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Enhanced Deposition and Reflective Properties of Thin Aluminium Films by Substrate Vibration

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Abstract: The influence of substrate's vibration during vacuum deposition of aluminium thin films on copper substrates was examined. Aluminium metal was evaporated in specially designed vacuum chamber using the hot-filament technique. Copper substrates were subjected to a vibration of 7.6 kHz during deposition. The Al coatings were identified using X-ray diffraction spectroscopy and scanning electron microscopy was used to examine the resulting microstructures deposited on the substrates. Coatings deposited under substrate vibration had fewer particles, spherical in shape and deposited over uniformly over the entire surface. This was not the case for the non-vibrated substrates, which tended to have much more densely packed granular shaped particles. The reflectivity experiments revealed that vibrated substrates were superior to the non-vibrated substrates by 28 %, while the difference in the thermal response was around 14 %.

Keywords: thin film deposition, reflection, vibrating substrate

1. Introduction

Many naturally occurring surfaces found in plants and animals show reflective properties which are used in a diverse range of applications such as assisting in heat regulation, camouflage and to highlight warning colourisation [1]. When engineering thin films for various applications, optical properties can also have a significant influence on the overall performance of the film and their application in devices, ultimately determines the device performance efficiency. Generally, optical properties of thin films are characterised by properties such as the refractive index, reflectivity and absorption index. The properties of vacuum deposited films are significantly influenced by the surface morphology of the thin film [2]. Studies have shown that the morphology of a thermally deposited thin film is dependent on the deposition rate [3], the type of substrate material [4, 5], the presence of surface contaminants and temperature [6]. The dominant parameter is the deposition rate which in turn is controlled by factors such as the formation of required species, temperature of the filament, mass transport in the vapour phase, temperature of the substrate, surface kinetics at the surface of the substrate, surface chemistry and processing parameters like pressure and temperature [2, 7-9].

More recently, studies have shown that the morphology and properties of thin films can be modified by *in situ* substrate vibration [10, 11]. These studies were able to show that *in situ* substrate vibration could be used as a controllable process parameter for changing overall film properties. However, the use of high frequency acoustic vibrations (sound and ultrasound) for refining crystallite morphology during metal solidification to improve material properties and product quality has been known for many years [12, 13]. Furthermore, studies have shown that sound and ultrasound vibrations can be used as an effective process

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treatment technique for enhancing both mass and heat transfer rates [14-17].

The *in situ* substrate vibration technique has been used successfully in forming both organic and inorganic thin films. Higher substrate temperatures maybe suitable for a variety of metallic thin films, since it promotes epitaxial growth, but in the case of semiconductor materials, the higher temperatures tend to reduce the sticking coefficient of the semiconductor molecules [18]. The lower sticking coefficient results in a significantly reduced nucleation rate, which in turn results in a less than satisfactory performance of the thin film based device [19, 20]. *In situ* substrate vibration as a method for modifying film morphology and properties is advantageous for organic compounds, because substrate temperatures must be kept relatively low to avoid any thermal degradation and instability common to many organic materials [11, 21, 22].

The optical properties of thin films deposited on *in situ* vibrating substrates have been the subject of a number of studies and have resulted in a number of potential device applications [23]. This preliminary study investigates a new application of this technique at 7.6 kHz and also studies the reflective and heating behaviour of thin aluminium films deposited on vibrating copper substrates. Aluminium (Al) is an important engineering material that is currently used in a wide range of industrial applications such as aerospace, automotive, ship-building and construction. Its wide range of applications comes from its high strength to weight ratio, excellent heat and electrical conductivities, and its corrosion resistance once it forms a protective oxide layer that forms naturally as Al reacts with atmospheric oxygen. This study in particular looks at the morphology of Al thin films deposited on non-vibrating and vibrating copper (Cu) substrates. The frequency of vibration used in this study was 7.6 kHz and falls mid way between the low frequencies studied by Parades *et al* (0 to 800 Hz) [11] for organic thin films, the fixed frequency of 22 kHz of Mohanchandra and Uchil [5] for semiconductor thin films and the 0–110 kHz range by Karim *et al* [6] for diamond-like carbon thin films. In this study, the improved reflectivity of the substrates with the deposited Al film is examined; a property that enables this coated material to be used in potential solar thermal applications.

2. Materials and Methods

2.1. Materials

All chemicals used in this work were supplied by Chem-Supply (Australia) and all aqueous solutions were made using Milli-Q® water (18.3 MΩ cm^{-1}) produced by an ultrapure water system (Barnstead Ultrapure Water System D11931; Thermo Scientific, Dubuque, IA). Cu substrates used in this study were made from oxygen free, high conductivity 70 µm thick copper foils. The Al wire used in the resistive heating circuit was 0.76 mm in diameter and 99.97% in purity was supplied by BDH Laboratory Supplies Australia.

2.2. Experimental

The evaporation of Al was carried out using a conventional in-house built bell jar type vacuum deposition system using a hot filament resistive circuit to provide thermal evaporation. For *in situ* vibration of the Cu substrate, the substrates were first cut from the Cu foil sheet, (80 x 50 x 0.07 mm) and then fixed between two piezoelectric disks, (38 mm and 15 mm in diameter and both 5 mm in height) which are electrically connected in parallel. The Cu substrate and piezoelectric disk assemble was then placed into the vacuum chamber and evacuated to a pressure of $8x10^{-2}$ mbar. A BWD Instruments Wave generator Model 141 generated the sound vibration signals, before undergoing amplification (x10). The amplified 7.6 kHz signal was then transferred to the piezoelectric/substrate assembly inside the chamber. There was also provision to monitor the frequency and amplitude of the generated signal via an oscilloscope, which was connected in parallel with the signal generator.

2.3. Characterisation techniques

2.3.1. X-ray diffraction (XRD) spectroscopy

X-ray diffraction (XRD) spectroscopy technique was used to examine and to identify the crystalline size and phases present in the pure Cu control substrates and the subsequent Al depositions on the sample Cu substrates. Spectroscopy data was recorded at room temperature, using a GBC® eMMA X-ray Powder Diffractometer [Cu K_α = 1.5406 Å radiation source] operating at 35 kV and 28 mA. The diffraction patterns were collected over a 2θ range of 20° to 60° with an incremental step size of 0.02° using flat plane geometry with 2 second acquisition time for each scan, with only the relevant 50º to 52º shown in this study. The crystalline size of the particles in the powders was calculated using the Debye-Scherrer equation [Equation 1] from the respective spectroscopy patterns.

2.3.2. Scanning electron microscopy (SEM)
The SEM technique was used to study the size, shape and morphological features of both the untreated Cu substrates and the substrates after Al deposition. All micrographs were taken using a JCM-6000, NeoScope™ with samples being individually mounted on SEM stubs using carbon adhesive tape.

2.4.3. Optical and thermal characterisation
The reflective properties of Al coated Cu substrates were carried using a Vernier SpectroVIS Spectrophotometer [Model SVIS] over a spectral range from 400 to 720 nm at room temperature. Each substrate was examined at least four times, each time a different location was selected. The laser light source used in the reflection experiments was a standard Red Laser Pointer, Class 2, 630-680 nm, <1 mW. Temperature measurements were made using a handheld Fluke IR Ti25 camera.

3. Results and discussions
3.1. XRD spectroscopy analysis
Analysis of the respective XRD patterns was used to identify and estimate the size of crystalline Al deposited on the underlining Cu substrate. The dominant peaks present on a representative sample occurred over a narrow 2θ range, which ranged from 50.2º up to a maximum of 51.0º as seen in Figure 1 and were identified as Al and Cu as listed in the ICDD database. Inspection of the XRD patterns presented in Figure 1 reveals the two peaks associated with the copper substrate located at 50.60º and 50.72º. The Al deposited without substrate vibration has one dominant peak located at 50.5º, while the Al deposited with substrate vibration has two distinct peaks. The first peak is located at 50.5º and has the largest intensity of the two. It is also located in the same 2θ position as the non-vibrating substrate. The second peak located at 50.62º is of particular interest since its intensity is around two thirds of the main peak located at 50.5º and can be clearly seen, unlike the case of the non-vibrating substrate. The crystallite size, $t_{(hkl)}$, of the Al depositions were calculated using the 50.5º peak for both vibrating and non vibrating substrates from the respective XRD patterns using the Debye-Scherrer equation [24]

$$t_{(hkl)} = \frac{0.9\lambda}{B\cos\theta_{(hkl)}} \quad (1)$$

where, λ is the wavelength of the monochromatic X-ray beam, *B* is the Full Width at Half Maximum (FWHM) of the peak at the maximum intensity, $\theta_{(hkl)}$ is the peak diffraction angle that satisfies Bragg's law for the (h k l) plane and $t_{(hkl)}$ is the crystallite size. Analysis revealed that the mean Al crystallite size for the non-vibrating samples was calculated to be 76 nm, while the crystallite size on the vibrating substrates was found to be 42 nm. XRD analysis has confirmed a significant reduction in crystallite size of around 44.7 % in the presence of substrate vibrations. During the XRD analysis, no characteristic peaks normally associated with impurities were detected and confirms successful pure Al deposition was achieved under the currently prescribed experimental conditions.

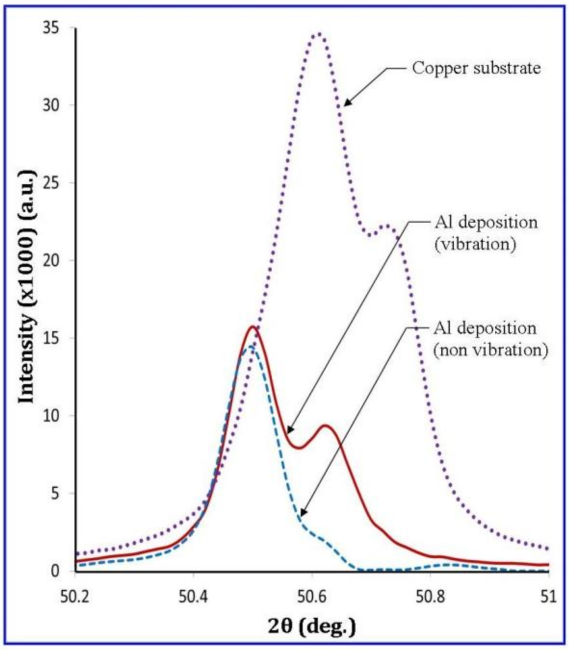

Figure 1 XRD analysis of aluminium deposited on copper substrate with and without substrate vibration.

3.2. SEM investigations
Figure 2 presents a comparison of representative micrographs of Al coated Cu substrates taken under the influence of vibration and without vibration. The top left-hand micrograph (a) presents a typical image of a surface coated with Al without substrate vibration and the top right-hand micrograph (b) is the coated surface deposited during substrate vibration

(7.6 kHz). Enlarged surface views of the respective substrates are shown in (c) non vibration and (d) vibration. Inspection of the coating deposited while the substrate was under vibration (d) reveals pronounced spherical morphology with particles typically around 100 to 150 nm in diameter. The non-vibrated substrates have a similar particle size range, but have different particle morphology. The particles tend to be more granular in shape and tended to be more elongated. Inspection of the coated surfaces in Figure 2 also revealed coatings deposited under vibration had fewer particles deposited over a given area, and as seen in Figure 2 (d) the deposited particles tended to be uniformly distributed over the surface. This was not the case for the non-vibrated substrates, which tended to have much greater coverage as seen in Figure 2 (c).

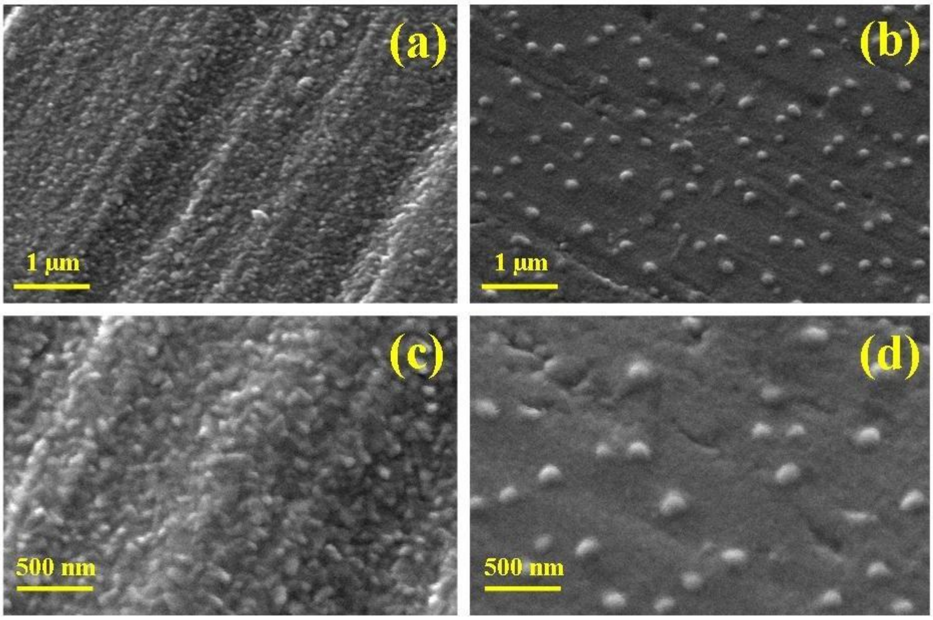

Figure 2 SEM micrographs of Al deposited on Cu substrates without (a) & (c) and with substrate vibration (b) & (d) at (7.6 kHz).

3.3. Reflective properties and thermal response

The reflective properties of Al films deposited on Cu substrates with and without substrate vibration were investigated using a standard 100 W incandescent light bulb as a light source. The results of two representative sets of spectroscopy measurements made during this study are presented in Figure 3. The upper set of four plots was taken at different locations on a substrate that was vibrating during Al deposition. The lower set of four plots was taken on a substrate that was not vibrated during deposition. Inspection of Figure 3 reveals that there was significantly more reflectivity over the entire spectrum from substrates that were vibrated during deposition. Maximum reflectivity occurred at 680 nm for both sets of substrates. Substrates that received vibration during deposition had a reflectivity on average 28 % higher than those which did not receive vibration.

Another interesting feature of the substrates was their thermal response and in particular the significant difference between substrates exposed to vibration to those that experienced no vibration. Figure 4 presents a graphical representation of the thermal response of two typical substrates exposed to a single 100 W incandescent light bulb. In each case the substrates were irradiated on one side by the light source, while the reradiated infrared radiation emmitted from the oposite side of the substrate was measured. Comparing the two plots it is clearly evident that the substrate exposed to vibration during Al deposition has improved thermal emmision properties. Calculating the temperature gradient for both substrates from the graph reveals that the vibrated substrate has a value of 0.07 ºC/min while the non vibrated substrate has a value of 0.06 ºC/min. This equates to an improvement of around 14 % if the substrate is subjected to a 7.6 kHz vibration during Al deposition.

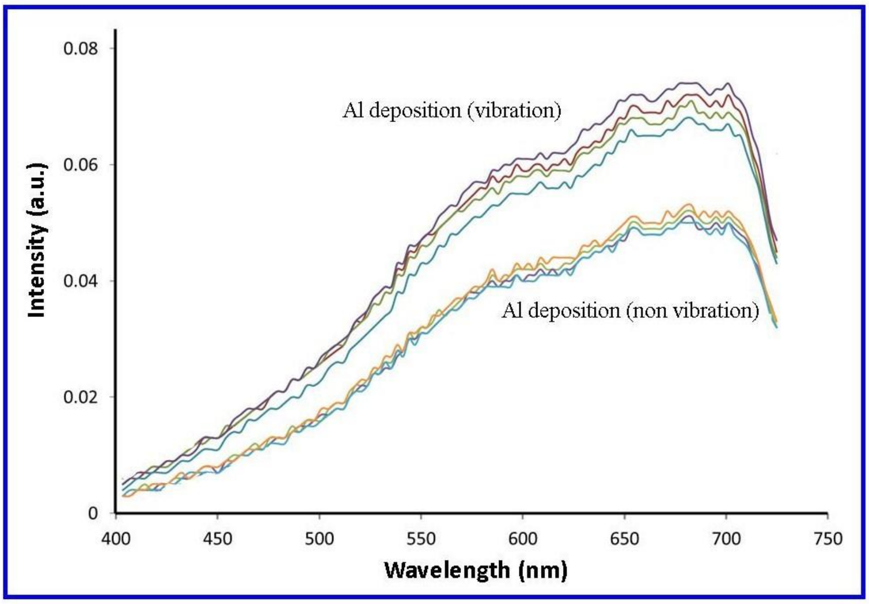

Figure 3 Representative reflectivity measurements taken of two types of Al coated substrates, (vibrated at 7.6 kHz and non-vibrated during Al deposition) subjected to irradiation from a 100 W incandescent light bulb.

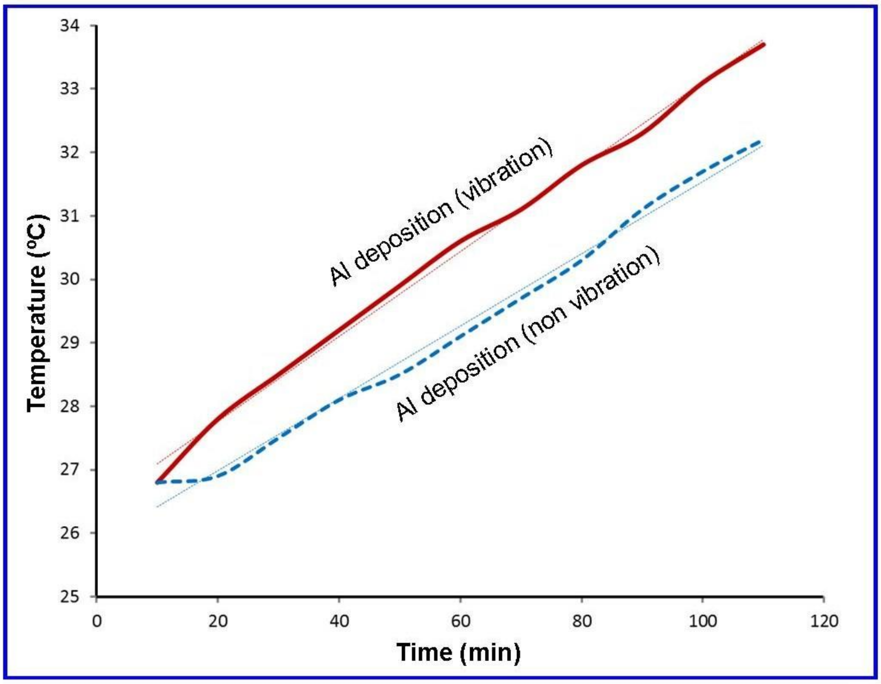

Figure 4 Temperature rise in Al coated substrates (vibrated at 7.6 kHz and non-vibrated during Al deposition) subjected to irradiation from a 100 W incandescent light bulb.

3.4 Laser Light reflections from substrate surface

A narrow band of laser light was also used to examine the reflective response of both types of substrate surfaces. The results of two representative substrate types used in the laser response measurements carried out in this study are presented in Figure 5. Inspection of Figure 5 reveals that both the vibrated substrate and non-vibrated substrate have the same reflective intensity, with both peaks located at 648 nm. However, there is a significant difference between the two peak widths. The peak width for the vibrated substrate is on average 39% broader compared to the peak width for the non-vibrated substrate. The result suggests that the Al deposited during substrate vibration has an enhanced reflective over the 620 nm to 680 nm wavelength range, with the most significant response occurring between 640 nm and 660 nm range. This result also confirms the significant difference in reflective intensity seen in the 100 W incandescent light bulb measurements between 620 nm and 680 nm range discussed earlier.

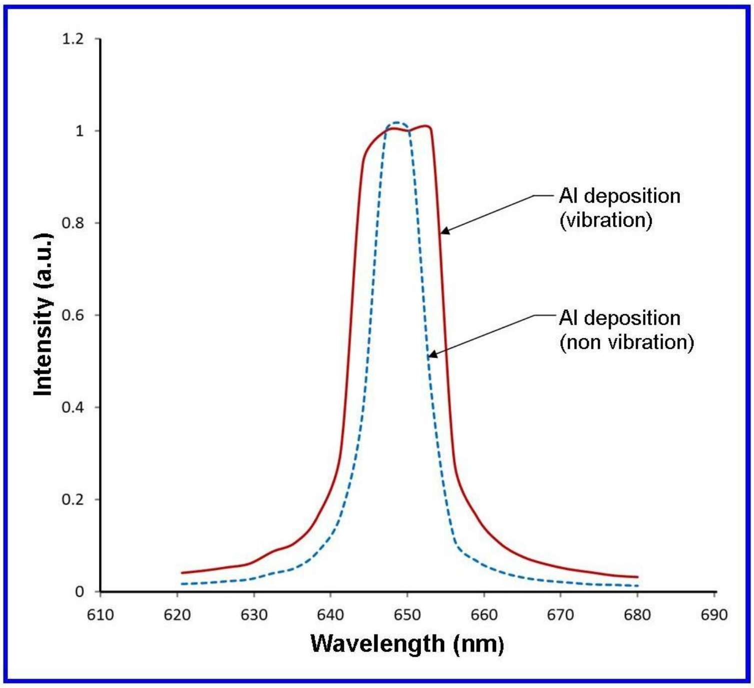

Figure 5 Surface reflectivity of laser light (630-680 nm, 1 mW) of typical vibrated and non-vibrated substrates coated with a thin film of aluminium.

4. **Conclusions**

The results of the present study have shown that during the vacuum deposition of Al on Cu substrates subjected to a vibration of 7.6 kHz there were fewer particles deposited. The SEM studies also revealed that the particles were spherical in shape and tended to be uniformly distributed over the entire substrate surface. Similar SEM studies on non-vibrated substrates revealed that the surface morphology was significantly different with much more densely packed granular shape particles. This result indicates that *in situ* vibration of a substrate can be used to influence surface morphology. Reflectivity and thermal response studies also confirm that changes in surface morphology can significantly influences surface reflectivity properties of the substrates. For example, reflectivity experiments revealed that vibrated substrates were superior to the non-vibrated substrates by 28 %, while the difference in the thermal response was around 14 %. These preliminary studies clearly indicate that substrate vibration can have a significant effect in controlling the overall surface properties of thin aluminium films deposited on copper substrates. However, further studies are required to fully investigate the potential use of substrate vibration as a controlling experimental parameter in determining surface properties.

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Disclosure

The authors report no conflict of interest in this work.

References

[1] Knox B, Ladiges P, Evans B, Biology, McGraw-Hill: Australian, 1st edition, 1994.

[2] Bunshah RF, Handbook of Deposition Technologies for Films and Coatings, NJ, USA, 2nd edition, 1994.

[3] Cheng LF, Liao LS, Lai WY, Sun XH, Wong NB, Lee CS, Lee ST, Effect of deposition rate on the morphology, chemistry and electroluminescence of tris-(8-hydroxyqiunoline) aluminium films. Chem. Phys. Lett, 2000; 319: 418-422.

[4] Koini M, Haber T, Werzer O, Berkebile S, Koller G, Oehzelt M, Ramsey MG, Resel R, Epitaxial order of pentacene on

Cu(110)-(2x1)O: One dimensional alignment induced by surface corrugation. Thin Solid Films, 2008; 517: 483-487.

[5] Mohanchandra KP and Uchil J, Electrical properties of CdS and CdSe films deposited on vibrating substrates, J. Appl. Phys., 1998; 84(1): 306-310.

[6] Karim SI, Chowdhury MA, Helali MM, Influence of Sound Vibration on Diamond-Like Carbon Deposition Rate. ISRN - Mechanical Engineering, Volume 2012, Article ID 676751, 8 pages.

[7] Regel LL and Wilcox WR, Diamond film deposition by chemical vapour transport, Acta Astronautica, 2001; 48(2-3): 129–144.

[8] Burakowski T and Wierzchon T, Surface Engineering of Metal, CRC Press, New York, NY, USA, 2000.

[9] Fu Y, Sun CQ, Du H, and Yan B, From diamond to crystalline silicon carbonitride: Effect of introduction of nitrogen in CH4/H2 gas mixture using MW-PECVD, Surface and Coatings Technology, 2002; 160(2-3): 165–172.

[10] Mohanchandra KP, Shanbhogue HG, and Uchil J, Effect of substrate vibration during evaporation on the optical properties of cadmium chalcogenide films. J. Phys. D, 1992; 25: 853-856.

[11] Paredes YA, Caldas PG, Prioli R, Cremona M, Quality improvement of organic thin films deposited on vibrating substrates. Thin Solid Films, 2011; 520: 1416-1421.

[12] Abramov OV, Action of high intensity ultrasound on solidifying metals. Ultrasonic, 1987; 25: 73-82.

[13] Djordjevic S, The effect of ultrasonic solidification of aluminium, Journal of Mining and Metallurgy, Section B: Metallurgy, 2003; 39(3-4): 527-532.

[14] Alasser RS, Badr HM, Mavromatis HA, Heat convectionfrom a sphere placed in an oscillating free stream. Int. J. Heat Mass Trans. 1999; 42: 1289-1304.

[15] Mackley MR & Stonestreet P, Heat transfer and associated energy dissipation for oscillatory flow in baffled tubes. Chem. Eng. Sci., 1995; 50: 2211-2224.

[16] Copper WL, Yang KT, Nee VW, Fluid mechanics of oscillatory and modulated flows and associated applications in heat and mass transfer – a review. J. Energy Heat Mass Transfer, 1993; 15: 1-19.

[17] Richardson PD, Effect of sound and vibration on heat transfer. Appl. Mech. Rev. 1967; 20: 201-217.

[18] Ohring M. The materials science of thin films. Academic Press, California, USA, 1991.

[19] Virkar AA, Mannsfeld S, Bao Z, Stingellin N, Organic semiconductor growth and morphology considerations for organic thin-film transistors. Adv. Mater, 2010; 22(34): 3857-3875.

[20] Krause B,Durr AC, Schreiber F, Dosch H, Seeck OH, Thermal stability and partial dewetting of crystalline organic thin films: 3, 4, 9, 10-perylenetetracarboxylic dianhydride on Ag(111). J. Chem. Phys. 2003; 119(6): 3429-3435.

[21] Manjunatha P, Uchil J, Rao MK, Thermoelectric properties of CdS & CdSe films deposited on vibrating substrates. Thin Solid Films, 1997; 305: 124-129.

[22] Viji VR, Kumar KS, Stella MM, Vaidyan VK, Studies on the properties of dip-coated indium tin oxide films considering substrate vibration. Indian J. Pure Appl. Phys. 2005; 43: 368-371.

[23] Uchil J, Shanbhogue HG, Thutupalli GKM, Optical properties and durability of thin films deposited on vibrating substrates at ultrasonic frequencies. Thin Solid Films, 1989; 170(1): 27-34.

[24] Barrett CS, Cohen JB, Faber J, Jenkins JR, Leyden DE, Russ JC, Predecki PK, Advances in X-ray analysis, Vol. 29, New York: Plenum Press, 1986.