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Li$^+$ alumino-silicate ion source development for the Neutralized Drift Compression Experiment (NDCX)

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We report results on lithium alumino-silicate ion source development in preparation for warm-dense-matter heating experiments on the new Neutralized Drift Compression Experiment (NDCX-II). The practical limit to the current density for a lithium alumino-silicate source is determined by the maximum operating temperature that the ion source can withstand before running into problems of heat transfer, melting of the alumino-silicate material, and emission lifetime. Using small prototype emitters, at a temperature of ≈1275 ^{0}C, a space-charge-limited Li$^+$ beam current density of J ≈1 mA/cm^2 was obtained. The lifetime of the ion source was ≈50 hours while pulsing at a rate of 0.033 Hz with a pulse duration of 5-6 μs.

I. INTRODUCTION

The ion source requirements for heavy ion fusion research [1], as well as warm dense matter (WDM) experiments [2] have similar attributes. In these cases, a low duty factor ion beam pulse must be radially focused (compressed) to a small spot [3] and longitudinally compressed to a short pulse [4]. Thus, the ion beam must be launched from an ion source that simultaneously has high current and high brightness [5], and also has on-off gating control. To uniformly heat targets to electron-volt temperatures for the study of warm dense matter with intense ion beams, low mass ions, such as lithium, have an energy loss peak (dE/dx) at a suitable kinetic energy [6]. Using the Neutralized Drift Compression Experiment (NDCX)-II [7] we will create warm dense matter conditions in thin-foils [2] with a Li$^+$ with energy 1.2-4 MeV to achieve uniform heating up to 0.1 - 1 eV. The required beam charge is about 50 nC. The accelerator physics design require that the pulse length at the ion source should be about 500 ns [8]. Thus for producing 50 nC of beam charge, the required beam current is about 100 mA, and the required emittance is about <2 mm-mrad. For NDCX-II, a high current-density emitter is generally preferred so that the source is reasonably compact. In previous studies [9–13], Li ion beams have been used as a diagnostic in magnetically confined fusion experiments. But there appears to be no description of a bright, high current, space-charge limited lithium beam in the literature which meets the needs of our WDM experiments. Several methods to produce a lithium beam are known. With lithium-vapor sources, challenges are posed to the vacuum system, beam reproducibility is not reliable, and beam quality (emittance) is difficult to maintain. Surface ionization sources developed for spacecraft propulsion (usually Cs$^+$) have used a porous, sintered tungsten plug which allowed Cs to be fed from the rear of the assembly to the emitter [14]. The ratio of the emitted alkali ions and neutral atoms is given by the Saha-Langmuir equation. This approach has the advantage of enabling a continuous supply of alkali atoms to the surface. A different approach for supplying alkali atoms to the contact ionizer has been used [15, 16] for the generation of intense ion beams of K$^+$ and Cs$^+$. In this approach an aqueous solution K$_2$CO$_3$ (or Cs$_2$CO$_3$) is deposited on a porous tungsten or iridium substrate. The substrate surface serves as the contact ionizer, as well as a reservoir of alkali atoms. The CO$_3$ is driven off at a temperature below the operating temperature of the emitter. However, in the case of lithium ions, the ion yield and lifetime from these surface ionization sources is unfavorable due to the greater ionization energy of Li. The alumino-silicate source approach appears to be a better choice for beam reproducibility and beam quality; moreover it is easier to handle in a low vacuum pressure. Li$^+$ has been produced by thermionic emission from the alumino-silicates compounds β-Spodumene and β-eucryptite [17–19], but it requires a higher operating temperature than for heavier alkali ions. For example, potassium ion sources, with an ionization energy of 4.34 eV, deliver a current density J ≈20 mA/cm^2, at a temperature of 1050^{0}C. The lithium ionization energy is 5.39 eV, and a temperature greater than 1200^{0}C is required for J=1 mA/cm^2 beam emission. This high temperature can significantly reduce the lifetime of the source, and therefore it is desirable to identify a method to extend the lifetime or to replenish the supply of ions at the surface. The observed Li$^+$ beam current density in previous work [9–13, 17–24] has been in the range of 1 to 1.5 mA/cm^2, but the dependence of the lifetime on temperature, extracted current and repetition rate have not been extensively described. Moreover, process control of the source fabrication is critical due to the tighter tolerances (temperature, duration) for calcination, the leucite phase preparation and sintering, and the significant anisotropy in thermal expansion coefficients. Figure 1 shows some of the fabrication steps of a Li$^+$$\beta$ eucryptite source. Several small (0.64 cm diameter) sources have been operated in a pulsed mode, with similar repetition rate and pulse duration as needed for NDCX-II. This paper describes the beam current density, and lifetime, as functions of
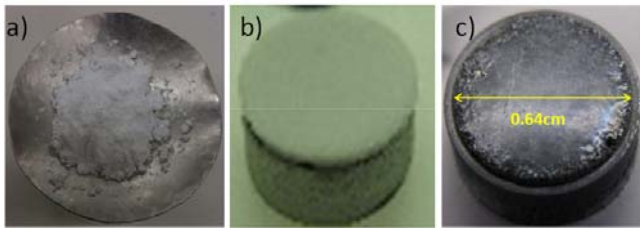
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FIG. 1. (Color online) (a): A sample of reacted Li β-eucryptite ($Li_2O - Al_2O_3 - 2SiO_2$); (b) a 70-80% density tungsten substrate's top surface is coated with Li β-eucryptite materials; and (c) a sintered Li β-eucryptite source, sintered in a vacuum furnace with 96% Ar and 4% H_2 mixture at 4.57×10^{-2} Torr at 1410 ^{0}C, prior to beam extraction measurements.

temperature for lithium alumino-silicate ion sources in preparation for a larger ($\leq$ 11 cm) source for NDCX-II.

II. CHARACTERIZATION OF THE ION GUN

A. Beam emission

In an ion gun injector [25], a thermionic source may be heated by Ohmic heating [26, 27], laser radiation [28], induction heating or other methods [29]. The source surface temperature places a lower bound on the required input power, $P = A\sigma\varepsilon T^4$; where A is the cross sectional area (πr^2) of the emission surface, σ is the Boltzmann constant, ε is the emissivity of material and T is the source surface temperature. This is less than the required input power due to conductive and radiative heat loss from the sides and back of the source assembly. Once operating temperature is achieved, ions are extracted by applying an electric field.

For a pulsed ion source, the total beam charge per pulse is $Q = JA\tau$, where Q is the charge of the beam pulse, J is the current density, A is the emission area (πr^2), with beam radius r, and τ the pulse length. Thus, in general, the ion source specifications are described by the parameters: J, r, τ. The field between the source surface and the extraction electrode influences to define the extracted current density. The space-charge limited current density of an ion gun is defined by the Child-Langmuir law:

$$J\left(\chi, V, d\right) = \chi \frac{V^{3/2}}{d^2}, \qquad (1)$$

where, $\chi = \frac{4\varepsilon_0}{9}\sqrt{\frac{2q}{m}}$, d is the distance between source and extraction electrode, V is the beam extraction voltage, m is the mass of an ion, and q is the ion charge. The space-charge limited beam current, or Child-Langmuir current is
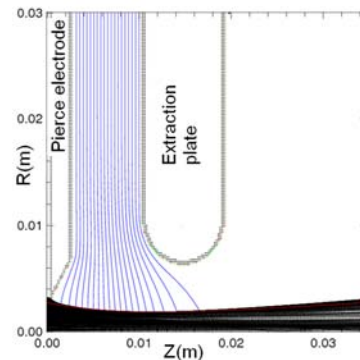
$$I_E = PV^{3/2}, \qquad (2)$$

FIG. 2. (Color online) 10 kV Li$^+$ beam trajectories with equipotential lines calculated using the WARP code. The diameter of the source is 0.64 cm. The extraction electrode aperture plate has a 1-cm diameter aperture. The distance between the source surface and the mid-plane of the extraction electrode is 1.48 cm.

where, $P = \chi\left(\frac{\pi r^2}{d^2}\right)$ is the gun perveance which characterizes the geometry, charge state, and ion mass of the injector. 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 $V^{3/2}$. If the intrinsic emission limit exceeds the space-charge limit everywhere on the surface, then the extracted beam is insensitive to variations in the intrinsic emission properties of the emitter.

In order to operate a source with uniform extraction, it is preferable to operate a source with low enough V to obtain space-charge limited extraction. The space-charge limit effectively smoothes out spatial variation in emission.

Figure 2 shows a particle in cell simulation of the 0.64 cm diameter lithium source test gun using the WARP particle-in-cell code [30]. The axial distance between the inner edge of the 60^0 conical electrode and the emitter surface is 0.4 mm, the non-ideal Pierce electrode angle is a source of some nonlinear aberrations. A 10 kV extraction voltage (V) predicts a beam current and current density of 2.5 mA and 7.9 mA/cm^2, respectively. A single-particle transit time inside the injector for a Li^7 is $\tau = 3d/\sqrt{(2qV/m)} = 85$ ns.

B. Ion species identification

Admixtures of contaminant ions in the extracted beam are due to contaminants in the source materials and vacuum system. Common contaminants are the other common alkalis: K$^+$, Na$^+$, Cs$^+$, which will be easily extracted due to their lower ionization potential. Since the emitter surface also contains Al, O, and Si, these are also possible contaminants. Therefore, it is important to verify ion species purity of the extracted beam. An ExB filter was used as a spectrometer to analyze the ion
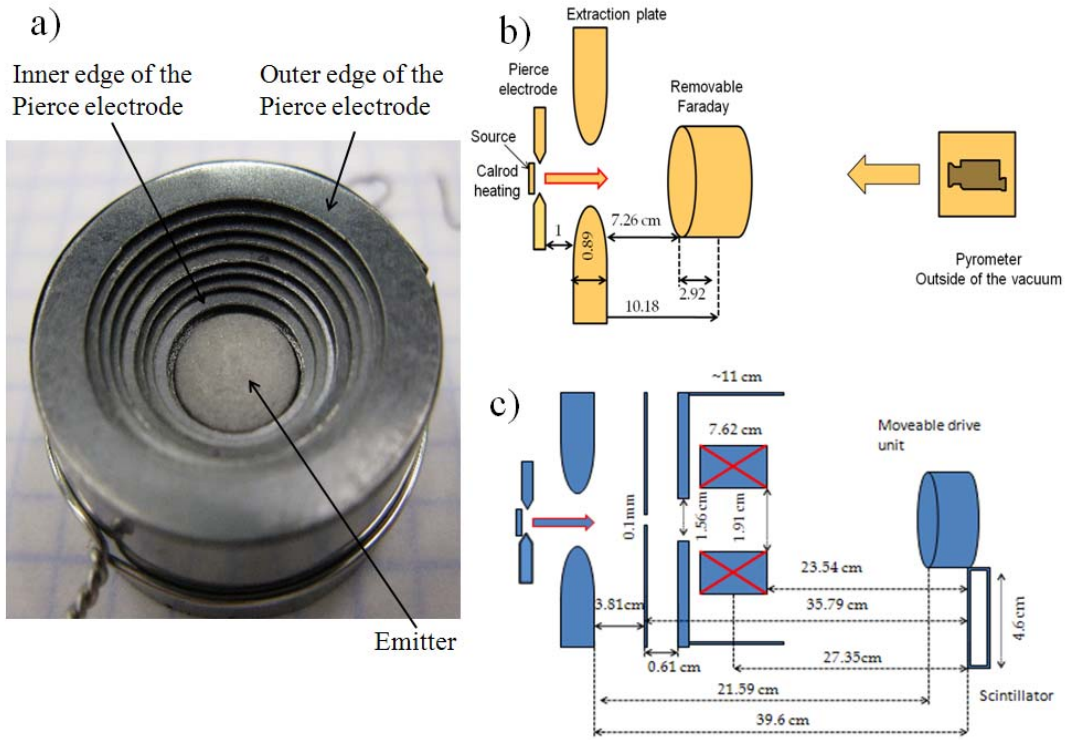
FIG. 3. (Color online) (a) Ion gun: the Pierce electrode was made of stacked molybdenum plates but still maintained a 60 ^{0} opening angle normal to the source surface. The emitter diameter was 0.64 cm. (b) setup for beam current and temperature measurements; and (c) The ExB filter setup.
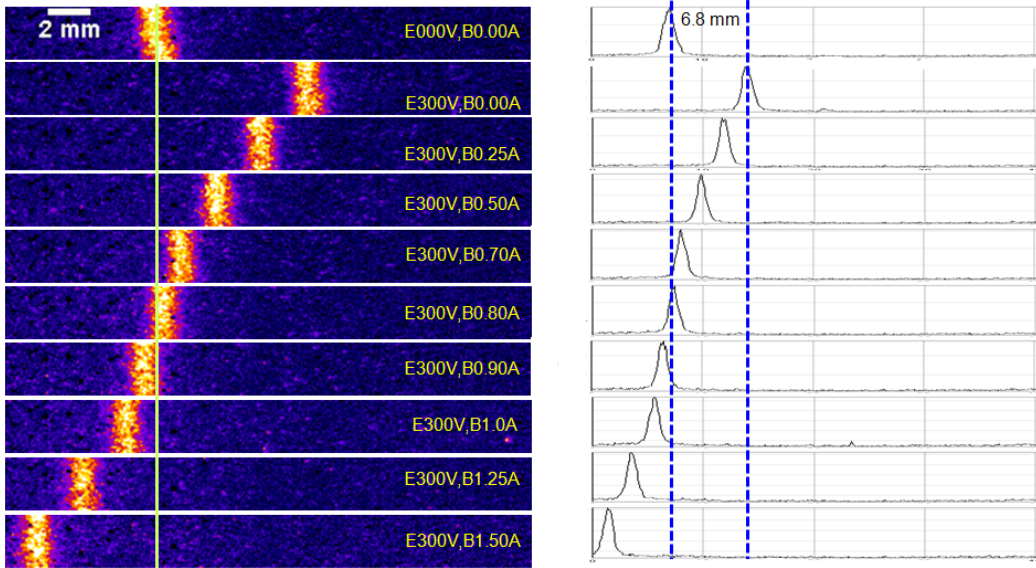

FIG. 4. (Color online) 20 kV Beam images through a 0.1 mm slit on the YAP scintillator. When a magnetic field of 168.5 G (0.83 A) and ±150 V (in total 300 V) with the dipole of the filter were applied, the beam was returned to its original field-free position.

species in the beam. It consists of a dipole magnet and a pair of electrostatic deflection plates. The plates are mounted between the magnet poles to produce an electric field, E, perpendicular to the magnetic field B. The unknown mass of an element is determined by balancing the electric force against the magnetic force as

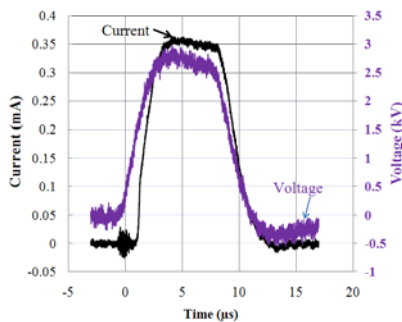$$m = 2qV \left(\frac{B}{E} \right)^2,\tag{3}$$

FIG. 5. (Color online) A typical beam extraction voltage and current waveforms. A 0.64 cm diameter lithium alumino-silicate source was operated at 1260 ^{0}C - 1275 ^{0}C.

where, qV is the ion kinetic energy with potential V across the electrostatic deflector plates.

III. EXPERIMENTAL SETUP TO CHARACTERIZE A FABRICATED ION SOURCE

Figure 3 shows the ion source test stand setup used for 0.64 cm diameter lithium alumino-silicate sources. The length of the beam diagnostics column is $\leq$ 0.5 m, accompanied with a 0.1 mm slit, E x B filter, scintillator, and a Faraday cup. To meet the NDCX-II ion beam brightness requirements, the lithium alumino-silicate source operating temperature was about 1250 ^{0}C-1300 ^{0}C. A tungsten filament was used to heat the emitter to this temperature. In order to minimize heat loss, the Pierce electrode was made of stacked molybdenum plates but still maintained a 60 ^{0} opening angle normal to the source surface (Fig. 3a). The heater package was encased by heat shielding materials (molybdenum foils and ceramics). The beam diagnostics column consisted of a 2.9 cm long (4 cm diameter aperture) Faraday cup to measure beam current; an ExB filter with YAP scintillator to detect the beam; and a pyrometer to measure the source surface temperature. Temperature in this study was measured using a disappearing filament-type brightness pyrometer with null-balance, lamp-current measuring circuit, made by Leeds and Northrup Co, (8632-C series) and calibrated with emissivity 1.0. The pyrometer is sensitive to the brightness at λ = 0.65 microns. The 'brightness temperature' measured using the pyrometer is affected by the emissivity of the alumino-silicate material, and we note that the emissivity of the alumino-silicate at λ=0.65 microns may not be 1.0. The Faraday cup is temporarily removed from the beam axis, without breaking vacuum, when doing the temperature measurement with the pyrometer (Fig. 3b). A source surface temperature of 1250 ^{0}C was measured when 110 to 120 watts rms electrical heating power was applied.

When using the ExB filter, a 0.1 mm slit was installed at the entrance to enable accurate species identification at the detection plane (scintillator). The location of the

filter with respect to the ion source and the scintillator is shown in Fig. 3(c). A higher extraction voltage, such as 20 kV, was used to transport the beam to the scintillator location. A gated, 2 μs gate width, image intensified CCD camera was used to view light emission from the scintillator. Figure 4 shows slit beam images on the scintillator with different values of E and B fields using the EXB filter. When a magnetic field of 168.5 G (0.83 A) and electrostatic potential of $\pm$150 V (in total 300 V) with the dipole of the filter were applied for a 20 kV beam, the beam was returned to its original field free position on the scintillator (E=0, B=0); this identified the element emitted as Li^7. To illustrate the sensitivity to contamination from other alkalis, we note that Na$^+$ would appear at the field-free position for 1.5 A (bottom trace). At this magnetic field strength there was no signature of the beam at the field free location.

IV. LITHIUM BEAM CURRENT DENSITY MEASUREMENTS

Figure 5 shows a 5-6 μs beam extraction voltage and beam current waveforms when the source surface temperature was at 1270 $\pm$ 7 ^{0}C. The beam current was recorded by the Faraday cup (+300 V on the collector plate and -300 V on the suppressor ring).

While keeping the source surface temperature constant, the lithium ion beam current was measured by varying the extraction voltage, up to 10 kV. Figure 6 shows measured beam current density vs. $V^{3/2}$, measured at 1240 ^{0}C, 1260 ^{0}C, 1270 ^{0}C and 1275 ^{0}C. A space charge limited current density of 1.1 mA/cm^2 was measured with extraction voltage of 2.5 kV at 1275 ^{0}C temperature, after allowing a conditioning (surface cleaning) time of about 12 to 30 hours. At the same temperature, the current density was raised to 1.47 mA/cm^2 when the extraction voltage was increased to 10 kV. Beam emission stability with these current density levels was observed for more than 72 hours for a pulse repetition rate of 0.033 Hz. However, the beam current density decreased gradually after this period, leading to the data shown as dotted lines in Fig. 6. Thus the maximum space-charge limited beam current density was reduced to 0.6 - 0.7 mA/cm^2 corresponding to an applied extraction voltage of 1.88 kV (shown as the dotted line with hollow circles in Fig. 6). The same data line (still at 1275 ^{0}C) shows an emission-limited current density of 1.37 mA/cm^2 with 10 kV extraction voltages due to electric field enhancement. The solid line with hollow circles, at the left, colored in red, represents the space-charge limited current density, calculated using the Child-Langmuir law (Eq. 1) for the gun geometry. Beam current density at lower extraction voltage follows the space-charge limited (SCL) Child-Langmuir law. At a higher extraction voltage, there are not enough ions, at a given temperature, on the source surface to extract, and thus extracted current fall below the Child-Langmuir law. It is preferable to run an injec-
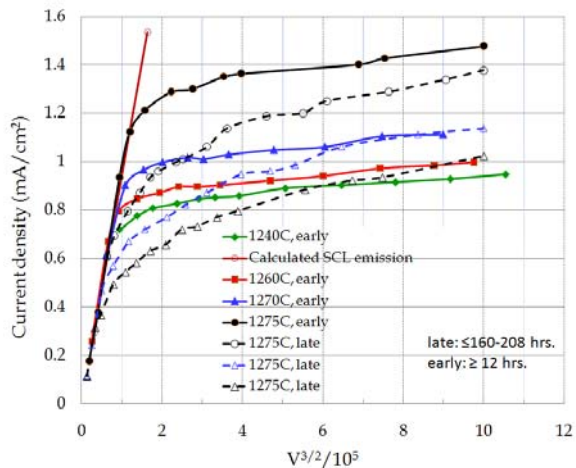
FIG. 6. (Color online) Measured beam current density vs. $V^{3/2}$, measured at 1240 ^{0}C, 1260 ^{0}C, 1270 ^{0}C and 1275 ^{0}C temperatures. The solid lines are for measurements early in the source lifetime. The dashed lines are for measurements late in the source lifetime and shows signs of depletion.

FIG. 7. (Color online) Life test of a Li β-eucryptite source at 10^{-6} Torr pressure. These cases are for pulsed extraction. The duty factor is 10^{-7}.

tor system in the space-charge limited extraction mode, as mentioned earlier.

These data demonstrate that a minimum temperature of 1275 ^{0}C is required for lithium alumino-silicate ion source operation with $\approx$1 mA/cm^2 current density in the space-charge limited mode. The practical limit to the current density for a lithium alumino-silicate source is determined by the maximum operating temperature that the ion source can withstand before running into problems of heat transfer, melting of the alumino-silicate material, and short emission life time.

V. LIFETIME MEASUREMENT

A. Pulsed beam extraction

Figure 7 shows an example of measured lithium beam current density vs. time. At $\approx$1275 ^{0}C and with a 10 kV extraction voltage, the source was delivering a current density $\geq$1 mA/cm^2 for more than $\approx$ 150 hours at a repetition rate of 0.033 Hz. In another test, a current density of $\geq$1 mA/cm^2 was measured for $\approx$ 72 hours with the same beam pulse rate at a higher temperature (1285 ^{0}C). Other measurements of different 0.64 cm sources show that there is a wide variation in conditioning time (12 to 40 hours) to reach $J \geq$1 mA/cm^2, and in lifetime (40 to 200 hours) when $J \geq$1 mA/cm^2. These may be due to the time required for surface mono-layer cleaning, variation of thickness of the coating, or variation of source chemical processing time and temperature from an ideal range. However, these experiments suggest that a larger lithium alumino-silicate ion source could be used for at least a week, without further re-coating, when operating
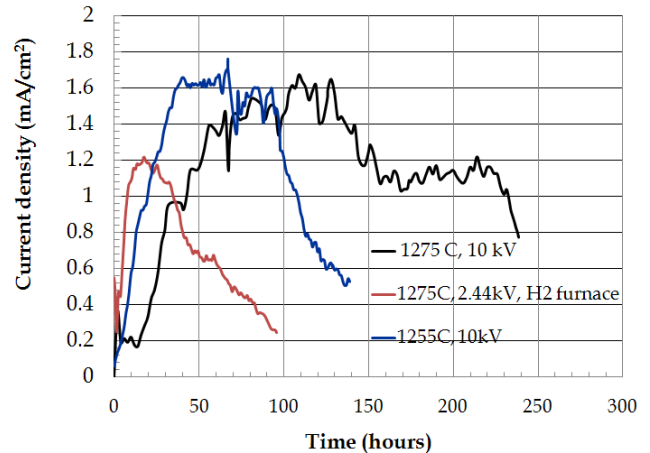
at a current density level of $\approx$1 mA/cm^2 at $\geq$1250 ^{0}C. The lifetime of lithium alumino-silicate ion sources is not as long as for other sources such as potassium [31, 32]. Low duty factor K$^+$ sources have operated for $\approx$ 2 years, several hours per day, 3-4 days per week, at 1100 ^{0}C. The K$^+$ extraction voltage yields a space-charge limited current density of 9 mA/cm^2, while the emission limit is 30-50 mA/cm^2, giving a comfortable margin to accommodate emission variations over the surface.

The lifetime of a lithium source is determined by the loss of lithium from the alumino-silicate material either as ions or as neutral atoms. If the lithium loss is dominated by ions, for high duty factor, then the lifetime will depend on the extracted ion current. On the other hand, if the loss is dominated by neutral atoms (due to the typically low duty factor, even a relatively low neutral evaporation rate can become significant), then the lifetime is simply proportional to the time that the ion source is kept at elevated temperature. Our measurements suggest that for the low duty factor required for NDCX-II, the lifetime of a lithium ion source depends mostly on the duration that the emitter spends at elevated temperature, that is, at $\geq$1250 ^{0}C. That is, lithium loss is due mostly to neutral loss (not charged ion extraction).

B. Continuous beam extraction (DC mode)

In the interest of enabling a more rapid sequence of experiments, we built an in-situ sintering and beam extraction test stand. A known quantity of Li-alumino-silicate material is placed in a $\sim$1.5 mm diameter dimple of a 0.5 cm width molybdenum filament. The source material was melted at 1400 ^{0}C, and then cooled down to a source operating temperature (1250 - 1300 ^{0}C). At that point, the Li-alumino-silicate was nearly flush with the nearby flat part of the filament. Beam was
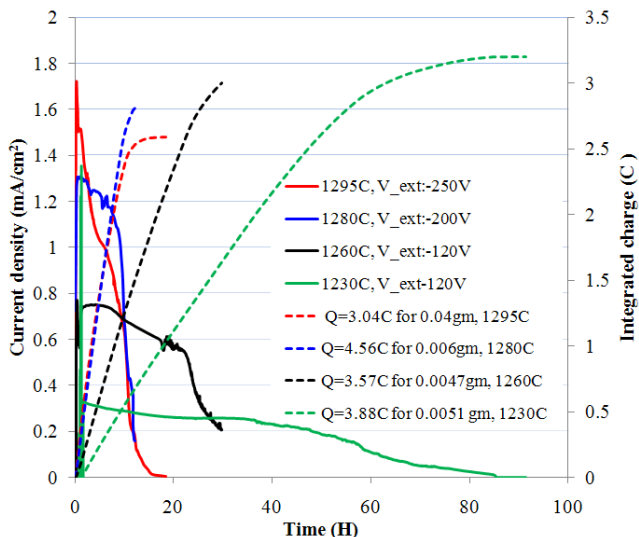
FIG. 8. (Color online) Li$^+$ beam current density, and integrated charge vs. time at various temperatures (1230, 1260, 1280 and 1295 ^{0}C for DC extraction.

VI. NDCX-II ION SOURCE DESIGN AND FABRICATION

The ion source diameter for NDCX-II would be 10.9 cm for 93 mA beam current, assuming 1 mA/cm^2. This source diameter was a compromise between the desire to have higher current and the beam optics challenges of matching a larger diameter beam into the NDCX-II beamline.

In order to understand the experimental heater power requirement for such a large diameter lithium source, we have considered the heater power used to heat a 10 cm diameter K$^+$ source in the high current transport experiment (HCX) [31, 32]. HCX experiments required <2 kW for T=1100 ^{0}C. Extrapolation of the HCX data suggests that ~3 kW power is required for NDCX-II operation at 1300 ^{0}C. The engineering design of the NDCX-II source heater system is underway.

In order to fabricate a large diameter NDCX-II Li$^+$ source, it is necessary to show scalability from the 0.64 cm test samples to $\leq$ 11 cm. At present we are fabricating 2.54 cm test samples, using our experience to control the fabrication process to attach a smooth alumino-silicate layer, about 0.5 mm thick, on top of a 70 to 80% porous tungsten substrate.

VII. CONCLUSION

Characterization studies of a prototype lithium alumino-silicate sources have been presented. Using 6.35 mm diameter prototype emitters (coated and sintered on a $\simeq$ 75% porous tungsten substrate), at a temperature of $\approx$ 1275 ^{0} C, a space-charge limited Li$^+$ beam current density of $\geq$1 mA/cm^2 was measured. At higher extraction voltage, the source is emission limited at around $\geq$ 1.5 mA/cm^2, weakly dependent on the applied voltage. The lifetime of the ion source is $\simeq$ 50 hours when pulsing the extraction voltage at 2 to 3 times per minute. Measurements under these conditions show that the lifetime of the ion source does not depend only on beam current extraction; rather, lithium loss may be dominated by neutral loss or by evaporation. It is inferred that pulsed heating, synchronized with the beam pulse, may increase the lifetime of a source. It is more difficult to make lithium ions than potassium or cesium ions, and the current density of Li$^+$ is typically lower than that of K$^+$. For a given beam current, the current density is inversely proportional to the ion source area. Since both radiation heat loss and beam optics considerations lead us to prefer a smaller ion source, the NDCX-II design seeks to operate the ion source at the maximum current density without running into heat management and lifetime problems. While the operating temperature of a potassium alumino-silicate ion source is about 1050 ^{0}C, for $J \approx 9$ mA/cm^2, the operating temperature of a lithium alumino-silicate ion source is about 1275 ^{0} C for $J \approx 1$ mA/cm^2. This high temperature is part of

extracted to a negative bias plate, located 4 mm from the emitting surface. Figure 8 shows the DC extraction beam current density (primary vertical axis), and the extracted total beam charge (secondary vertical axis) vs. time of beam emission for several samples. The expected charge was calculated from the known mass of lithium in the β-eucryptite mixture. For example, a sample of a ^{7}Li β-eucryptite is 0.0028 gm, the available charge is $[(0.0028gm)(5.56\%)]/[(7)(1.6X10^{-24})]1.6X10^{-19}$ Coulomb $\equiv$ 2.13 Coulomb. For each individual sample, this calculated charge was compared with the measured integrated charge over a period of time until emission was significantly reduced. The space-charge limited Li$^+$ beam current density is about 1 mA/cm^2, at a temperature of >1250 ^{0}C. By comparing pulsed beam extraction and continuous beam extraction, it was observed that there is no significant difference of beam current density level. Interestingly, it was observed that the lifetime of a source is around 10 hours in a DC mode extraction, and the extracted charge is ~75% of the available Li in the sample. One of the samples was operated at 1230 ^{0}C, and its continuous emission lifetime was increased to ~48 hours with current density limit of <0.4 mA/cm^2, with $\sim$ 80% charge extraction. In DC mode extraction, it is also possible to enhance the lifetime of a source by reducing the extraction voltage, i.e., operating in the region of space charge limited field, instead of high field emission extraction. This is not true for a pulsed beam (duty factor $\sim 10^{-7}$) due to neutral Li loss.

the reason for a shorter lifetime of the lithium source in comparison with potassium and cesium ion sources. Our data suggest that a design current density of $1 \, \mathrm{mA/cm^2}$ is satisfactory, in terms of operating temperature, lifetime, and stable emission.

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