Temporal response of a lithium nitrate electrohydrodynamic ion source

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(Received 2 November 1993; accepted for publication 26 January 1994)

The first measurements of electrohydrodynamic (EHD) ion emission from thin films of molten lithium nitrate under pulsed field conditions are presented. Pulse widths from ~20 ns to 50 μ s were used to initiate ion emission from the apex of a stainless-steel field emitter tip coated with molten lithium nitrate. Current densities >100 A/cm² were obtained from a single emission site. As the pulse width was increased from 20 ns to several microseconds the electric field required to initiate ion emission decreased and asymptotically approached the electric field value characteristic of dc operation. In contrast to dc operation, Li⁺ was the dominant species (>90%) for pulse durations less than 1 μ s. The abundance of Li⁺ was independent of pressure between 10⁻³ and 10⁻⁸ Torr of water vapor. The use of a lithium nitrate EHD ion source for the production of intense lithium ion beams in light-ion inertial confinement fusion reactors is foreseen.

Inertial confinement fusion (ICF) employing singly charged lithium ions accelerated in a pulsed electric field remains an attractive fusion alternative despite the difficulty in obtaining an ion beam with suitable characteristics.¹ For example, Sandia's Pulsed Beam Fusion Accelerator II (PBFA II) requires a relatively pure lithium beam (>90% Li^+), with a nanosecond turn-on time and small divergence.² Current densities of \sim 5 kA/cm² of anode surface must be established during a typical 20 ns voltage pulse in order to create a focused beam intensity of approximately 100 T W/cm² that will be required to initiate a significant thermonuclear burn.² In principle, a two-dimensional electrohydrodynamic (EHD) instability in a thin, liquid lithium film at the anode surface (by the voltage pulse) would satisfy these requirements. Theoretical predictions show that in PBFA II a 10 MV, 20 ns voltage pulse (with a rise time of several nanoseconds) could create an array of Taylor cusps in a conducting film at the anode surface having a characteristic cusp spacing of ~ 1 μ m.³ If each cusp produced an emission current of 10 μ A (of Li⁺), currents of 1 kA/cm² would be produced. Unfortunately, vacuum and handling conditions in PBFA II may preclude the use liquid lithium because the EHD instability in thin lithium films will extinguish when the ambient vacuum contains water vapor at a pressure greater than $\sim 10^{-6}$ Torr.⁴ On the other hand, EHD behavior in molten lithium nitrate (LiNO₃) has been observed under dc conditions in high vacuum, and could persist in poor vacuum because the material is intrinsically oxidized.⁵ Although large current densities could be extracted from a single cusp, only a small yield of Li⁺ [less than 10% that of the dominant salt species, $(\text{LiNO}_3)_n \text{Li}^+ n = 1...4$] was reported.^{5,6} Discussions of possible production mechanisms for the dominant salt species suggested that the abundance of Li⁺ might be favored if the EHD source was operated in pulsed mode.⁷ This letter presents the first measurements of EHD emission from molten lithium nitrate under pulsed field conditions.

The molten lithium nitrate ion source used in this study was described previously (for dc operation).⁵ It consists of a field-emitter tip etched from 0.2-mm-diam type 302 stainless-steel wire spot welded to a 6 mm \times 25 mm \times 25- μ m-

thick ribbon of type 302 CRES foil. After etching, the tip is annealed at $\sim 10^{-8}$ Torr to smooth and clean its apex, and to improve its wettability by depleting chromium from its surface.⁸ The tip surface is coated with lithium nitrate by immersing the ribbon and the tip into molten salt, heated in air to ~ 300 °C. Ions are generated when the ribbon is resistively heated in high vacuum (10^{-8} Torr) at a fixed dc bias with a negative high voltage extraction pulse applied to a counterelectrode. A commercial pulse generator was used to produce pulses between 500 ns and 100 μ s.⁹ A cable discharge pulse generator (triggered by a gas-filled spark-gap switch) was used to generate 20 ns pulses. By varying the bias-to-pulse voltage ratio as a function of pulse width, the temporal characteristics of the source could be investigated between dc and 20 ns. The imaging atom probe time-of-flight mass spectrometer was used to record mass spectra at a microchannel plate (MCP) that imaged the source during operation.¹⁰ The distance from the ion source to the extraction electrode was adjusted to obtain ion emission at a dc threshold bias between +2.7 and +2.8 kV. To determine the temporal characteristics of the source as it was operated in pulsed mode, the tip bias was reduced to $V_{dc} = +1.0$ kV, and the extraction electrode was pulsed negative (relative to ground). V_{dc} was chosen to ensure that the electric field (enhanced at the tip apex), in the absence of a pulse, was insufficient to generate Taylor Cone formation. For time-of-flight mass spectroscopy, the tip bias was typically set between +2.0 and +2.5 kV.

The behavior of liquids in high electric fields has been well characterized theoretically and experimentally for many years by Rayleigh, Taylor, and others.^{11–13} When a fluid surface is exposed to a high electric field, the surface tension of the fluid will balance the electrostatic field stress to create a two-dimensional instability at the surface. When a minimum field is established, the instability will grow into a "Taylor Cone" with a characteristic half-angle~49°.¹⁴ As the field is increased, ions will be emitted from the apex of the Taylor Cone primarily by field evaporation.¹⁵ The temporal response of the ion emission process to pulsed extraction fields has been reported for liquid metal films.^{4,16–18}

 Appl. Phys. Lett. 64 (16), 18 April 1994
 0003-6951/94/64(16)/2175/3/\$6.00
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FIG. 1. Negative extraction pulses (inverted for clarity) and the corresponding EHD current from molten lithium nitrate at 300 °C. As the width of the extraction pulse is decreased (at constant pulse amplitude) a minimum pulse width τ is reached comparable to Taylor Cone formation times. Under these conditions, EHD emission becomes erratic, and is extinguished if the pulse width is further decreased.

However, the temporal characteristics of molten salts have not been previously reported.

Taylor Cone formation (and subsequent ion emission) is limited by the rate at which fluid can be drawn to the tip apex. When an electric field is generated at the tip apex, the time to form a Taylor Cone is determined by several parameters, including the viscosity of the liquid, its surface tension, and the value of the electric field.¹⁵ If the electric field is held constant (i.e., the tip bias and extraction pulse amplitude are fixed), ion emission remains stable as a function of decreasing pulse width until the pulse width becomes comparable to the cone formation time. At this point, ion emission becomes erratic, which defines a minimum pulse width (τ) below which a Taylor Cone is not formed. Figure 1 shows this effect for space-charge limited emission (the amplitude of each current profile would remain constant for the duration of each pulse in the absence of space charge). In Fig. 2(a),



FIG. 2. (a) Extraction pulse width (τ) for EHD ion emission as a function of electric field strength relative to the electric field required for stable dc operation (E_{dc}) . (\blacksquare)(\spadesuit)(\spadesuit)(\spadesuit)(\blacksquare)) are successive runs at 10^{-8} Torr, $T \sim 400$ °C; 10^{-8} Torr, T = 298 °C (\triangle); 10^{-4} Torr H₂O, $T \sim 400$ °C(\bigcirc). (b) Data of (a) in the form $\tau(E/E_{dc})^n$, (n = 4.16).

Figure 2(b) shows the functional dependence of τ on the relative electric field strength $E/E_{\rm dc}$. The least-squares fit to the data (for $\tau > 20$ ns) has the form

$$[\tau][E/E_{\rm dc}]^n = \text{constant.} \tag{1}$$

Note that the single data point obtained with the cable discharge pulse generator (τ =20 ns) is accurately predicted by the behavior of the source at much longer pulse widths. The functional dependence exhibited by Eq. (1) is independent of pressure as indicated in Fig. 2 by the insensitivity of the data to source operation at 10^{-4} Torr of H₂O vapor. It is interesting to note that EHD emission from liquid gallium has the same functional dependence exhibited by Eq. (1) for molten lithium nitrate.¹⁶ Perhaps this is not surprising since molten lithium nitrate has resistivity of $\sim 1 \Omega$ cm at its melting point of 298 °C. The ion emission pattern, as viewed on the fluorescent screen of the MCP assembly, changed from a single emission site at longer pulse widths, to multiple emission sites with 20 ns extraction pulses (initiated at a correspondingly higher field strength). When a molten lithium nitrate EHD source is operated under dc conditions, a single emission site is observed at low field strengths, with multiple emission sites appearing as the electric field strength is increased. Both the dc and the pulsed observations are consistent with a theoretical analysis of the growth of twodimensional EHD instabilities which predicts that for given boundary conditions the instability wavelength (i.e., the number of emission sites per unit area) depends on the electric field strength.¹⁹

A time-of-flight mass spectrometer with a field-free drift region of 72 cm was used to analyze the mass species gen-



FIG. 3. (a) Mass species generated by a 500 ns extraction pulse. Notice the small current of ions at amu=76 relative to the Li^+ current. (b) Ion current as viewed on a shorter time scale clearly resolving the ⁶Li⁺ and ⁷Li⁺ peaks in the correct natural isotopic abundances.

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FIG. 4. Mass spectra for EHD ion emission from molten lithium nitrate as a function of extraction pulse widths (τ) between 500 ns and 3 μ s. The salt species (LiNO₃)Li⁺ (m/z=76 amu) is the dominant species at longer pulse widths. As the pulse width decreases, Li⁺ becomes the dominant species.

erated by the source. Figure 3 shows a typical mass spectrum with $\tau \sim 500$ ns. Singly charged lithium dominated the mass spectra. The instantaneous current corresponding to the lithium signal is 200 μ A (10 mV into 50 Ω , or approximately 1000 ions imaged at the CEMA). This corresponds to a current density of $\sim 100 \text{ A/cm}^2$ (using an estimated emission area of 0.1 μ m). The isotropic abundance of lithium, ${}^{6}\text{Li}^{+}/[{}^{6}\text{Li}^{+}+{}^{7}\text{LI}^{+}]=8\%$, is in agreement with the natural abundance (7.5%) expected for these isotopes. Other species in the mass spectra include NO⁺ at m/z=30, Fe⁺ at m/z=56, (LiNO₂) Li^+ at m/z=60, and (LiNO₃) Li^+ at m/z=76. These species are also observed for dc operation of the source.¹³ However; during dc operation of the source the salt species (LiNO₂)Li⁺, and (LiNO₃)_nLi⁺ (n=1...4) are dominant, with the larger cluster ions appearing at least an order of magnitude more abundant than lithium.¹⁸ To explore this unexpected observation, mass spectra were recorded as a function of extraction pulse width (500 ns $<\tau<3$ µs) as shown in Fig. 4. For pulse widths greater than 2 μ s, (LiNO₃)Li⁺ dominates the mass spectrum. As the pulse width is decreased, the relative abundance of (LiNO₃)Li⁺ to

Li⁺ decreases. For $\tau = 500$ ns, the abundance ratios are reversed and Li⁺ dominates the mass spectrum. The data suggest that the salt species (LiNO₂)Li⁺ and (LiNO₃)_nLi⁺ (n = 1...4) require several microseconds to form prior to EHD extraction. Future studies will investigate the lithium ion abundance at $\tau = 20$ ns.

The authors wish to acknowledge many helpful discussions with Bob Gerber and Regan Stinnett of Sandia National Laboratories (Albuquerque). This work supported an Advanced Undergraduate Honors Project (for M.B.) under Grant No. DE-FG04-88ER45348 from the United States Department of Energy (Office of Basic Energy Sciences). Additional support was provided by Sandia National Laboratories under Contract No. 75-4246.

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