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HYGROSCOPIC PROPERTY OBSERVED IN $Nd_2 Si_2 O_7$ COATINGS

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Abstract: $Nd_2 Si_2 O_7$ coatings have been prepared on Si (100) wafers by screen-printing method. The 80μm thick coatings were prepared from a mixture paste of high grade SiO_2 and Nd_2O_3 powders. The samples were characterized, by X-ray diffraction, IR-Spectrometry and optical microscopy. Tetragonal Type-A structure of $Nd_2 Si_2 O_7$ coatings with lattice constants a=b=6.8Å and c=24.7Å was obtained for samples sintered at 1000°C for 24 hrs. The porous $Nd_2 Si_2 O_7$ coatings were found to be hygroscopic. The moisturized coatings indicated strong IR absorption in the 1600-3000cm^{-1} range.

Keywords: Coatings, hygroscopic, IR-spectroscopy, screen printing, sintering, Type-A structure

INTRODUCTION

Due to superior magnetic [Maqsood *et al.* 1994] and optical [Maqsood 1981] properties, the rare-earth compounds have attracted considerable interest of the scientific community during the last two decades. The unique properties of these compounds emerge from the gradual filling of the $4f^n$ electronic states of the rare-earth elements. These compounds, which also include rare earth silicates, have nearly identical chemical character. So the study of any one rare-earth would serve as the representative of the whole family. In the present study Nd was chosen as the representative of the rare-earth family. Although fairly large number of reports have appeared in the scientific literature about the bulk [Felsche 1970, Sun *et al.* 1987] and single crystal [Ito and Johnson 1968, Maqsood *et al.* 1979] rare-earth silicates yet reports about rare-earth films or coatings are scarce [Maqsood 2000]. In the present paper we report the hygroscopic property observed in the screen-printed $Nd_2 Si_2 O_7$ coatings.

MATERIALS AND METHODS

Reagent grade powders (all from Aldrich USA; 99.99% pure) of Nd_2O_3 and SiO_2 were dried at 250°C, weighed by a digital balance (having 10μgm resonation) were mixed to give a final chemical formula of $Nd_2Si_2O_7$. After thorough grinding by an agate mortar and pestle, a paste was prepared by adding a small quantity of ethanol. The paste was applied to a 30μm mesh size printing screen to give 80μm thick coatings on ultrasonically cleaned single crystal silicon wafers with (100) orientation. The samples were dried in vacuum at 160°C for 1hr. The dried samples were then shifted to a muffle furnace equipped with automatic temperature controller. The samples were sintered at 800-1200°C for varying time intervals (10-140hrs). The film structure was studied by monochromatic X-ray diffraction (XRD) method. The bonding information and effect of moisture contents were investigated by IR spectrometry in the 650-4000 cm^{-1} range. The surface topography was studied by optical microscopy.

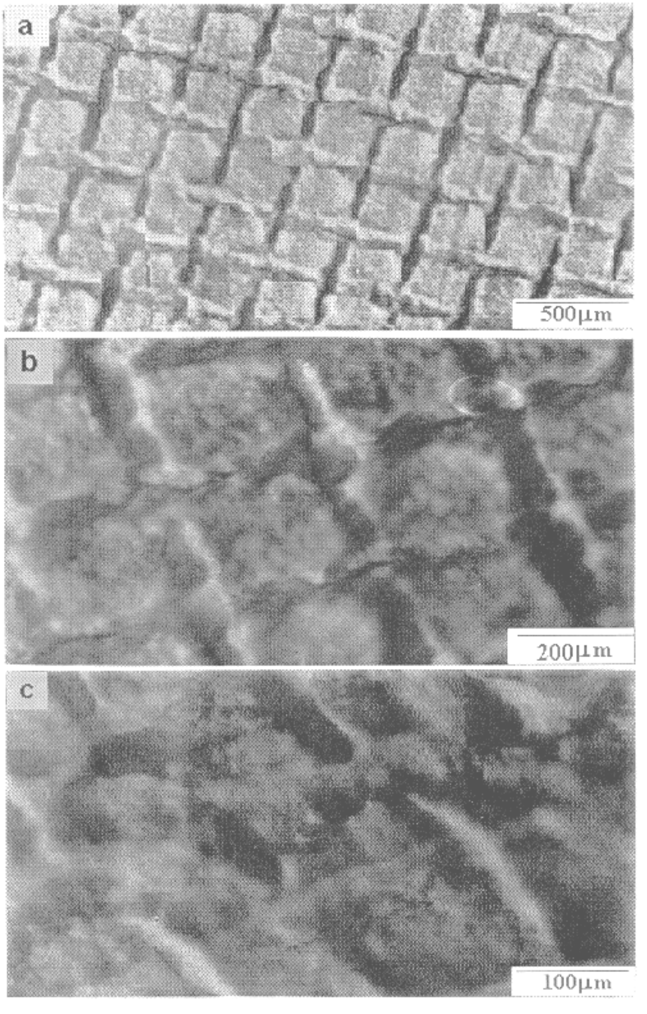

Fig. 1: Optical micrographs of 80μm thick $Nd_2Si_2O_7$ coatings sintered for 24 hrs: (a) at 1000°C, (b) and (c) at 800°C.

RESULTS AND DISCUSSION

Chemically or mechanically pretreated silicon (100) wafers showed better adhesion to the coatings. Although as deposited coatings were rough and crack free yet sintering above 800°C made the coating surface smooth but occasionally developed micro-cracks at the grid positions. Fig.1a shows the topography of the coating sintered at 1000°C for 24 hrs while Figs. 1b and 1c are the optical micrographs of the samples sintered at 800°C for 24hrs in air at two different magnifications. Coatings sintered below 800°C did not show any appreciable change in the topography of the samples. X-ray diffraction studies did not indicate any sign of solid-state reaction as no tetragonal or orthorhombic phases were detected at this temperature. However above 800°C a clear change, both in surface morphology as well as in microstructure crystallinity, as indicated by X-ray diffraction measurements, was obtained. Fig. 2 shows X-ray diffraction patterns of the $Nd_2Si_2O_7$, samples sintered at 1000, 1100, and 1200°C all for 24 hrs in air. It was critically noted that tetragonal phase starts appearing around 900°C while tetra- to ortho- phase transformation takes place at 1100°C. The two phases co-existed up to 1200°C. Presence of prominent XRD peaks for the tetragonal phase as well as orthorhombic phase in the samples sintered at 1100^0C and 1200°C can be clearly seen in Figs. 2b and 2c respectively.

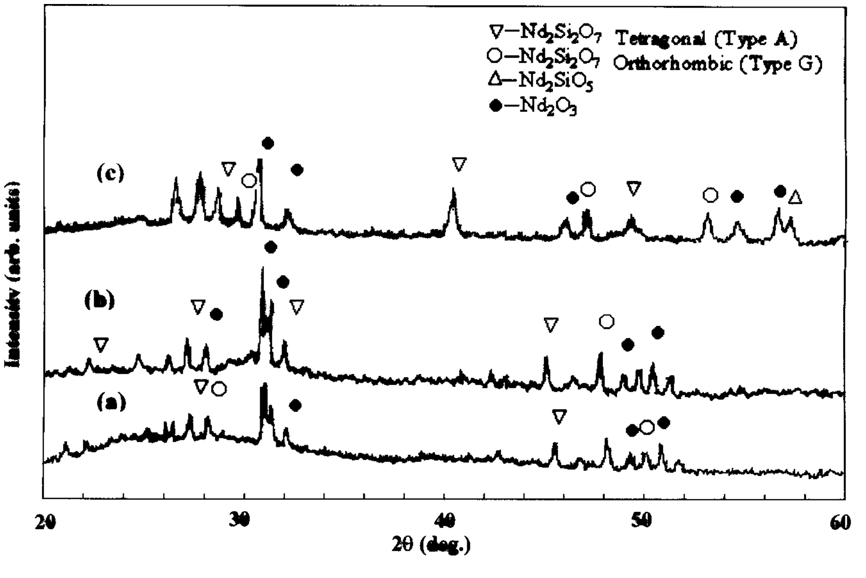

Fig. 2: X-ray diffraction patterns of $Nd_2Si_2O_7$ coatings deposited on the Si (100) substrates and sintered for 24hrs at (a) 1000ºC (b) 1100ºC and (c) 1200ºC

Moreover, it was observed that prolonged sintering gives better crystallinity for a given sintering temperature at and above 1000°C [Saeed *et al.* 2001, 2002]. IR-spectrometry of a specimen sintered at 800°C for 24hrs and exposed to water vapors showed broad absorption band between 1600-3000cm^{-1} (Fig. 3b). For the purpose of comparison IR-spectra for the dry sample and for pure water are also included in Fig. 3. Hence we critically note that porous coatings possess the hygroscopic property. The fabrication of porous samples was preferred as it facilitates the low temperature phase transformation of the coatings. In $Nd_2Si_2O_7$ compound the tetragonal phase normally appears at 1150°C [Felsche 1970] in dense bulk samples while the same phase in porous coatings appears at 900°C. Nonetheless the porous coatings are found to be hygroscopic, holding water vapors in the micro-pores of the coatings.

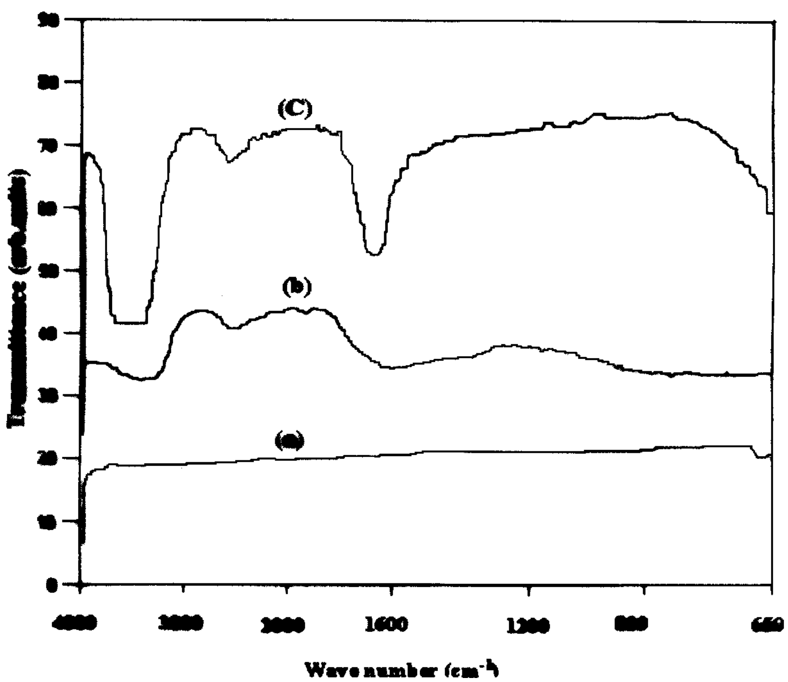

Fig. 3: Infrared transmittance of a $Nd_2Si_2O_7$ coating on Si (100) wafer: (a) Unmoisturized, (b) Moisturized, and (c) pure water sample (included for the purpose of comparison).

Monochromatic X-ray diffraction measurements for the hygroscopic sample, initially sintered at 1000°C for 24hrs, indicated that the coating mainly belong to tetragonal- type A phase with unit cell dimensions as a=b=6.8Å and c=24.7Å. The unit cell dimensions were not affected by the absorbed moisture, indicating that moisture is not altering the atomic arrangements of the tetragonal unit cells rather staying in the micro-pores of the sample.

CONCLUSIONS

In conclusion porous and nonporous $Nd_2Si_2O_7$ coatings have been successfully made on Si (100) wafers. The porous coatings show hygroscopic nature while non-porous coatings did not show any sign of moisture absorption. The porous coatings absorb oxygen during sintering more efficiently and facilitate tetragonal to orthorhombic phase transformation at relatively low temperature of 1000°C. However, due to porosity the coatings become hygroscopic, a property not usually desirable in the coating technology. The work is now in progress to overcome these difficulties and to achieve high-density high quality non-porous coatings on Si (100) and stainless steel substrates.

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References

Felsche, J. (**1970**) "Polymorphsim and crystal data of the disilicates of the form R.E. Si_2O_7", *J. Less-Common Metals*, 21, 1-14.

Ito, J. and Johnson, H. (**1968**) "Growth of $Nd_2Si_2O_7$ single crystals" *Am. Mineralogist*, 53, 1940-1947.

Maqsood, A. (**1981**) "Optical properties of $Er_2 Si_2 O_7$", *J. Mater. Sci.,* 16(11), 2198-2204.

Maqsood, A., Wanklyn, B.M. and Graton, G. (**1979**) "Growth of $Er_2Si_2O_7$ single crystals by flux method", *J. Crystal Growth*, 46(5), 671-679.

Maqsood, A., Arshad, M. and Maqsood, M. (**1994**) "Thermal transport properties of D- $Er_2Si_2O_7$ at room temperature", *J. Phys. D: Appl. Phys.*, 27, 1830-1834.

Maqsood, A. (**2000**) "Single crystal growth of polymorphic $Er_2Si_2O_7$ ceramics", *J. Mater. Sci. Lett.*, 19, 711-712.

Saeed, M., Aleem, K., Ghouri, S. and Chaudhary, K. (**2001**) "Structural properties of screen printed $Nd_2Si_2O_7$ thick films", *Proceedings of 7th International Symposium Advanced Matererials*, September 17-21, Islamabad, Pakistan, 459-461.

Saeed, M., Aleem, K., Ghouri, S. and Chaudhary, K. (**2002**) "Low temperature phase transformation in screen printed $Nd_2Si_2O_7$ thick films", *J. Mater. Sci. Lett.*, 21(21), 1649-1650.

Sun, K., Lee, H. and Risen, W.M. (**1987**) "Thermal properties of rare-earth silicates", *J. Non-Cryst. Solids*, 92, 145-149.