

# Surface characterization of float glass using Indus-1

G.S. Lodha, M.K. Tiwari, K.J.S. Sawhney, M.H. Modi, R.V. Nandedkar  
Synchrotron Utilization Section, Centre for Advanced Technology, Indore 452 013.

## Abstract

*The production of window glass is through float technique, which causes diffusion of tin into glass surface in contact with molten tin. Using Indus-1 storage ring, angle dependent reflectivity of the top and the bottom surface of float glass were measured in vacuum ultra violet/ soft x-ray region. Remarkable differences in refractive index were observed between the two sides. Surface impurities on the top and bottom surface were quantified using grazing incidence x-ray fluorescence measurements. The higher optical density of the bottom side was attributed fractional enrichment of iron along with tin diffusion.*

## INTRODUCTION:

Casting the molten soda lime glass on a molten tin bath produces window glass, popularly known as float glass. This process of fabrication leads to tin diffusion on the face of the glass in contact with molten tin. The other face, which is in contact with inert atmosphere, is weakly contaminated with tin. Thus float glass is composed of two composite surface layers and a bulk. It is important to know if change in optical properties and concentration profile of tin are correlated. Furthermore it is important to study if tin is the only impurity responsible for the change in optical properties.

The two surfaces of float glass have been extensively studied using a large range of surface sensitive techniques [1-7]. We have performed surface optical studies on the two sides of float glass using angle dependent EUV reflectivity measurements. These studies permit non-destructive investigation of vertical in-homogeneity and surface roughness. The technique has a high surface sensitivity of a few Å and is not hampered by surface charging. Surface impurities have been measured using grazing incidence x-ray fluorescence (GIXRF). This technique is ideal for the determination of high atomic number surface impurities.

The angle dependent reflectivity measurements in the wavelength range 70 to 200Å were carried on the EUV/soft x-ray reflectometry station [8] on Indus-1 synchrotron radiation source. The Indus-1 reflectometry beamline facility has been described elsewhere [9]. For GIXRF studies we have used indigenously developed setup described elsewhere [10].

The samples used were bottom and topside of 5 mm thick float glass. The bottom side was in contact with the tin bath during solidification. Observing the fluorescence under UV illumination easily identified the tin side. The samples were cleaned ultrasonically in methanol. To remove organic contamination on the surface, the samples were heated at  $\approx 140$  °C in an oxygen rich environment under UV illumination.

## RESULTS AND DISCUSSIONS:

GIXRF spectra of top (non-tin) and bottom (tin) side of float glass are shown in Figure 1. The excitation energy is 8.04 keV. The measurements were performed at a grazing

angle of  $0.21^\circ$ . This angle corresponds to the critical angle of silicon dioxide (a major constituent of float glass). At this angle, the penetration depth of the incident wavelength (1.54Å) is 400Å. Thus the fluorescence signals were highly surface sensitive. On the topside of float glass, only Ca K $\alpha$  peak was observed in the fluorescence spectrum. Calcium is part of the bulk composition of float glass. Bottom side fluorescence measurements show peaks of Sn L $\alpha$ , Ca K $\alpha$ , Fe K $\alpha$  and Fe K $\beta$  in addition to scattered peaks of incident radiation. The fluorescence of Sn L $\alpha$  (3.44 keV) and Ca K $\alpha$  (3.69 keV) overlap, due to limited resolution of 250eV of the energy dispersive detector. The spectrum unfolding was done using non-linear least square-fitting program. Concentrations of tin and iron were determined using iron and tin thin film standards. On the bottom side, tin and iron concentrations were 6.26  $\mu\text{g}/\text{cm}^2$  and 2.65  $\mu\text{g}/\text{cm}^2$  respectively. On the topside iron concentration was 0.015  $\mu\text{g}/\text{cm}^2$  and tin concentration was below the detection limit. These concentrations were in the top 400 Å of the float glass surface. The tin fluorescence on the bottom side decreased, at angles above the critical angle. The estimation of tin above the critical angle, becomes difficult, as the calcium concentration increases in the bulk, and there is a strong overlap of small Sn L $\alpha$  and strong Ca K $\alpha$  peaks.

The iron concentration on the bottom side increased sharply above the critical angle of  $0.21^\circ$ , indicating enrichment of iron in the bottom surface up to a few microns. At  $\theta=0.5^\circ$ , the probing depth is few microns. The iron concentration on topside was considerably lower and did not increase significantly as we moved from the surface to bulk.

Reflectivity measurements at 190 Å, of non-tin (top) and tin (bottom) side of float glass, along with the fitted reflectivity profiles are shown in Figure 2. The measurement wavelength was reasonably away from the silicon L edge (123Å). To avoid higher order contamination in the incident energy, Al L edge filter (170 Å cutoff) was used after the monochromator. The measured reflectivity showed higher critical angle for the bottom side, clearly indicating higher substrate density on this side. The (R –  $\theta$ ) curves were fitted by stratified layer formalism of Parratt [11]. Nevot-Croce [12] model was

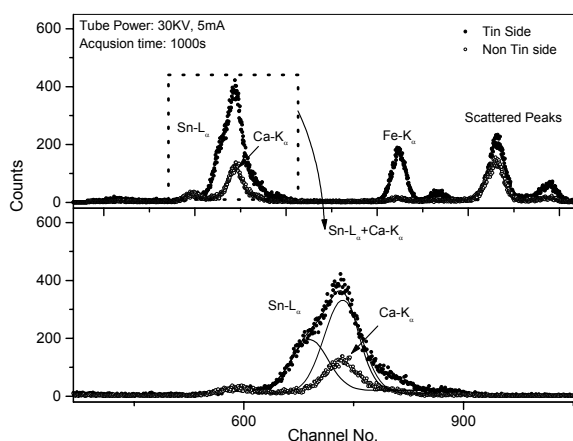


Figure 1: GIXRF measurement at incident energy of 8.04KeV of top and bottom side of float glass measured at an incident angle of  $0.21^\circ$

used to incorporate the surface and interface roughnesses in layered system with infinitely absorbing substrate.

Fitting the measured reflectivity at various wavelengths showed that a chemisorbed layer with an average thickness of 25 Å and a density of about 1.1 gram/cm<sup>3</sup> exists on both sides of float glass. The reflectivity of the topside float glass was fitted reasonably well using a substrate density of 2.2 gram/cm<sup>3</sup> and substrate composition of SiO<sub>2</sub>. The values of optical constants of the non tin side, derived by fitting the R-θ profile, are in good agreement with Diel et al [3]. For fitting the reflectivity of the bottom side, we modeled two layers above the substrate. The top most layer was modeled as a low density layer of thickness 28Å. GIXRF analysis clearly shows that Sn concentration on this side is more than 50 times the concentration on the top side. Iron concentration on this side is considerably higher. A high-density layer was modeled with a second layer of thickness of 17 Å and a composition of FeSn<sub>3</sub>. This composition was based on Fe and Sn concentration determined by grazing incidence XRF. The fitted value of density for the bottom side substrate was 2.8 gram/cm<sup>3</sup>. To account for these changes in density, we needed to change the composition to 0.95 SiO<sub>2</sub>, .025 Fe<sub>2</sub>O<sub>3</sub> and 0.025 SnO. These model compositions derived by EUV reflectivity fitting were in agreement with high concentration of tin and iron observed by grazing incidence XRF. The fitted profile in Figure 2 for the tin side is based on the above model.

The present study showed that the combination of EUV reflectivity and GIXRF provided a tool to study the variation in EUV optical constants of float glass, side. The comparative changes in EUV reflectivity are distinct compared to x-ray reflectivity measurement. [5] High atomic number impurities are increased in bottom side of float glass. Due to increase in tin and iron on the bottom side, there is an increase of refractive index in the EUV region. upto a few tens of Å. Fitting of EUV reflectance profile at multiple

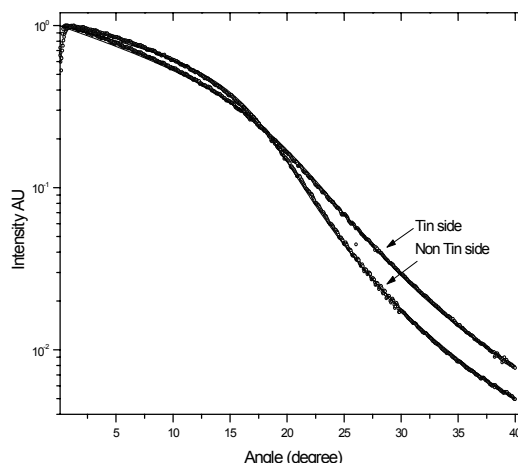


Figure 2: EUV reflectivity of tin and non tin side of float glass measured at 190Å. Open circle represents measured profile whereas solid line represents fitted model profile.

wavelength, clearly suggest that a low density layer of about 25 Å, and with a density of about 1.1 gram/cm<sup>3</sup>, exists on both sides of float glass. The optical constants of the bottom side are significantly higher compared to the top

#### REFERENCES:

1. B. Dugnoille, O. Virlet, Applied Optics, 33 (1994) 5653.
2. M.Hüppauff, B.Langel, J. Appl. Phys., 75 (1994) 785.
3. I.Diel, J.Friedrich, C.Kunz, S.Di Fonzo, B.R.Müller, W.Jark, Appl. Opt., 36 (1997) 6376
4. B.Yang, P.D.Townsend, S.A.Holgate. J.Phys.D:Appl. Phys. 27 (1994) 1757.
5. P.J.LaPuma, R.L.Snyder, S.Zdzieszynski, R.Brückner, Advances in X-ray Analysis, 38 (1995) 705.
6. K.F.E.Williams, C.E.Johnson, J.Greengrass, B.P.Tilley, D.Gelder, J.A.Johnson, J.Non-Crystalline Solids 211 (1997) 164.
7. P.D.Townsend, N.Can, P.J.Chandler, B.W.Farmery, R.Lopez-Heredero, A.Peto, L.Salvin, D.Underdown, B.Yang, J. Non-Crystalline Solids, 223 (1998) 73.
8. G.S.Lodha, V.K.Raghuvanshi, M.H.Modi, P.Tripathi, A.Verma, R.V.Nandedkar, Vacuum, 60 (2001) 385.
9. M.K.Tiwari, B.Gowrishnkar, V.K.Raghuvanshi, R.V.Nandedkar, K.J.S.Sawhney, (communicated).
10. R.V. Nandedkar et al (to appear in Current Science)
11. L.G.Parratt, Phys. Rev., 95 (1954) 359.
12. L.Nevot, P.Croce, Revue.Phys.Appl., 15 (1980) 761.

