Studies for determining thermal ion extraction potential for aluminium plasma generated by electron beam evaporator

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Abstract: For effective evaporation of refractory metal, electron beam is found to be most suitable vapour generator source. Using electron beam, high throughput laser based purification processes are carried out. But due to highly concentrated electron beam, the vapour gets ionised and these ions lead to dilution of the pure product of laser based separation process. To estimate the concentration of these ions and extraction potential requirement to remove these ions from vapour stream, experiments have been conducted using aluminium as evaporant. The aluminium ingots were placed in water cooled copper crucible. Inserts were used to hold the evaporant, in order to attain higher number density in the vapour processing zone and also for confining the liquid metal. Parametric studies with beam power, number density and extraction potential were conducted. In this paper we discuss the trend of the generation of thermal ions and electrostatic field requirement for extraction.

1. Introduction

For achieving better efficiencies in the laser based process under consideration, the desired atomic vapour density is of the order of $10^{12}$-$10^{13}$ atoms/cc at the photo-interaction zone. In this process, the desired material is evaporated using electron beam, laser beam or resistive heating. The process is carried out in vacuum environment. Once the vapour stream reaches the photo interaction zone, the laser photons interact with the vapour stream. Using suitable laser frequency, only the desired species is selectively excited and then ionized. The selectively ionized species is extracted using suitable extraction potential. The extraction zone is so designed that the ions are collected with minimum loss, to ensure the process efficiency. Electron beam heating is found to be more efficient for generating high vapour flux with greater efficiency [1]. In the electron beam heated source ions are generated through various ionization processes, namely, electron impact ionization, Saha ionization etc. Since these ion species are non-selective in nature, their presence in photointeraction region, and subsequent collection with selective laser photoions reduces the process selectivity. We are in the process of characterizing these ions and their effects on the total laser separation/purification process. This study will lead to designing an effective ion collection scheme for better efficiency of the process.

In our previous work [2] we have conducted experiments while placing aluminium ingot in a water cooled copper crucible and could achieve vapour density of the order of $10^{11}$ atoms /cm$^2$ near the photo interaction zone (~200-300mm above the evaporation surface), using the available electron beam source of 10kW (10 kV, 1A). The location of the photointeraction zone was decided as per the process requirement. The minimum distance of the photointeraction zone from the source is based on the magnetic field extent of the electron beam system and the vapour parameters. To achieve the desired vapour density, one has to either use a higher electron beam power or utilise the available power more effectively. As the electron beam source is limited to 10kW, the effective use of energy was achieved using alumina coated stainless steel insert.
2. Experimental Setup

The experimental setup is as shown in the Figure 1. The aluminium ingot is kept in an alumina coated stainless steel insert. This insert is placed in a water cooled copper crucible (40cc) and the aluminium vapour is produced by 270° bend electron beam, inside a vacuum chamber. During the experiment the vacuum of the order of $2 \times 10^{-5}$ mbar was maintained. The vapour emanates in forward direction and expands as it moves upward. A pair of stainless steel plates [300mmx 50mmx 2mm (thick)] are kept in parallel configuration (~30mm separation) along the vapour flow direction to extract the ions using electrostatic field. The height could be varied between 200 and 300mm from the source surface. Below these plates, two horizontal plates are kept with a separation of ~10mm, to collimate the directional vapour stream leading to ion extraction zone. To estimate the ion density, an in-house made Langmuir probe was used. The Langmuir probe is made of Ta wire of dia 0.5mm. The exposure length of the probe was ~6mm. The remaining parts of the probe and signal wire were suitably protected from being coated with the vapour. Arrangement is made to keep the isolation between signal and ground wire to get the correct signal for ion current [2, 3]. For every experiment, two probes were used, one below the beam collimator plates and one above the ion extraction plates to measure the ion densities at these locations. Typical Langmuir probe characteristic curve is shown in Figure 2. The characteristic curve was used for determining electron temperature and density. No shutter for the probes has been provided since repeatability of the readings with multiple exposures was verified and found within experimental error limits for the experimental duration of ~10min. A stainless steel token is also kept at the location of the top probe, to determine the atom density at various source temperatures. A horizontal plate was kept above the extraction plates to prevent the vapour from migrating to other areas of the vacuum chamber. The source temperature is measured using a two color pyrometer. During feeler experiments for finalizing the operating parameters, it was observed that for a given source condition, the ion current is the best representative of the identical source condition. For ensuring similar experimental conditions during the set of parametric experiments, where the vapor behavior was studied for different geometrical variations, a reference Langmuir probe was used at fixed location with a fixed bias. The probe current is maintained by maneuvering the electron beam parameters, to ensure identical vapour properties.

The ion density is determined using standard Langmuir probe characteristic curve as follows. The probe current measured with the probe is related to the electron density [4-7] as,

$$I_e = n_e e A \left( \frac{kT_e}{2\pi m_e} \right)^{\frac{3}{2}}$$  \hspace{1cm} (1)

where $T_e$ is the electron temperature, $I_e$ is the saturation electron current, $A$ the probe exposure area, $n_e$ is the electron density, $e$ the electronic charge, $m_e$ the mass of the electron and $k$ is the Boltzmann constant. Since quasi-neutral condition was assumed, the ion density was assumed to be same as the electrons.

The vapour density was calculated from the surface temperature (obtained using the pyrometer) and the deposition rate on the token.

Average velocity, $c^* = \sqrt{\frac{8kT}{\pi m}}$

Vapour Density, $n_a = \frac{\rho}{C' m}$  \hspace{1cm} (2)

where $k$ is the Boltzmann constant, $T$- the source temperature, $m$- the atomic mass of the evaporating element, aluminium. The flux, $\rho$, is determined by the weight of the material deposited per unit area of the token surface during experiment. The ions generated due to Saha ionization can be evaluated from
\[ \frac{\alpha^2}{1 - \alpha^2} = 5 \times 10^{-4} \times \frac{5}{T} \times e^{\frac{-e\epsilon}{kT}} \times \frac{1}{P} \]  

(3)

Where \( \alpha \)-degree of ionization due to thermal process, \( T \)-Temperature in K, \( \epsilon \)-the ionization potential of the element, \( k \)-Boltzmann constant, \( P \)-the saturated vapour pressure in atm. The saturated vapour pressure (mmHg) for temperature (K) was evaluated using Eq.4 [8] as follows,

\[ \log P = -\frac{16450}{T} + 12.36 \]  

(4)

Figure.1: The Experimental setup for aluminium evaporation experiments

Figure.2: Typical Langmuir probe characteristic curve.
3. Discussion

3.1 Characterisation of plasma

During the evaporation of aluminium, ionization of the vapour takes place due to various mechanisms, such as, electron impact ionisation, Saha ionization, etc. To determine the contribution of these non-selective ions with enhanced vapour density, the vapour and plasma density distribution along the centre line height at various distances (230mm to 350mm) from the source were determined [Figure.3(a)]. These experiments were carried out with 2 kW incident electron beam power. Both the atom density and the ion density follow the similar trend and hence the percentage of ionization (~0.6%) does not vary much along the height within the experimental range.

The behavior of electron temperature within the experimental variations of electron beam power for different heights from the source are presented in Figure.3 (b). The electron temperatures along the height from the source are almost constant for the range of measurement, which matches with the results presented by Nishio et. al. [8]. The variation of electron temperature (0.36 eV – 0.41 eV) for different electron beam power is also not much for the experimental range (1.5 kW to 3 kW), as expected. Some variations in electron temperature (0.11eV to 0.18eV) could be observed by Nishio et al. [9] for drastic variation of electron beam power (172 kW to 300 kW).

![Figure.3(a): Variation of aluminium vapour density and thermal ion density at different heights from the source](image1)

![Figure.3(b): Variation of electron temperature for different electron beam power along the height from the source](image2)

3.2 Prominence of electron impact ionization:

To determine the effectiveness of various processes for causing ionization in the vapour, the contribution of Saha ionization to non selective ions was evaluated using Eq.3. In one of the reported result [10] it was presented that the Saha ionization is the main source of ion generation and the rest is linked to electron impact ionization. Since the Saha Ionisation is predominantly governed by the temperature of the source, it was perhaps dominating in those experimental conditions. In our experiments with aluminium, the maximum source temperature was ~1473K, which gives vapour pressure of the order of 2.8x10^5 atm. Aluminium has ionization potential of 5.986 eV. The percentage of Saha ionization is of the order of 10^-9. This is considered negligible compared to the total ionization determined in these experiments. This indicates that the major source of vapour ionization is the
electron impact ionization due to electrons from both the electron beam source and from the backscattered/secondary electrons. Probability of ionization also increases with higher vapour density and could be seen in the results reported.

3.3 Determination of ion extraction potential with different evaporating conditions

Experiments were conducted to determine the extraction potential for removing the non-selective ions using ion extraction plates. For this the probe which was kept above the extraction plates (LP1) was biased at its ion saturation current. The bias potential was determined from the probe characteristic curve. The evaporation experiment was started with pre-determined operating conditions as per experimental plan. Firstly, a stable ion current is observed in the Langmuir probe (LP1). Then the negatively biased extraction plate potential was increased slowly and the probe ion currents were noted. With increase in negative bias of extraction plate potential, reduction in probe current was observed. This was the indication of attraction of ions toward the extraction plate. At a specific extraction plate potential, the direction of probe current was reversed (the probe started measuring mainly electron current). This potential was considered as the extraction potential required for extracting the ions available in the ion extraction region for that experimental condition. To confirm the behavior, the top of the extraction zone was covered by a horizontal top collector plate, very close to the top of the ion extraction zone, so that escape of vapour was minimal. This plate collected all the vapour along with ions escaping the ion extraction zone. The behavior of the top collector current was also observed in the same manner as that of the Langmuir probe (LP1), by varying extraction plate potential. Similar behavior was observed in case of top collector current measurement. The typical behavior of Langmuir probe (LP1) and top collector current are presented in Figure 4.

\[\text{Figure 4: Variation in probe (LP1) and the top collector current along with the application of extraction plate potential.}\]

We could conduct experiments for vapour densities in the range $8.3 \times 10^{10}$ to $3.4 \times 10^{12}$ /cm$^3$. For higher number densities inserts lined with alumina were used. In Table-1, the first three data were for without inserts/liner evaporation experiments while the last three were for experiments conducted with liner/insert. The variation of electron temperature for different electron beam power is very small as expected, for the range of variations in experimental parameter. The degree of ionization
monotonously increases with beam current for source of one configuration (Figure 5), that is, with inserts or without inserts; as the electron impact ionization is higher at higher beam currents.

Table-1: Comparison of percentage ionization of aluminium vapour with and without Alumina coated insert (data were for height of 300mm from the source).

| Sl. No | Vapour Density (atoms/cm³) | Ion Density (Ions/cm³) | Beam Power (kW) | Degree of Ionisation, (%) | Electron Temp, T_e (eV) |
|--------|----------------------------|------------------------|-----------------|---------------------------|-------------------------|
| 1      | 8.3 x 10^{10}             | 6.2 x 10^{7}           | 6(10kV/600mA)   | 0.07                      | 0.36                    |
| 2      | 1.9 x 10^{11}             | 4.6 x 10^{8}           | 7(10kV/700mA)   | 0.24                      | 0.39                    |
| 3      | 4.6 x 10^{11}             | 4.7 x 10^{9}           | 8.5(10kV/850mA) | 1.0                       | 0.39                    |
| 4      | 5.2 x 10^{11}             | 2.9 x 10^{8}           | 1.5(10kV/150mA) | 0.06                      | 0.38                    |
| 5      | 2.3 x 10^{12}             | 2.3 x 10^{9}           | 2(10kV/200mA)   | 0.1                       | 0.41                    |
| 6      | 3.4 x 10^{12}             | 1.9 x 10^{10}          | 3(10kV/300mA)   | 0.6                       | 0.412                   |

Figure.5: Aluminium vapour density with and without alumina lined stainless insert and their respective thermal ion densities

Similarly a set of experimental data for electron temperature and extraction potential requirement as a function of ion densities are presented in Figure-6. The electron temperatures were nearly same for the range of ion density observed in this set of experiments, as expected. The ion extraction is a complicated process, which is a function of drift velocity (electron temperature related), sheath formation in plasma, ion density, etc. In these experiments, the behavior of ion extraction potential verses ion density does not lead to any definite conclusion regarding the contribution of
different parameters. Set of experiments are planned to study the effects of these parameters in near future.

Figure-6: Behavior of ion extraction potential and electron temperature for different ion density

4. Conclusion
Experiments were carried out to study the plasma behavior with enhanced vapour density using ingot of evaporant in insulated liner. The methodology was devised to determine the ion extraction potential for a selected process condition, which will help in designing the ion extraction set-up. Experiments are planned with different materials of interest, and also to derive correlations between ionization and process parameters, which will be useful for designing the ion extraction system of the laser based purification process.

5. Acknowledgement
Authors acknowledge the suggestions from Dr. N.K. Joshi for determining plasma parameters, which helped in designing the experiments. Authors are grateful to Dr. A.K. Das for his constant support in carrying out the study.
References

[1] S. Schiller, U. Heisig and S. Panzer, Electron Beam Technology (Wiley, N.Y, 1982)

[2] Dileep Kumar V et al, Langmuir probe diagnostic studies of charged particles in vapour generated by 270° bend electron beam evaporation, presented at SEBTA 2005, BARC Mumbai.

[3] E. Besuelle and J.-P. Nicolai, Study of expansion of plasma generated by electron-beam evaporation, J. Appl. Phys., Vol. 84(8), 1998

[4] F. F. Chen, Plasma Diagnostic Techniques, Ed, R. H. Huddleston and S. L. Leonard (Academic, N.Y. 1965)

[5] F. F. Chen, Use of Electrostatic Probes in Plasma Physics, PPL, Princeton Univ, Princeton, N.J (August 1961)

[6] F. F. Chen, Lecture Notes on Langmuir Probe Diagnostics, Electrical Engg Dept, Univ of California, L.A, Mini-course on Plasma Diagnostics, Jeju, Korea (2003)

[7] H. M. Mott-Smith and I. Langmuir, The Theory of Collectors in Gaseous Discharges, Physical Review, Vol. 28, 1926.

[8] Smithell’s Metals Reference Book, 7th edition, Ed, E. A. Brandes and G. B. Brook, Reed Educational and Professional Publishing Ltd., 1992

[9] Nishio et al, J. Appl. Phys. 72(10), 1992

[10] Dikshit et al. J of Phys. Conference Series 114 (2008)