On-line Characterisation of Copper Vapour Evolution from Linear Vapour Source Generated Using Strip Electron Beam

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Abstract. In electron beam assisted physical vapour deposition (EB-PVD) technique, the online characterization of the evaporator is essential for process optimisation and control. In applications such as decorative and corrosion resistance coating, the knowledge of time average distribution of vapour is essential, whereas in some real time applications such as isotope purification, surface hardening and alloying etc., real time knowledge of vapour distribution and vapour propagation is important. The online characterization of various parameters related to the evaporator and associated processes using least expensive techniques is necessary to know the process throughput. Measurement of atom flux using quartz crystal thickness monitor can be one such techniques. The experimental studies were carried out to characterize the evaporator using thickness monitor by measuring copper vapour propagation and distribution over the two dimensional source. The experimental data measured at two heights corresponding to aspect ratio 2 and 3 are presented and the behaviour of expanding vapour is discussed. This technique can also be used to estimate the source temperature from the deposition rate data, which is discussed in the paper with its validation using measured temperature using two-colour pyrometer.

1. Introduction
The electron beam assisted evaporation technique is well established in the coating industry due to its ability to attain high evaporation rate, temperature beyond the melting point of many refractory and reactive metals without containment problems and its ability to be used in a continuous mode of operation [1-3]. It is used for non-corrosive and decorative protective layer coating, surface alloying etc. During coating, real time variations of evaporation parameters get averaged out. Thickness monitor provides coating rate under dynamic condition.
In large-scale applications, the generation of wedge shaped atomic beam from a two-dimensional source like linear or strip type electron beam has already been reported [4-7]. Unlike point source, atomic beam generated from a 2D source does not run out of collisions and never attains free molecular flow regime [7]. The atomic collision processes along the length and the width directions are different and vapour flux distribution can be explained in terms of most generalized function having different beaming coefficients along the respective directions [8-9]. This type of evaporation has application in isotope purification process. In this process, knowledge of the real time evaporation parameters, vapour propagation and atomic relaxation is important. The online characterization using quartz crystal thickness monitor provides some information regarding these features. Measurement of thermal ion content using Langmuir probe is used to investigate the charge particle content and distribution. It is presented in another paper by our group in this conference.
Localized heat source to be used for evaporation requires extensive characterization of vapour distribution that is governed by the flow characteristics, which in turn are decided by a number of source parameters. During electron beam heating, the temperature rises from water-cooled crucible boundary to the vaporizing hot zone under the beam. Under this situation, the entire liquid pool is set into bulk convective current as a result of interplay of various forces such as temperature gradient-driven surface tension, vapour-thrust and buoyancy. Under these combined forces, the molten liquid has circulation up under the hot zone and down to low temperature outer boundaries with detached vortices under the hot zone. For this reason the source parameters changes due to time variation in the eddy structure, so that the properties such as surface temperature do not come to steady state. Thus the entire evaporating surface gets deformed and with extreme conditions of high evaporation rates, a trench is formed under the impinging electron beam [6, 7]. As a result of this, the evaporating source conditions such as dimensions and temperature are time variant and need due consideration in characterization of atomic vapour propagation in real time applications. Apart from these molten pool instabilities, generated directly heated segmented hot tantalum cathode filament can have non-uniform filament temperature that results in non-uniform current density and temperature of the evaporating source.

The data of atomic vapour evolution as a function of aspect ratio (Height/ Length of the source) and Knudsen number (decreases as source temperature increases) for any element can be used as an online characterization technique for evaporator’s performance. The ion content evolution and propagation measured using Langmuir probe and is reported in another paper in this conference. For neutral atom propagation, quartz crystal monitor is installed on a Wilson seal provided with vertical motion and rotary motion. The vertical motion is for changing aspect ratio and to know variation in vapour flux in propagation direction. The rotary motion is for spatial variation on one plane to know vapour flux distribution as a function of ‘θ’ and ‘ϕ’. Using these devices several experiments of atomic vapour flux measurements were carried out over a wide range of aspect ratio and Knudsen number. Various evaporation parameters such as beaming coefficients, source temperature were estimated using experimentally measured parameters such as deposition rate at various locations, source temperatures etc. for the validation of the technique. This technique can be applied for characterization of the vapour evolution of any element in any EB-PVD evaporator.

2. Experimental Measurement

Design details of 100kW evaporator developed in our laboratory have been reported along with its performance [4]. Briefly, this facility is a double-walled water-cooled stainless steel vacuum (operating pressure = 6x10^-5 mbar) chamber along with a pair of magnetic coils used in Helmholtz configuration and other components. The uniform axial magnetic field deflects the strip electron beam through 270° and also focuses electron trajectories in the post anode region. A variety of strip type electron gun with Ta filament has been developed for this evaporator. A fixed length gun of 80 mm length as well as another gun, in which the filament length can be varied from 130 mm to 160 mm are consistently used. The width and the thickness of Ta filament are 3.7 mm and 0.7 mm respectively. The evaporator is divided into two compartments using a separating plate. In order to avoid metal vapour entering into anode-cathode space of the strip electron guns and reduce filament life they are installed in the lower compartment. The water-cooled copper crucible along with the target and other diagnostic accessories for study are located in the upper compartment. In this study the electron gun was operated up to 90kW (48kV x 1.9A) and the filament length was 130 mm.

The vapour evaporating from the melt pool under the electron beam focal strip expands into the volume above the source. To measure the vapour flux at various locations, a quartz crystal thickness monitor is used. It is placed on a vacuum feed through which can be moved along vertical direction up to 300 mm and can also be given 360° rotational motion about the vertical mounting axis. The quartz crystal thickness monitor (SIGMA SQM-160; accuracy 0.1 A/sec) used has a water-cooled crystal mounting head and a pneumatically operated shutter to cover crystal surface during idle time. The tubing for water and pressurised air is made out of 6mm copper tubes which can be carefully given
shape to mount the thickness monitor head at desired location. The axis of rotation is 200 mm offset from the crucible centre along the axis of chamber and the quartz crystal head is mounted vertically above the centre of crucible. Thus the rotational movement of the vacuum feed-through moves the crystal head in circular path. The rotation of the feed through can be made with an accuracy of 1°. The shutter is opened for short time (~5secs) for each measurement to avoid saturation of the crystal. The schematic diagram of the experimental set up is shown in figure-1

The wedged shaped copper atomic beam was generated using 130mm-strip electron gun operated at power range of 40 to 90kW. The deposition rate was monitored in a horizontal plane at a vertical height of 240mm (aspect ratio 2) and 290mm (aspect ratio 3) measured from the evaporating source. At each of these aspect ratios, the thickness monitoring crystal head was rotated in the range of 0 to 90° with the resolution of 10° for each incident powers varying from 40 to 90 kW. The size of electron beam on the copper target generally has nearly 1:1 correspondence with the size of the filament. The strip beam dimension is measured using the photograph of the melt pool taken along with internal standard through a periscope arrangement. The photograph is analysed using image-processing software (ImagePro) to get the evaporating source dimension. Proper neutral density filter were used while taking photograph so as to avoid saturation of any pixel of the photograph. The length of the beam is measured from equal intensity contour. As there is a variation of width along the strip length, the average width is obtained by taking ratio of area enclosed by the equal intensity contour and the length. The dimension of the focal strip is measured to be 120 mm x 6 mm.

At the point of impingement of electron beam, the molten liquid has detached vortices under the hot zone due to which evaporating surface temperature does not attain a steady state value. Also, due to the convection current there is a variation in thermal contact between copper ingot and water-cooled crucible. This results in real time variation in temperature of the 2D evaporating source. The temperature of the 2D evaporating source is measured as a function of incident electron beam power using a two-colour pyrometer (Keller Make) through the periscope arrangement. The minimum and maximum temperature at any particular electron beam power is thus recorded to find the range of temperature variation.

### 3. THEORY

The experimental characterization of a wedge shaped atomic beam from a 2D evaporating source generated using a strip electron gun and its validation has been rarely reported in the literature. Holland has provided analytical expressions for the spatial distribution of the deposition thickness for various source geometries assuming Knudsen's cosine law for vapour flux distribution [10] applicable to free molecular flow. Rosengard [11] has also attempted to characterize the atomic flow from a 2D
source in terms of $K_n$ and a deviation from Knudsen cosine law using the beaming exponent $q_n$, expressed in terms of $K_n$. The need to express a beaming exponent in terms of $K_n$ and $\theta$ has already been pointed out to account for the flux propagating at larger angles [12]. The validation of these results are generally carried out by applying the Direct Simulation Monte Carlo (DSMC) technique with source conditions defined in terms of the Knudsen number ($K_n$) [13]. The spatial distribution of atomic flux from a two-dimensional source having width ‘D’ and length ‘L’ strongly depends on the radial distance of measurement ‘r’. Specifically it depends on the relative values of these parameters. In our earlier studies [14], we had reported that the Knudsen’s cosine law was inadequate to validate experimental results and showed that only over a limited free space vapour flux could be explained using the vapour distribution function $((n+1)/2\pi)\cos^n\psi$ reported by Chaleix et.al.[15]. Subsequently we have reported [8] more generalized spatial distribution function for vapour propagating from a 2D source with different beaming coefficients along the width and the length of the evaporating source and validated it with experimental results covering larger free space. This generalized vapour distribution function is defined as

$$f = \frac{(n_1 + n_2 + 2)}{4\pi} \cos^{n_1}\theta \cos^{n_2}\phi$$

Where $n_1$ is beaming exponent in the ‘$\theta$’ direction along the width and the beaming exponent $n_2$ is along the ‘$\phi$’ direction along the length of a 2D evaporating source. The evaporation of metals from a two dimensional strip source (length $(L) \gg$ width $(D)$) can be thought of as an ensemble of closely packed spot sources of width $D$ and length $dL$. To get the total vapor flux passing through any location above the strip source, the flux emanating from each elemental source area is to be integrated for the entire 2D source.

The location of the quartz crystal thickness monitor where the atom deposition rate is intended to be measured can be specified by $(r,\theta,\phi)$ (Figure-2), where ‘$r$’ is the source to collector distance, ‘$\theta$’ is the angle between normal plane passing through strip source and plane containing the collector and ‘$\phi$’ is the angle between the line joining an elemental source and a collector area with respect to the normal to the elemental source area in any given ‘$\theta$’ plane.

The total vapour emanated from the source depends on the source temperature $(T)$ and Knudsen number $(K_n)$. A fraction of vapour is reflected back into the source, which depends on Knudsen number [11]. The vapor further gets distributed over the space according to generalized distribution function reported earlier [8,9].

With this general vapor flux distribution function, the vapour flux (atoms/sec) on the horizontal collector of area ‘$a_s$’ in SC direction (having length SC) from a elemental source area, ‘$a_s$’ is given by

$$F = \frac{Q}{DL} a_s \frac{(n_1 + n_2 + 2)}{4\pi} \cos^{n_1}\theta \cos^{n_2}\phi \frac{\cos \theta \cos \phi}{SC^2}$$
Where $n_1$ and $n_2$ are the beaming coefficients along ‘$\theta$’ and ‘$\phi$’ directions respectively. $Q$ is the total evaporation rate (atoms/sec) from the source of dimension ($D \times L$). The beaming coefficients depend on the Knudsen number. The deposition rate at the location $(r, \theta, \phi)$ will be

$$F = \frac{M}{N_A \rho}$$

Where $N_A$ is Avogadro’s number, $\rho$ is density and $M$ is atomic mass in amu.

Depending on the source conditions, defined by the Knudsen number, and point of measurement, defined in terms of aspect ratio, the beaming exponents $n_1$ and $n_2$ are expected to show different behaviour. When $n_1 = n_2 = n$ and $\cos \theta \cos \phi = \cos \psi$ the distribution function becomes $((n+1)/2\pi)\cos^n \psi$ as reported by Chaleix et.al.[15] and when $n_1 = n_2 = n= 1$ this function becomes $(1/\pi)\cos \psi$, which is the Knudsen cosine law.

4. Results and Discussion

The atomic vapor deposition rate in $A^0/sec$ as measured by thickness monitor for various incident electron beam power and aspect ratio 2 and 3 for each position of thickness monitor rate is measured as a function of incident power. It varies exponentially with incident power as per the following relation $d=a \times P^b$, where $a$ and $b$ are constants and $P$ is incident e-beam power in kW. The typical plot of deposition rate as a function of incident power at various locations, for aspect ratio 2, is given in figure-3.

The deposition rate data is analyzed using various combination of source temperature $(T)$, beaming exponent $n_1$ and $n_2$. The combination that fits with the experimental data within specified accuracy is taken as a possible set. The best 500 such sets are used to calculate these parameters for spatial distribution of vapour flux. The beaming coefficients $n_1$ and $n_2$ are varied from 1 to 3 and 1 to 7 respectively in steps of 0.02. The temperature is varied in steps of 2 K within 50K around the expected temperature value. The results are presented in table- 1 & 2 for aspect ratio 2 and 3 respectively. The calculated temperature from the deposition rate data is in good agreement with the instantaneously measured temperature of the evaporating source using two-colour pyrometer. It should be noted that for the reasons mentioned earlier, the temperature of the evaporating source never reaches steady state value [7]. The measurements using thickness monitor are carried out for 5seconds only, whereas during our earlier studies we have presented experimental results wherein vapour flux distribution was measured by carrying deposition for large time duration so that variation in source temperature gets averaged.
Table-1: Height from source 24 cm (aspect ratio: 2); Source dimension (120mm x 5mm); Angle covered 38° perpendicular to length. (Rotation of crystal head 0° to 90° in steps of 10°)

| E-Beam Power | Temp (K)   | n_1 (±) | n_2 (±) | Error in fitting (±) | K_e |
|---------------|------------|---------|---------|----------------------|-----|
| 40 kW         | 1771 (±2.9)| 1.15 (±0.1)| 1.31 (±0.21)| 4.67 (±0.38) | 0.44 |
| 48 kW         | 1798 (±3.1)| 1.09 (±0.07)| 2.00 (±0.25)| 4.13 (±0.49) | 0.32 |
| 60 kW         | 1843 (±2.9)| 1.16 (±0.11)| 2.83 (±0.24)| 3.99 (±0.3) | 0.2 |
| 72 kW         | 1881 (±3.5)| 1.45 (±0.16)| 3.58 (±0.32)| 4.81 (±0.27) | 0.134 |
| 80 kW         | 1906 (±2.9)| 1.61 (±0.16)| 4.04 (±0.25)| 3.98 (±0.25) | 0.103 |

Table-2: Height from source 39 cm (aspect ratio: 3); Source dimension (120mm x 6mm); Angle covered 38° perpendicular to length. (Rotation of crystal head 0° to 90° in steps of 15°)

| E-Beam Power | Temp (K)   | n_1 (±) | n_2 (±) | err (±) | K_e |
|---------------|------------|---------|---------|---------|-----|
| 40 kW         | 1776 (±3.0)| 1.15 (±0.11)| 1.3 (±0.2)| 4.56 (±0.32) | 0.41 |
| 48 kW         | 1809 (±3.4)| 1.21 (±0.16)| 3.43 (±0.32)| 3.62 (±0.18) | 0.28 |
| 60 kW         | 1866 (±1.9)| 1.18 (±0.14)| 4.39 (±0.26)| 2.74 (±0.29) | 0.16 |
| 72 kW         | 1899 (±2.8)| 1.12 (±0.1)| 5.1 (±0.31)| 3.78 (±0.25)| 0.11 |
| 80 kW         | 1930 (±3.2)| 1.19 (±0.12)| 6.4 (±0.35)| 4.32 (±0.14)| 0.08 |

5. Conclusions

Based on our earlier studies [8], it was concluded that for aspect ratio ≈1, the collimation of atomic beam and beaming exponent along the width remain constant whereas those along length of the 2D source increases monotonically for different source temperatures as a function of 1/K_e ranging from 0.85 to 4.35.

It was anticipated that at higher aspect ratios evaporating source should appear as a point source and vapour propagation parameters should cease at certain values determined by the source Knudsen number, as is the case for point source. To study the difference in the vapour propagation from a point and the linear source, the spatial distribution of copper vapour from a 2D source at aspect ratio ≥ 3 was studied and reported. It was observed that the vapour collimation along the width of the evaporating source ceases with beaming coefficient in the range of 2 whereas collimation along the length direction continues and the beaming coefficient along the length increases beyond 6 for inverse Knudsen number in the range of 0.75 to 6.67[9].

Present study was carried out to demonstrate that the quartz crystal monitor could be used as an online technique for characterization of the EB-PVD evaporator. The beaming exponents along width and the
length direction of the evaporating source and evaporating source temperature are estimated from the vapour flux distribution measurements. The beaming exponents are plotted as a function of inverse Knudsen number for aspect ratio 2 and 3 as shown in figure-4. The plots presented in figure-2 as a function of incident e-beam power can be used as database for online characterization of the evaporator. Experimental results obtained in this studies using online measurement vapour flux distribution at aspect ratio 3 are consistent with our earlier reported beaming coefficients along the length and the width of the evaporating source and its temperatures as function of inverse Knudsen number within experimental error. This clearly indicates reliability of this technique. Subsequently we have conducted vapour flux distribution studies from 2D source at aspect ratio ≈2 with increasing electron beam power from 40 kW to 80 kW; wherein the source temperature increases from 1776K to 1930K and $1/\text{Kn}$ increases from 2.4 to 12.5. For these variations in source condition, the beaming exponent along the width of the 2D source $(n_1)$ practically remains constant at ~1.2. This clearly indicates that, within the standard deviation, the beaming exponent along the width of the 2D source $(n_1)$ remains constant depending on vapour flow conditions in the vicinity of the 2D source and the range of $1/\text{Kn}$. The case is same for beaming exponent $n_1$ at aspect ratio>3. At the same time, the beaming exponent along the length direction of the 2D source $(n_2)$ rises substantially from 1.3 to 6.41 with increase in $1/\text{Kn}$ at aspect ratio >3. These values of $n_2$ are substantially higher than those for corresponding range $1/\text{Kn}$ at aspect ratio ≈ 2. This clearly indicates that lengthwise collisions, atomic beam collimation and hence beaming coefficient do not stabilize and the wedge-shaped atomic beam generated from a 2D source does not run out of collisions in the lengthwise direction even at an aspect ratio >3. In addition the atomic beam generated from the 2D source does not attain the free molecular regime even at radial distance of 390mm and above the source.

It is interesting to note that, during these EBPVD experiments with the strip electron gun and a 2D source over a wide range of Knudsen number, the vapour flux in the free space covering ±L/2 and ±θ has uniformity better than 90%. Thus, by increasing length of the 2D evaporating source, it is possible to handle large size substrates for EBPVD.

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7. Reference

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