductivity analysis results, anode composites A3 and A4 have shown better micro hardness and dc conductivity than other composites with respect to the variation of NiO at sintering temperature 1300°C. Hence, probable and better anode composite candidates are A3 and A4 for intermediate operating SOFC's.

The impedance spectrum is usually represented as real part of impedance ( $Z'$ ) versus imaginary part of impedance ( $Z''$ ) at different temperatures, which is called Nyquist or cole-cole plot. Each point in the complex impedance plot corresponds to different frequency in the measuring range. The Nyquist plots of the anode samples A3 and A4 are shown in Fig. 8 and Fig. 9 respectively. Obviously, these plots shows the variation of impedance as semicircle nature and decrease the size of the semicircle with increasing temperature.

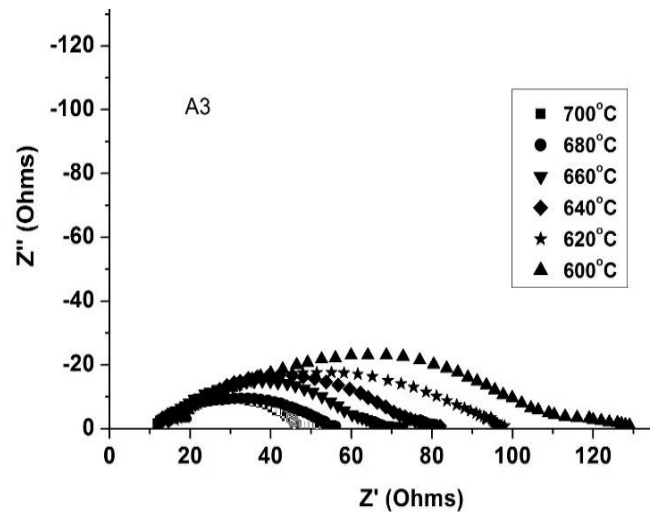


Fig.8 Nyquist plot of anode composite A3 at different temperatures

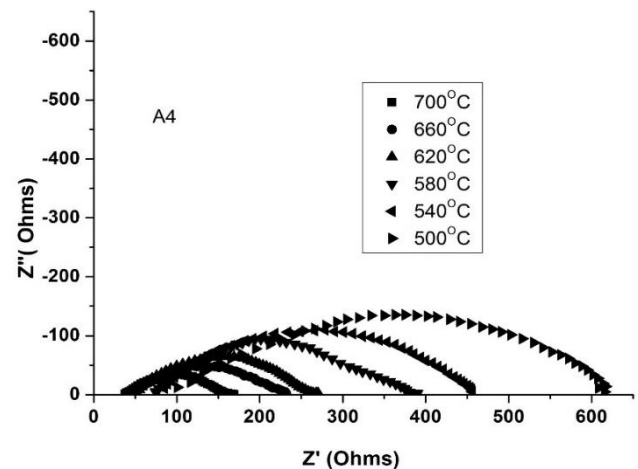


Fig.9 Nyquist plot of anode composite A4 at different temperatures

The complex impedance plots of A3 and A4 from the Fig.8 & Fig. 9 are resolved into two semicircles from the equivalent circuit model fit using impedance analysis software (Z view version 3.5) for extract the grain interior and grain boundary impedances to know the total conductivity of the samples at the measured temperatures. The equivalent circuit model used here is a series of  $R_s$ ,  $(R_g C_g)$ ,  $(R_{gb} C_{gb})$ , where  $R_s$  is a series resistance, RC is a parallel combination of resistance and capacitance for fit semicircles shown in Fig.10 and Fig.11. The plots in the complex

plane fit into two semicircles (not shown here) in the temperature range 500 - 700°C.

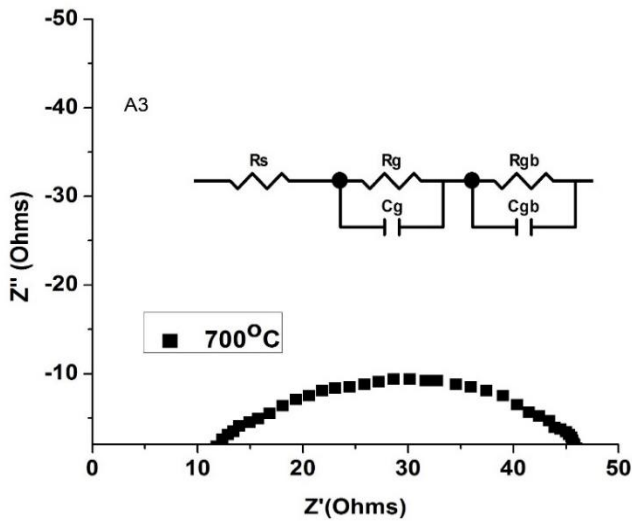


Fig.10 Nyquist plot of anode composite A3 at 700°C temperature and inset equivalent circuit fit.

A series resistor,  $R_s$  is added to account for the high frequency intersection of the first semicircle with the real part of  $z^1$  axis. The high frequency semicircle represents the grain interior property of the material arising due to the parallel combination of grain resistance  $R_g$  and capacitance  $C_g$ , another semicircle at low frequency domain of the impedance is due to the grain boundary effect which is equivalent to the parallel combination of grain boundary resistance,  $R_{gb}$  and capacitance,  $C_{gb}$ .

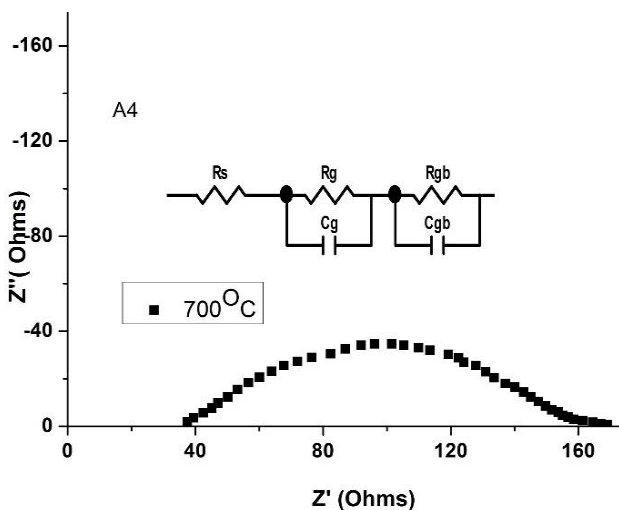


Fig.11 Nyquist plot of anode composite A4 at 700°C temperature and inset equivalent circuit fit.

The conductivity of anode composite contributions of the microstructural inhomogeneity's can be separated by assigning the values of resistance (R) and capacitance (C) to each of them in equivalent circuit for good fit from the temperature range

500-700°C. Calculated resistances of grain interior,  $R_g$  and grain boundary,  $R_{gb}$  of the samples A3 and A4 are shown in Table IV and V.

The ac conductivities of grain interior and grain boundary of the samples were calculated by

$$\sigma = \frac{t}{Z' \times A} \quad (4)$$

Where  $t$  is thickness and  $A$  is the surface area of the sample

$Z'$  is the impedance on the real axis ( $= R_g$  or  $R_{gb}$ )

$\sigma$  is the conductivity of grain or grain boundary

The anode composite of A3 pellet sintered at 1300°C having thickness 1.9 mm and diameter 9.6 mm was used for complex impedance analysis. Conductivities of grain interior (GI) and grain boundary (GB) are calculated using equation (4) from the temperature range 500-700°C are shown in table IV.

Table IV: Grain interior and grain boundary resistances and conductivities of anode composite A3

Temperature (°C)	Resistance ( $\Omega$ )		Conductivity(S/cm)	
	$R_g$	$R_{gb}$	$\sigma'_g$	$\sigma'_{gb}$
700	13.64	20.47	0.0193	0.0128
660	19.98	29.42	0.0131	0.0089
620	30.86	45.28	0.0085	0.0058
580	49.29	67.63	0.0053	0.0039
540	79.20	102.2	0.0033	0.0026
500	124.8	152.9	0.0021	0.0017

The conductivity of grain interior (GI) and grain boundary (GB) at 700°C is  $1.93 \times 10^{-2}$  S/cm and  $1.28 \times 10^{-2}$  S/cm respectively.

The plot linearly grain and grain boundary conductivities with inverse temperature as  $\ln(\sigma')$  Vs.  $1000/T$  i.e. Arrhenius curves for anode composites A3 are shown in Fig.12. where GI is Grain interior, GB is Grain boundary and  $T$  is the total conductivity.

The slope of the  $\ln(\sigma')$  Vs.  $1000/T$  of grain interior (GI) and grain boundary (GB) are calculated from the linear fit for the activation energies ( $E_a$ ) of the grain interior and grain boundary of anode composites A3. The activation energies of the grain interior, grain boundary and total ac conductivity of A3 are 0.63 eV, 0.69 eV and 0.62 eV respectively.

The anode composite of A4 pellet sintered at 1300°C having thickness and diameter is 1.76mm and 9.42 mm respectively, used for complex impedance analysis. Similarly, Conductivities of grain interior (GI) and grain boundary (GB) are calculated using equation (4) from the temperature range 500-700°C are shown in Table V.

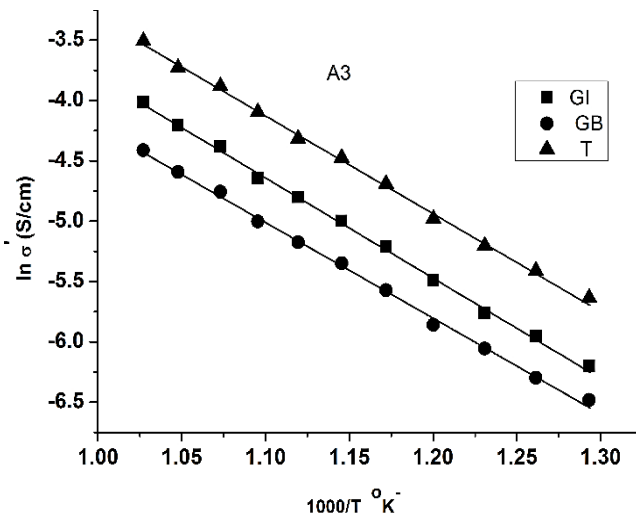


Fig.12 Arrhenius plot of grain interior, grain boundary and total conductivities of anode composite A3

Table V Grain interior and grain boundary resistances and conductivities of anode composite A4

Temperature (°C)	Resistance (Ω)		Conductivity(S/cm)	
	$R_g$	$R_{gb}$	$\sigma'_g$	$\sigma'_{gb}$
700	29.12	54.79	0.0086	0.0046
660	39.44	90.02	0.0064	0.0028
620	55.75	131.68	0.0045	0.0019
580	83.47	192.87	0.0030	0.0013
540	121.84	272.51	0.0021	0.0009
500	187.56	412.70	0.0013	0.0006

The conductivity of grain interior (GI) and grain boundary (GB) at 700°C is  $8.6 \times 10^{-3}$  S/cm and  $4.59 \times 10^{-3}$  S/cm respectively. The plot linearly grain and grain boundary conductivities with inverse temperature as  $\ln(\sigma')$  Vs.  $1000/T$  i.e. Arrhenius curves for anode composites A4 are shown in Fig. 13.

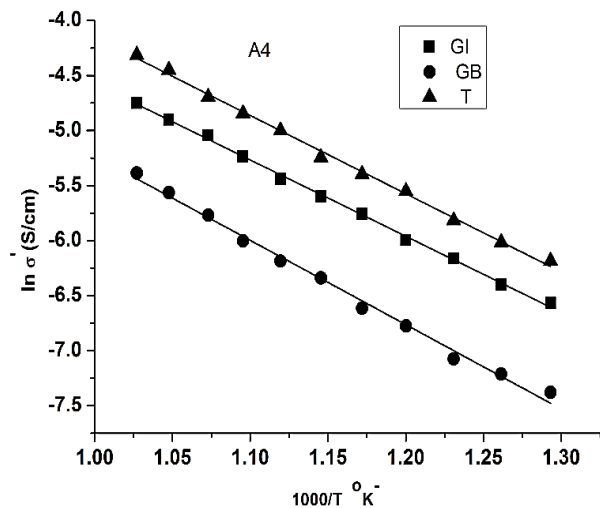


Fig.13 Arrhenius plot of grain interior (GI), grain boundary (GB) and total conductivities (T) of anode composite A4

The slope of the  $\ln(\sigma)$  Vs.  $1000/T$  of grain interior and grain boundary are calculated from range of temperature 500-700°C for the activation energies ( $E_a$ ) of the grain interior and grain boundary of anode composites A4. The activation energies of the grain interior, grain boundary and total ac conductivities are 0.68 eV, 0.70 eV and 0.67 eV respectively. The activation energy for total conduction should come from mainly from three sources such as the enthalpy of migration of oxygen ions ( $\Delta H_m$ ), the association enthalpy of complex defects ( $\Delta H_a$ ) and the activation energy for the grain boundary conduction ( $E_{gb}$ ).

The thickness of the samples also plays the critical role in impeding the electrical performance of samples.

The total AC conductivities of A3 and A4 from the grain interior and grain boundary conductivities are  $3.21 \times 10^{-2}$  S/cm and  $1.32 \times 10^{-2}$  S/cm respectively.

### CONCLUSIONS

- XRD reveals that NiO-GDC20 anode composite has multiphase. NiO and GDC20, both have cubic structure.
- Micro-hardness measurements given that better strength or chemical stability of anode composite are A3 and A4 that is  $x = 45\text{wt}\%$  and  $50\text{wt}\%$  of NiO, its hardness is 11.25 GPa and 12.02GPa respectively at 500g load and more than other NiO compositions sintered at 1300°C.
- Electrical characterisations (dc and ac conductivity measurements) given that good conductivity and least activation energy anode composite is A3 ( $x=45\text{wt}\%$ ) and A4 has better conductivity and least activation energies at measured temperature range 500 - 700°C.
- If we compromise a little less micro hardness, anode A3 is promising candidate or compromise less conductivity than A3, anode composite A4 is better candidate for anode supported SOFC's at intermediate operating temperatures.
- Further, research of symmetric cell characterization for anode supported SOFC and over a functional ceramic anode support such as Ni-GDC20 (reduction of NiO-GDC20 in H<sub>2</sub> atmosphere) decides, better candidate of anode composites is A3 or A4 and finds promising application in intermediate temperature SOFC's.

### ACKNOWLEDGEMENTS

The authors are very much thankful to the Department of Physics, Osmania University and DMRL, Hyderabad, Department of Physics, NIT Warangal and Department of Material Science and Metallurgy, IIT Mumbai and Department of Physics, R.T.M Nagpur University, Nagpur for providing experimental support.

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