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Source fabrication and lifetime for Li⁺ ion beams extracted from alumino-silicate sources

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A space-charge-limited beam with current densities \( J \) exceeding 1 mA/cm² have been measured from lithium alumino-silicate ion sources at a temperature of \( \sim 1275^\circ \) C. At higher extraction voltages, the source appears to become emission limited with \( J \geq 1.5 \) mA/cm², and \( J \) increases weakly with the applied voltage. A source with an alumino-silicate coating 6.35 mm in diameter and \( \leq 0.25 \) mm thick, has a lifetime of \( \sim 40 \) hours at \( \sim 1275^\circ \) C, when pulsed at 0.05 Hz and with pulse length of \( \sim 6 \) \( \mu \)s each. At this rate, the source lifetime was independent of the actual beam charge extracted due to the loss of neutral atoms at high temperature. Furthermore, the source lifetime increases with the amount of alumino-silicate coated on the emitting surface, and may also be extended if the temperature is reduced between pulses.

I. INTRODUCTION

Low-mass ions, such as lithium (Li⁺), have an energy loss peak \( (dE/dx) \) at a suitable kinetic energy [1] for heating targets to electron-volt temperatures for studies of warm dense matter (WDM) [2]. The Heavy Ion Fusion Sciences (HIFS) program [3] at Lawrence Berkeley National Laboratory will carry out WDM experiments using Li⁺ ion beam with energy 1.2 - 3 MeV to achieve volumetric heating up to 0.1 - 1 eV. Experiments will be carried out at the Neutralized Drift Compression Experiment-II (NDCX-II) facility [4, 5]. The conceptual design of NDCX-II was presented by A. Friedman et al. in Ref. [6] using a 10.9 cm diameter source of lithium ions (Li⁺).

A Li⁺ ion beam can be produced by thermionic emission from the alumino-silicates compounds \( \beta \)-spodumene and \( \beta \)-eucryptite [7–9] at above 1200° C. This type of ion source is also being used in magnetically confined fusion experiments for plasma diagnostics [10–14]. Table I shows Li⁺ current density data presented by several authors [7, 9, 11, 13, 15, 16]. Unfortunately, crucial details of the source fabrication and emission characterization are not always published. Krupnik et al. [15] demonstrated beam current density of 4 mA/cm² at 1400° C to 1500° C. However, this temperature range is near sintering (melting) of the lithium alumino-silicate compound and emission at this temperature is unstable due to the phase transitions of the compound. There is also concern about de-lamination of fragments from the substrate depending on the size of the ion source. In contrast to the potassium alumino-silicate sources, the lifetime for lithium sources is short when operating at a very high temperature, and therefore is an important issue for our application. This article describes preparation of lithium \( \beta \)-eucryptite compound, typical current density and lifetime, and variation of lifetime with temperature and mass of the alumino-silicate. These results reflect recent experimental work to characterize lithium alumino-silicate ion sources.

The total lithium ion charge produced by a source depends on the beam pulse length and extraction voltage, and on the ratio of the emitted alkali ions and neutral atoms. In general, for a pulsed ion source, the total beam charge per pulse is

\[
Q = J_b A \tau,
\]

where \( Q \) is the charge of the beam pulse, \( J_b \) is the beam current density (assumed uniform), \( A \) is the emission area, and \( \tau \) is the pulse duration. The total charge available \( (Q_a) \) by a source is

\[
Q_a = \left( \frac{M_s \eta}{m_i} \right) e,
\]

where \( M_s \) is the mass of lithium \( \beta \)-eucryptite compound (in gm) that is used to fabricate a source, \( \eta \) is the concentration of lithium atom by weight within the total compound, \( m_i \) is the mass of the ion, \( \eta \) is the concentration of lithium atom by weight within the total compound, \( m_i \) is the mass of the ion, \( (\text{for Li}^+, m_i=1.16 \times 10^{-23} \text{ gm}) \), and \( e \) is the ion charge. Therefore, the theoretical lifetime of a source for complete extraction (neglecting emission of neutrals) is related to the compound mass by

\[
T_{\text{life}} = \left( \frac{M_s \eta}{m_i} \right) \left( \frac{e}{I_b} \right),
\]

where \( I_b = J_b A \) is the beam current for the source surface area \( A \). For example, 0.4 mg lithium alumino-silicate compound that has a 5.56\% concentration of lithium atom can provide a beam current \( (I_b) \) of 25 \( \mu \)A with a lifetime of 3.4 hours.

In comparison with potassium alumino-silicate, the lithium alumino-silicate has much shorter lifetime and the coating is more difficult to make. In order to achieve a good coating, it is necessary to melt alumino-silicate into the porous tungsten substrate during the sintering (melting) process. In fact, the variation of the amount of absorbed material may affect the lifetime. Also, fabricating a source with a thick coating is problematic due to the nature of anisotropic thermal-expansion coefficients of the lithium alumino-silicate resulting in material stresses.
There are surface cracks if the material is too thick and the material occasionally flakes off from the substrate surface. Figure 1(a) and Fig. 1(b) show 500 times and 2000 times magnified tungsten substrate surfaces, respectively; Fig. 1(c) shows a cracked and partially flaking source surface, and Fig. 1(d) shows a smooth surface of a coated substrate. The magnified images were taken with a 15 kV electron microscope.

The earlier facility NDCX-I [17, 18] used a 2.54 cm diameter potassium-ion ($K^+$) source [19]. $K^+$ ion sources, with an ionization energy of 4.34 eV, deliver a space-charge limited current density $J \approx 20$ mA/cm$^2$ at a temperature of $\sim 1050^\circ C$. In comparison, the lithium ionization energy is 5.39 eV, and a temperature greater than $\sim 1200^\circ C$ is required to produce a space-charge limited current density of 1 mA/cm$^2$.

<table>
<thead>
<tr>
<th>Density (mA/cm$^2$)</th>
<th>Temp. ($^\circ C$)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 ($\beta$-eucryptite)</td>
<td>$\approx 1230$</td>
<td>[7]</td>
</tr>
<tr>
<td>0.02 ($\beta$-spodumene)</td>
<td>$\approx 1200$</td>
<td>[9]</td>
</tr>
<tr>
<td>1.5 ($\beta$-eucryptite)</td>
<td>$\approx 1200$</td>
<td>[11]</td>
</tr>
<tr>
<td>1 ($\beta$-eucryptite)</td>
<td>$\approx 1300$</td>
<td>[13]</td>
</tr>
<tr>
<td>4 ($\beta$-eucryptite)</td>
<td>$\approx 1500$</td>
<td>[15]</td>
</tr>
<tr>
<td>1.5 ($\beta$-eucryptite)</td>
<td>$\approx 1275$</td>
<td>[16]</td>
</tr>
</tbody>
</table>

II. PREPARATION OF LITHIUM $\beta$-EUCRYPTITE ALUMINO-SILICATE SOURCE

There are several steps to prepare a lithium alumino-silicate source: (1) produce the chemical compound, (2) grind the compound into powder, (3) apply a “green coating”, (4) sinter the material to form a hard surface layer.

A. Preparing powder, coating and drying process

A mixture of Li$_2$CO$_3$, Al$_2$O$_3$, and SiO$_2$ is prepared following the stoichiometric ratio for $\beta$-eucryptite type source [7], and calcined at 600$^\circ C$ for one hour to drive off CO$_2$. This temperature has been chosen after testing several samples at different temperatures (400 to 1200$^\circ C$) and analyzing under electron microscope to detect carbon. The resulting carbon-free calcined powder is ground to obtain a homogeneous mixture, and then heated to $1200^\circ C$ to form Li$_2$O-Al$_2$O$_3$-2SiO$_2$ leucite crystalline phase. This $\beta$-eucryptite crystalline compound is ground to particulate size in the range of 100 $\mu$m, ready for use in coating. For example, in making a 0.63 cm diameter Li$^+$ source, we use 0.037 gm of $\leq 37$ $\mu$m alumino silicate powder. After mixing the powder with several drops of de-ionized water, the mixture is agitated for several minutes in an ultrasonic cleaner to break up the clumps. A 70-80% density tungsten button is used as substrate after it has been clean fired at 1700$^\circ C$ for one hour in a vacuum furnace. Just before coating, the surface of the tungsten button is wetted with de-ionized water, and then a small amount of alumino-silicate suspended in plenty of de-ionized water is allowed to be self-absorbed into the porous tungsten substrate to “prime” the surface. The next step is to spread alumino-silicate paste on the substrate surface and build that up to the desired thickness. A Teflon spatula can be used to create a smooth finish. The coated tungsten button is placed into a vacuum oven for drying. The oven temperature is set to 100$^\circ C$ with rise rate of 2$^\circ C$/minute starting from room temperature. Drying takes about an hour.

B. Vacuum furnace melting procedure

The furnace is first backfilled with Argon and a small amount of Hydrogen (negative 677 millibar pressure of 96% Argon plus 4% Hydrogen). The idea of using a partial pressure of inert gas was to minimize lithium vapor loss. The following temperature cycle was used in sintering the alumino-silicate sources. At first the system is heated to $\sim 80^\circ C$ at a ramp rate of 2$^\circ C$/minute. After that, the temperature is increased at 5$^\circ C$/min until 200$^\circ C$, followed by 10$^\circ C$/min to 1400$^\circ C$. The peak temperature is held steady for 5 minutes, and then ramping down at 7$^\circ C$/min to room temperature. After cooling to room temperature, the furnace is slowly backfilled with argon to atmospheric pressure.

FIG. 1. (a) 70% to 80% dense an enlarged tungsten substrate surface shown 500 times magnified, (b) same surface shown 2000 times magnified, (c) source surface with cracks and flakes, and (d) an example of a smooth source surface.
III. BEAM CURRENT DENSITY OF A PULSED BEAM

Several 0.63 cm diameter lithium-alumino silicate ion sources were installed successively in an ion diode for testing. The distance between the source surface and the mid-plane of the extraction electrode was 1.48 cm. The extraction pulse voltage could be varied from 1 kV to 10 kV. A tungsten filament coil was used to heat up the source to desire temperatures. The beam current was measured by a Faraday cup. A negative biased electrode in the cup was used to suppress unwanted electrons from entering the positively biased collector. The positively biased collector also functions to suppress secondary electrons once the beam impinges on the collector plate. The ion diode configuration was described in an earlier article [16] with the diagnostic details and identification of the elements. Figure 2 shows the measured beam current density as a function of the extraction voltage ranging from 0.5 kV to 10 kV for source temperatures set from 1220 °C to 1300 °C. The source temperature was measured using a disappearing filament-type brightness pyrometer with null-balance, lamp-current measuring circuit [20], calibrated with emissivity of tungsten. The pyrometer is sensitive to the brightness at \( \lambda = 0.65 \) microns. The “brightness temperature” measured using the pyrometer is affected by the emissivity of the alumino-silicate material. We note that the emissivity of the alumino-silicate at \( \lambda = 0.65 \) microns may not be the same as tungsten.

The space-charge limited current density of an ion gun is defined by the Child-Langmuir law:

\[
J (\chi, V, d) = \frac{\lambda V^{3/2}}{d^{2/3}},
\]

where \( \chi = \frac{4 \pi e^2}{m_e}, \) and \( d \) is the effective distance between source and extraction electrode. Thus, in the space-charge limited regime, the beam current is determined by the ion mass and the geometry of the ion gun, and is linearly proportional to \( V^{3/2} \). Figure 2 shows that a space-charge limited current density of 1 mA/cm\(^2\) can be obtained when the sources were operated at or above 1275 °C with a 2.52 kV extraction bias. At higher extraction voltages, the source appears to become emission limited with \( J \geq 1.5 \) mA/cm\(^2\), and \( J \) varies weakly on the applied voltage.

IV. LIFETIME WITH SOURCE TEMPERATURE AND LITHIUM MASS DEPOSITED

Two of the approximately 0.25 mm thick sources were operated at a temperature of \( \approx 1265 ^\circ \)C to extract 5 to 6 \( \mu \)s pulsed beams with \( V=1.75 \) kV. The pulses were repeated at a rate of 0.05 Hz until emission of the sources was significantly reduced. Figure 3 shows lifetime of the sources when the beam was extracted at the space-charge limited regime. A uniform current density profile was observed for the duration of 30 to 40 hours after 10 to 15 hours of initial operation. It is speculated that the initial \( \approx 15 \) hours was affected by contamination while the heat and pulsed extraction voltage gradually removed the contamination. It was observed that the lifetime varied from source to source even when the operating temperature, pulsed voltage and pressure of the chamber were not significantly altered. One possibility is due to the variation in the mass of the alumino-silicate on the sources. Therefore a separate experiment was set up to measure the lifetime of the ion source as a function of temperature and mass of Li alumino-silicate used. In this set up, a narrow molybdenum foil with an embossed spherical depression at the center is used as the source substrate and simultaneously a heater ribbon. A small amount of alumino-silicate material is deposited at the depression pocket to form an emitting area.

A. Preparation of pocket source and experimental setup

Figure 4 shows the experiment setup including the molybdenum ribbon. All the ribbons used were 0.075 mm thick, 4.9 mm wide, and 5 cm long. The pocket was accurately made, always has the same diameter of 2.1 mm regardless of depth. There were five different pocket depths: 0.25 mm, 0.43 mm, 0.50 mm, 0.63 mm, and 0.75 mm. The ribbons were cleaned with de-ionized water in ultrasonic cleaner. After being air-dried, the pockets of the ribbons were lightly blasted with aluminum oxide powder (“sand blasted”) to create a rough surface for anchoring alumino-silicate material when sintered (melted). The pockets of the ribbons were filled up with lithium alumino-silicate paste. Following the procedure given in Sec. II, the ribbons were dried in an
FIG. 3. Measured Li\(^{+}\) beam current density (\(J\)) of pulsed source versus time. Thickness of sintered alumino-silicate was about 0.25 mm, but not kept constant. The source surface temperature of \(\approx 1265^\circ\) C and extraction voltage of \(V = \approx 1.75 \text{ kV}\) were unchanged during the data collection.

FIG. 4. Setup of a continuous beam extraction system. The ribbon location with a source pocket is indicated. The extraction plate was 4 mm above the source surface during the beam measurements. The cut-way aperture with the extraction plate is to measure temperature, and far enough from the beam trajectories.

air-furnace at \(\approx 100^\circ\) C for a couple of hours, and then sintered in the vacuum furnace at \(1400^\circ\) C. The mass of alumino-silicate deposited was determined by measuring the weight of the ribbon with and without the alumino-silicate. A biased conducting plate, located at a distance of 4 mm from the source surface, was used as a collector to measure current \(I_c\). With the source at ground potential, the bias of the collector was set at negative 120 V to ensure space-charge limited emission. In this simple diode set up, secondary electrons coming off the collector plate are not suppressed and therefore the measured collector current will include secondary electron current. Fortunately, the secondary electron coefficient is very low [21] at 120 eV ion energy such that the collector current can be expected to be not more than a few percent higher than the actual beam current.

B. Lifetime variation with duty factor

The lifetimes of three ribbon sources were measured by varying their duty factors. The vacuum in the chamber was at \(\approx 10^{-6} \text{ Torr}\). There were 3 cases: (A) continuously on for beam extraction, (B) repeating on for 5 minutes followed by off for 10 minutes, (C) repeating on for 5 minutes followed by off for 20 minutes. Thus the corresponding duty factors are 1, 0.33 and 0.2. The amount of alumino-silicate used was \(\approx 6.9\) mg in case A, and \(\approx 5.8\) mg in both case B and C. The surface area of each of the sources was 3.14 mm\(^2\). For all sources, during on time the temperature was held at \(1265^\circ\) C and the applied extraction voltage was 120 V, whereas during off time the temperature was reduced to \(850^\circ\) C with the extraction voltage turned off. The experiment continued until the sources were nearly depleted. As shown in Fig. 5, after a short initial period the beam current in all 3 cases reached the same level (over 60 \(\mu\text{A}\)), and the steady emission lasted for many hours depending on the duty factor. The elapsed time of case A was \(\approx 16\) hours (from the time the current has approached maximum to the time when the current decreased to \(\approx 50\%)\). For case B and C, the corresponding elapsed time was \(\approx 29\) hrs and \(\approx 49\) hrs. Note that if lifetime is defined as the useful beam time, which is the product of elapsed time and the duty factor, then the measured lifetime for these 3 cases are 16 hrs, 9.6 hrs and 9 hrs. Since case A has a higher alumino-silicate mass than cases B and C, the lifetime comparison should be further normalized by their weights (see next section). The corresponding normalized ratio is 1 : 0.71 : 0.67.

FIG. 5. The beam current versus lifetime of three sources with different duty factors. During on time, the temperature was at \(\approx 1265^\circ\) C and an extraction voltage of 120 V, and during off time, the temperature was at \(\approx 800^\circ\) C and zero extraction voltage. The surface of each of the sources was 3.14 mm\(^2\).
C. Source lifetime depending on the mass of lithium alumino-silicate deposited

Five sources with different mass deposited were measured to compare their lifetime. The other parameters of the experiment such as the distance between the source to collector plate (4 mm gap), the operating temperature, the beam extraction voltage, and the vacuum pressure (10^{-6} \text{Torr}) were unchanged. The emitting surface area of the sources was slightly varied between 3.14 mm$^2$ to 4.15 mm$^2$ due to an unintended error during the source fabrication process. All of the sources were operated at $\sim$1265°C and the beams were extracted, continuously, with 120 V, until the beams current signal was reduced to a minimum detectable level relative to the peak current. The beam current density was expected to have little variation with mass because the space-charge limited current density should be dependent only on the surface area and independent of the source mass deposited in so far as there remain sufficient ions to be emitted. This result is shown in Fig. 6 where the measured beam current density ($J$) was plotted against the deposited source mass for a given area and operating temperature. Figure 7 shows the lifetime depending on the mass of lithium alumino-silicate deposited. The lifetime of the alumino-silicate is measured from the time beam current has transitioned from cleaning phase to operational mode to the time when the beam current has decreased to $\sim$50% of its current at the transition level. As expected, the lifetime increases almost linearly with the lithium alumino-silicate mass deposited in the source.

The theoretically available charge can be calculated from the known mass of lithium in the $\beta$-eucryptite mixture. For example, with 2.8 gm of a $^7$Li $\beta$-eucryptite, the available charge ($Q_a$) is \[
\frac{(0.0028)(5.56\%)}{(7)(1.67\times10^{-24})}1.6\times10^{-19} \text{ C} = 2.13 \text{ C}.
\] Figure 8 shows comparison of the calculated and measured time-integrated charge extracted from the sources. As expected, the extracted charge of the sources was proportional to the mass of the deposited alumino-silicate material. The difference between the measured charge and the theoretical limit is within about 10%.

V. CONCLUSION

In this paper, the procedures used to fabricate lithium alumino-silicate sources were discussed. Typically, a well made source would have glassy-type surface and the substrate surface fully coated without defects. Previously, it was observed that if a source surface was not glassy-type, the emission density could be as low as 1/3 - 2/3 of that with glassy-type surface. Here, a space-charge-limited
emission with current densities exceeding 1 mA/cm² was obtained from 0.64 cm diameter lithium alumino-silicate ion sources when operating at ∼1275°C. The lifetime of a thin coated (on a tungsten substrate) lithium alumino-silicate source was between 30 to 40 hours when pulsed at 0.05 Hz and with pulse length of ∼6 μs each, i.e., a duty factor of 3×10⁻², at an operating temperature of ≥1275°C. A longer lifetime, nearly ≥100 hours was reported recently [16] for similar sources. This time variation could be due to the variation of amount of lithium alumino-silicate mass deposition on the substrate surface as demonstrated in Fig. 7. Nevertheless, the total beam charge extracted during the 40 - 100 hrs with such low duty factor pulsed mode was very small. In comparison, the beam charge that was extracted in ∼16 hrs lifetime when operated in dc mode was near the theoretical limit of the amount of lithium ions contained in the mass of alumino-silicate. This discrepancy suggested that the loss of lithium as neutral vapor was at a similar rate as the ion current extraction when the ion source temperature was at 1265°C. Thus the lifetime of a lithium ion source depends mostly on the total elapsed time that the source is kept at high temperature. Consequently, one way to extend the useful lifetime is by momentarily reduce the operating temperature during the idling time between pulses. This could be a practical method if the pulse rate is very slow as once or twice per minute.

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