Study on the Thermionic-Emission-Type Cs-Ion Source

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A thermionic-emission-type Cs\(^+\)-ion source based on a \(\beta\)-eucryptite-like material was fabricated, and its emission characteristics were investigated. The results obtained by using a residual gas analyzer showed that the material had a small amount of outgassing in an ultra-high vacuum even at high source temperature. The estimated Cs\(^+\) ion emission energy was 4.03 eV, meaning that Cs\(^+\) ions could be easily emitted. The current density at the source surface was 0.46 A/m\(^2\), the temperature was 1173 K \(\sim\) 1373 K, and the filament potential was 300 V. The halflives of this specimen were \(2.31 \times 10^3\) min, \(1.98 \times 10^2\) min and \(2.77 \times 10\) min for filament temperatures of 1173 K, 1273 K and 1373 K, respectively, meaning that this material has a very long lifetime. The estimated decay time constant function is very useful for determining the ion beam current or the total dose in an ion implantation study. In conclusion, these results can be applied to various research fields, such as depth profiling and secondary-ion mass spectroscopy, and can be used to construct Cs\(^+\) emitting devices with a simple structure.

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I. INTRODUCTION

The Cs\(^+\) ion is very useful for various research purposes. In particular, because of its heavy mass number, it is effective for sputtering experiments [1] and secondary ion mass spectroscopy (SIMS) studies [2] and it is used to generate negative ions [3–5]. Although Cs\(^+\) ions can be applied widely in various kinds of research, it is too difficult to generate Cs\(^+\) ions because Cs metal is very reactive chemically, and so it is too difficult to handle. Therefore, we need special apparatus to generate Cs\(^+\) ions. The duoplasmatron is one of the well-known apparatus that is frequently used to generate Cs\(^+\) ions [6–8]. Although we can get a high Cs\(^+\) ion beam current using a duoplasmatron, it has some use restrictions because its structure is too complicated and it is too big. So, most duoplasmatrons are used in big experimental systems. Several studies have reported on replacing the duoplasmatron with simple Cs\(^+\) ion generating devices.

Field desorption is one of the good Cs\(^+\) ion generating methods [9]. But the structure of a field desorption Cs\(^+\) ion generating apparatus is also complicated and the Cs\(^+\) ion beam current generated is low. In particular, it is too difficult to handling this apparatus because the Cs\(^+\) ion emitter is very sharp needle. G. Kuskevics and B. Thompson [10] reported that they could get Cs\(^+\) ions using a surface ionization [11] method. They used a spherical powder tungsten ionizer as a neutral Cs atom adsorber. But the structure of their Cs\(^+\) ion source was also too complicated. Up to now, a simple Cs\(^+\) ion source has not been found. Therefore, it is strongly requested to develop a simple Cs\(^+\) ion emitting device like an electron emitting hot cathode.

In this study, I fabricated thermionic emission type Cs\(^+\) ion emitting material and investigated its emission characteristics. The merits of this material are its very simple ion emission process, no outgassing in ultra-high vacuum, high current density and long lifetime. Overall, this material can be used in various research fields such as SIMS, ion implantation and sputtering experiments.

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II. EXPERIMENT

Although alkali atoms are chemically reactive, the alkali atoms in a compound should be stable. If we can emit alkali atoms from a compound like alumina-silicate material in an ion state but not neutral atoms, that material could be a good ion source. It is known that Handley [12] used alumina-silicates of lithium as a Li\(^+\) ion emitter and the emitter was used as an ion source for an accelerator in 1927. Since Handley’s work, C. C. Lee and his coworkers [13] succeeded in constructing a Li\(^+\) ion source using \(\beta\)-eucryptite (\(\text{Li}_2\text{O}\cdot\text{Al}_2\text{O}_3\cdot2\text{SiO}_2\)) [14], which is chemically stable even in the atmosphere.

It is known that \(\beta\)-eucryptite (\(\text{Li}_2\text{O}\cdot\text{Al}_2\text{O}_3\cdot2\text{SiO}_2\)) has highly one-dimensional Li\(^+\) conductivity [15–17], and one negative coefficient of thermal expansion. Its linear coefficients in the crystallographic \(\alpha_c\) direction are \(-17.6 \times 10^{-6}/\circ\text{C}\), whereas \(\alpha_a\) is \(8.21 \times 10^{-6}/\circ\text{C}\) [18]. Because of this net effect of anisotropic expansion, there are central openings for the Li\(^+\) ions to be readily emitted from this type of crystal lattice. The elements in Group I have similar chemical properties. Therefore, I infer that if Li atoms in the \(\beta\)-eucryptite could be substituted by elements in Group I, those elements in the \(\beta\)-eucryptite-like materials could also be emitted. Cs is the element in Group I, and Li atoms in the \(\beta\)-eucryptite could be substituted by Cs atoms. To prepare the Cs\(^+\) ion emitting \(\beta\)-eucryptite–like material, (1) I mixed Cs\(_2\)CO\(_3\), Al\(_2\)O\(_3\), and SiO\(_2\) powders uniformly with molar ratios of 1, 1, and 2. Furthermore, I made this mixed powder into briquettes by compressing them with a pressure of \(1 \times 10^4\) N/cm\(^2\). (2) By considering the melting point of Cs\(_2\)CO\(_3\), 883 K, and decomposing from this temperature, a briquette was heated up to 1000 K using a tungsten filament in a high vacuum chamber for several minutes. In this process, the briquette was placed inside the spiral type tungsten. I measured the filament resistances and converted the resistance to temperature. I performed this process in the high vacuum chamber to prevent contamination by oxygen, nitrogen, or other gases and to get a high purity ion source.

It is suggested that Cs\(_2\)CO\(_3\) breaks down into Cs\(_2\)O + CO\(_2\) by thermal decomposition [19]. By heating the briquette, we can get \(\beta\)-eucryptite–like material. This process is shown in Eq. (1).

\[
\text{Cs}_2\text{CO}_3 + \text{Al}_2\text{O}_3 + 2\text{SiO}_2 \rightarrow \text{Cs}_2\text{O} \cdot \text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 + \text{CO}_2
\]  

(1)

If we heat Cs\(_2\)O-Al\(_2\)O\(_3\)-2SiO\(_2\), as explained above, the Cs\(_2\)O in this material decomposes and Cs can be emitted in an ion state. This process is shown in Eq. (2).

\[
2\text{Cs}_2\text{O} \cdot \text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \rightarrow 4\text{Cs} + \text{O}_2 + 2\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2
\]  

(2)

To investigate the emission characteristics of the \(\beta\)-eucryptite–like Cs\(^+\) ion emitting material, very fine Cs\(_2\)O-Al\(_2\)O\(_3\)-2SiO\(_2\) powder was painted on the tungsten filament like an electron emission filament in a vacuum tube. The dimensions of the spiral tungsten filament were 0.7 mm in diameter and 5 mm in length. I placed a copper plate in front of the filament to measure emission current. The filament and the copper plate were inserted in the vacuum chamber with a base pressure of \(1.3 \times 10^{-7}\) Pa. I measured emitted Cs\(^+\) ions as well as impurities using an electrometer and a residual gas analyzer (RGA) placed near the copper plate. Although most of the emitted Cs\(^+\) ions are adsorbed on the copper plate, a small part of the ions are scattered and the RGA detects these scattered ions.

III. RESULTS AND ANALYSIS

1. Confirmation of Cs\(^+\) ion emission

Fig. 1 shows the RGA spectra when the filament temperature is 1100 K and the filament potential with respect to the copper plate is 350 V. As shown in this figure, when the filament is heated and positive potential is applied to the filament, strong new Cs peak and Cu peak appear. The Cs atom is heavy, and so the sputtering yield by incident Cs atoms is high. Therefore, the Cu peak should be induced by the sputtering by incident Cs atoms even at low incident energy. This result means that this ion source is effective in the sputtering experiments. If the incident Cs\(^+\) ion energy is high, one can expect very high Cu peak. \(\text{O}_2\) peak is induced by the decomposing of Cs\(_2\)O, and small increments of H\(_2\) and N\(_2\) peaks are observed when some material is heated in a vacuum. Some of these increments may have been induced by desorbing phenomena from
the surroundings due to thermal radiation from the hot filament, and some of these increments are directly from the hot filament. Usually, these peaks disappear when there is heating for a long time. These spectra show that Cs\(^+\) ions were emitted from the specimen because Cs\(^+\) ions were not detected when the potential was not applied to the specimen, even when specimen was heated. However, the comparison of these two spectra (filament on and filament off) shows that the purity of the emitted Cs\(^+\) ion beam is high. The RGA detected only scattered Cs\(^+\) ions and Cs atoms, but not all of the emitted Cs\(^+\) ions. The Cs atom is heavy, and so most of the Cs\(^+\) ions implanted in the copper target or adsorbed on the walls of the vacuum chamber and a small amount of the emitted Cs\(^+\) ions can arrive at the RGA detector. However, the peak heights of elements are not proportional to the number of atoms in the elements. If the RGA can detect all of the emitted Cs\(^+\) ions, the Cs peak should be stronger than this Cs peak.

In summary, the RGA results show that the fabricated material (Cs\(_2\)O-Al\(_2\)O\(_3\)-2SiO\(_2\)) emits Cs\(^+\) ions by heating with ignoble impurities.

2. Cs\(^+\) ion beam current versus filament potential

Fig. 2 shows the plot of Cs\(^+\) ion current versus filament potential when the filament temperature is 1373 K. The filament potential is 300 V. The Cs\(^+\) current density at the filament surface is estimated to be 48 A/m\(^2\). This figure shows that the Cs\(^+\) current does not proportional to the filament potential, but increases exponentially. It is known that the relation between current density \(J\) and filament potential \(V\) calculated using Poisson’s equation is

\[
J_{cy} \approx \left( \frac{16\pi e}{9D^2} \right) \left( \frac{2e}{m} \right)^{1/2} V_0^{3/2}
\]

(3)

where \(J_{cy}\) is the current density calculated in cylindrical coordinate, \(D\) is the distance between the filament and the plate, \(e\) is the elementary charge and \(m\) is the mass of the emitted particle and \(V_0\) is the potential difference between the filament and the plate [19].

Fig. 2 does not show the detailed exponent of \(V_0\) in Eq. (3). The logarithmic plots are helpful to get the detailed exponent.

\[
\ln J = \ln \eta + \frac{3}{2} \ln V_0
\]

(4)

where \(\ln \eta\) is a proportional constant that depends on the shapes of the filament and the plate.

Fig. 3 shows the plot of \(\ln I\) versus \(\ln V\). The graph shows a straight line, and the slope of this line is 1.446. These values are in good agreement with Eq. (4). Although the current density is proportional to \(V^{3/2}\), the density should be saturated because the number of emitting Cs atoms in the specimen is finite. However, we could get \(\sim 50 \mu A\) under this experimental conditions. If
one changed the experimental conditions \((V, \text{dimensions of filament, temperature})\), they could get a higher current. But a higher current results in a shorter lifetime for the source.

3. \(\text{Cs}^+\) ion beam current versus filament temperature

In the case of electrons, it is known that the emission current from a hot cathode is given by following the Richardson equation \([21]\),

\[
J = A T^2 e^{\frac{E}{kT}}
\]  \(5\)

where \(A\) is a constant given by the physical or chemical properties of the emitter, \(E\) is the work function of the emitter, and \(k\) is the Boltzmann constant. Although Eq. (5) was derived to explain electron emissions from a solid, it is still possible to use for ions because when they derived Eq. (5), they did not distinguish electrons from positive ions. In the case of ions, the \(E\) in Eq. (5) is the minimum energy required for ions to escape from solid to vacuum.

Fig. 4 shows a plot of \(\text{Cs}^+\) ion current versus filament temperature. It shows the exponential increment of current as the filament temperature increases. It is convenient to use the following logarithmic plot to estimate the minimum energy \(E\) for \(\text{Cs}^+\) ions to escape from solid \((\text{Cs}_2\text{O-Al}_2\text{O}_3\cdot2\text{SiO}_2)\) to vacuum,

\[
\ln \left(\frac{J}{T^2}\right) = \ln A - \frac{E}{kT}
\]  \(6\)

Fig. 5 shows the plot \(\ln \left(\frac{J}{T^2}\right)\) versus \(1000/T\) when the filament potential is fixed at 300 V. The slope of linear fit is 34.8. Although the work functions of the good electron emitters are much smaller than this value, this value is still reasonable because the work function of \(W\) is \(4.5 \sim 5.3\) eV \([22, 23]\) and \(W\) is frequently used as an electron emitter.
Fig. 6. Normalized Cs⁺ ion current versus working time.

4. Cs⁺ ion beam current versus working time and the lifetime of this material

Fig. 6 shows the normalized Cs⁺ ion current versus working time. In this experiment, the filament potential is fixed at 300 V. The initial currents were 0.5 μA, 5.0 μA and 52 μA when the filament temperature was 1173 K, 1273 K and 1373 K, respectively. Note that these currents are obtained without focusing. If the emitted ion beam is focused, the current densities may be very high. However, the corresponding current densities at the emitter surface were 0.46 A/m², 4.6 A/m² and 48 A/m², respectively. The ion currents decreased as the working time increased. When the filament temperature was 1373 K, the ion beam current decreased about 99%, but when the filament temperature was 1173 K, the current decreased only 5% during 6 hours of working time. If we operate it with low temperature, we can keep an almost stable ion beam current density.

This is a logical result because the number of Cs atoms in the specimen is finite. The emitted Cs atoms per unit time is proportional to the number of Cs atoms in the specimen and it is given by \( \frac{dn}{dt} = -\alpha n \), where the negative sign means the decrement of Cs atoms in the specimen. If we set the initial number of Cs atoms in the specimen is \( n_0 \), we can get the following equation,

\[
n = n_0 \exp \left( -\frac{t}{\tau} \right) \tag{7}
\]

where the decay constant \( \tau \) is \( 1/\alpha \) and \( t \) is working time. The emission current should be proportional to the number of Cs atoms in the specimen. Therefore, the time-dependent current is given by,

\[
I = I_0 \exp \left( -\frac{t}{\tau} \right) \tag{8}
\]

Fig. 7 shows the plot \( \ln (I/I_0) \) versus working time to determine the decay constants. One can determine \( 1/\tau \) with the slopes of each straight line. The decay constants determined are 3.33 × 10³/min, 2.86 × 10²/min and 4.00 × 10/min for filament temperatures 1173 K, 1273 K and 1373 K, respectively.

Therefore, the half-lifetimes of this specimen are 2.31 × 10³ min, 1.98 × 10² min and 2.77 × 10 min for filament temperatures 1173 K, 1273 K and 1373 K, respectively. Using these values, the decay constant determined as a function of temperature is,

\[
\tau = 6.54 \exp \left( \frac{-T}{41.43} \right) \tag{9}
\]

where the unit of \( \tau \) is minutes and the unit of \( T \) is K.

If we integrate the current with time, we can get the total number of Cs atoms in the specimen, i.e.,

\[
n = \frac{1}{e} \int_0^\infty I dt = \frac{1}{e} \int_0^\infty I_0 e^{-\frac{t}{\tau}} dt = \frac{1}{e} I_0 \tau \tag{10}
\]

Substitution of Eq. (9) into Eq. (10) yields the following equation,

\[
n = \frac{6.54}{e} I_0 \exp \left( \frac{-T}{41.43} \right) \tag{11}
\]
The initial current $I_0$ depends on the filament temperature and filament potential. However, the total number of Cs atoms in this specimen is determined to be $4.5 \times 10^{17}$. This means that when there is low current-long time or high current-short time ion implantation, the dose converges at $4.5 \times 10^{17}$.

If we combine the three factors described above, the current is given by following equation,

$$I = AT^2V^{3/2}\exp \left(-\frac{E}{kT} + \frac{t}{\tau}\right)$$  \hspace{1cm} (12)

If someone wants to use this material as a dosing source in an implantation experiment, Eqs. (9) and (11) may be very useful because they can easily determine the total number of implanted ions, and if they want to use this material for a SIMS experiment, it is also useful because the sputtering yield of heavy Cs$^+$ ions is high.

Finally, I am sure that if someone wanted to construct a Cs$^+$ ion gun using this material, the structure of the gun could be very simple because the Cs$^+$ ion emitting part is very similar to the cathode of an electron gun.

**IV. CONCLUSION**

In this study, I fabricated a thermionic emission type Cs$^+$ ion source material. This material is helpful to construct a simple structured Cs$^+$ ion emitting apparatus. Furthermore, because the Cs atom is heavy and the ionization energy is low, this material can be applied to various experiments such as ion implantation SIMS and negative ion production. When I painted it on a filament and heated the filament, the RGA results showed a Cs peak, a Cu peak and no other peak except for a small amount of increments of N$_2$, H$_2$ and O$_2$ gases. The Cu peak is due to sputtering by incident Cs$^+$ ions. This result means that the purity of emitted Cs$^+$ ions is fine. The estimated Cs$^+$ ion emission energy was 4.03 eV, meaning that Cs$^+$ ions can easily be emitted compared with electron emission from a hot tungsten cathode. When the filament temperature was 1373 K and filament potential was 300 V, the initial ion beam current was 52 $\mu$A and decreased by 99%, but when the filament temperature was 1173 K, the initial current was 0.5 $\mu$A and decreased by only 5% during 6 hours of working time. However, this specimen has a long lifetime. The decay time constant function determined is very useful to determine the ion beam current and lifetime of the ion source material. Therefore, I conclude that the fabricated Cs$^+$ ion source has a good outgassing characteristics, a high purity, a high ion current density, a reasonable ion emission energy, a very long lifetime, and a good beam stability. If one operate it at a low temperature, one can keep almost stable ion beam current density. If these beam currents are focused, I think these current densities are high enough to use this material in a SIMS study as well as in other applications. Thus, the fabricated material can be widely applied in various research fields and I can conclude that this results provide meaningful implications and will help to construct simple structured Cs$^+$ ion emitting devices.

**REFERENCES**