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A Recipe to Make Zeolite Ion Source for Plasma Diagnostics Beam

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Abstract

A mathematical model is presented to describe replacement process to make zeolite containing a particular species of alkali ions for the beam source. The fraction of the alkali ions trapped in a zeolite lattice can be expressed as a function of the number of the times of substitution reaction in a concise recursion formula. The formula is used for a simple estimation of efficiency for making alkali zeolite in terms of time and cost. A rough comparison between the model and an experiment is presented.

keywords: alkali ion source, zeolite, substitution reaction, plasma diagnostics

Zeolite is a porous material containing (or trapping) alkali ions in its lattice, and the alkali ions can be extracted by heating up zeolite above an appropriate temperature. Owing to the property zeolite is often utilized as an alkali ion source such as sodium, cesium and thallium. In our laboratory, cesium zeolite is used for a heavy ion beam probe[1] to measure potential, density and their fluctuations in high temperature plasmas.

The cesium zeolite is produced by ourselves through a substitution reaction from molecular sieves (or sodium zeolite), since the zeolite allows to exchange its own ions with other ions. This is a convenient way for a laboratory to make an ion source. However, the self-made cesium zeolite is contaminated with the remaining sodium unnecessary for our purpose. It is essential to increase the target alkali fraction in the zeolite in order to obtain higher current. This short note presents consideration on an efficient manner to refine a zeolite source to contain more fraction of the favorable alkali element.

In our laboratory, the cesium zeolite is made as follows. Cesium compound is solved into water, then sodium zeolite is put into the solution. The solution is stirred for several hours in temperature of approximately $70^{\circ}C$ until the substitution is completed. The solution is percolated to weed the sodium compound out (although cesium compound is removed simultaneously). The zeolite is put into the water again, in which appropriate amount of cesium compound is newly soluted. The same process can be repeated for several times. Finally, zeolite with a certain amount of cesium fraction is produced.

In the first place, the process of substitution can be described in the following model, as

$$\frac{dN_1}{dt} = -\alpha N_1 + \beta N_2 + \gamma(N - N_1 - N_2), \quad (1)$$

$$\frac{dN_2}{dt} = +\alpha N_1 - \beta N_2 + \delta(N - N_1 - N_2), \quad (2)$$

where N , N_1 and N_2 are the total number of the *holes* to trap alkali ions, the number of the holes to be occupied by the first chemical element, and that by the second chemical element, respectively. The parameters α and β are the rates of replacement, and the γ and δ are the rates of trapping. Summation of these two equations proves that the total number of the holes is kept constant.

In equilibrium, the solution is described as

$$N_1 = \frac{\beta}{\alpha + \beta}N, \quad N_2 = \frac{\alpha}{\alpha + \beta}N \quad (3).$$

Note that the number of occupation for the first chemical element N_1 is identical with the

average value predicted from a Bernoulli distribution when the probability for the first alkali element to occupy a hole is assumed to be $\beta/(\alpha + \beta)$.

The coefficient α (or β) is supposed to increase as the total number of the second (or first) chemical element increases. The simplest assumption is $\alpha = \alpha_0 M_2$, $\beta = \beta_0 M_1$, where the variables M_1 and M_2 are the concentrations of the first and second elements in the water. Then, the formula can be rewritten into

$$N_1 = \frac{M_1}{M_1 + (1 + \varepsilon)M_2} N, \quad N_2 = \frac{(1 + \varepsilon)M_2}{M_1 + (1 + \varepsilon)M_2} N \quad (4)$$

where the ratio of $\alpha_0/\beta_0 = 1 + \varepsilon$ ($-1 < \varepsilon < \infty$) is used for simplicity.

Next, we assume that the newly added amount of the first alkali compound is $\Delta \cdot N$ in every substitution process, and that all the compounds are removed with the percolation process. Under this assumption, the alkali fractions can be expressed as a function of the number of times of substitution processes in a recursion formula. By introducing new variables $p(n) = N_1/N$ and $q(n) = N_2/N$ which represent the fractions of the first and second alkali ions after the n -th time substitution, respectively, the concentrations of alkali ions in solution for the n -th substitution are found to be $(p(n) + \Delta)N$ and $q(n)N$ for the first and the second components, respectively. According to Eqs. (3,4), the variables $p(n + 1)$ and $q(n + 1)$ can be connected to the fraction of n -th time, $p(n)$ and $q(n)$, as

$$p(n + 1) = \frac{p(n) + \Delta}{p(n) + \Delta + (1 + \varepsilon)q(n)}, \quad q(n + 1) = \frac{(1 + \varepsilon)q(n)}{p(n) + \Delta + (1 + \varepsilon)q(n)}. \quad (5)$$

This recursion formula can be solved to make the ratio $p(n)/q(n) (\equiv a(n))$ with utilizing the relation of $p(n) + q(n) = 1$; the recursion formula for $a(n)$ is quite simplified into $a(n + 1) = \Xi a(n) + \Delta(1 + \varepsilon)^{-1}$, where $\Xi = (1 + \Delta)/(1 + \varepsilon)$. Then the recursion formula can be easily solved.

The solution is explicitly written as

$$p(n) = \frac{\Xi^n [a(1)(1 - \xi) + 1] - 1}{\Xi^n [a(1)(1 - \xi) + 1] - \xi}, \quad q(n) = \frac{1 - \xi}{\Xi^n [a(1)(1 - \xi) + 1] - \xi}, \quad (\Delta \neq \varepsilon) \quad (6)$$

or

$$p(n) = \frac{n\Delta + a(0)(1 + \Delta)}{n\Delta + (a(0) + 1)(1 + \Delta)}, \quad q(n) = \frac{1 + \Delta}{n\Delta + (a(0) + 1)(1 + \Delta)}, \quad (\Delta = \varepsilon) \quad (7)$$

where $\xi = \varepsilon/\Delta$ and $a(0) = p(0)/q(0)$. In the case of $a(0) = p(0) = 0$, the formula is simplified into the following form,

$$p(n) = \frac{\Xi^n - 1}{\Xi^n - \xi}, \quad q(n) = \frac{1 - \xi}{\Xi^n - \xi} \quad (\Delta \neq \varepsilon), \quad (8)$$

or

$$p(n) = \frac{n\Delta}{n\Delta + (1 + \Delta)}, \quad q(n) = \frac{1 + \Delta}{n\Delta + (1 + \Delta)} \quad (\Delta = \varepsilon). \quad (9)$$

According to the analysis, two characteristic regions are identified by the sign of ε . When $\varepsilon < 0$, the complete replacement ($p \rightarrow 1$ for $n \rightarrow \infty$), is possible for any small amount of the substitution compound. When $\varepsilon > 0$, the final fraction of the first element is controlled by the amount of the compound. In the case of the latter, the complete replacement is possible for $\Delta > \varepsilon$, while the maximum fraction is limited below $\Delta/\varepsilon (< 1)$ for $\Delta < \varepsilon$. When $\Delta = \varepsilon$, the complete replacement is possible but in the slowest manner. Figure 1 shows changes of the first element fraction as a function of the number of times of substitution processes for four cases; i) $\varepsilon = -0.5$, $\Delta = 0.5$, ii) $\varepsilon = 0.5$, $\Delta = 1.0$, iii) $\varepsilon = 0.5$, $\Delta = 0.5$, and iv) $\varepsilon = 1.0$, $\Delta = 0.5$.

The efficiency can be evaluated in terms of time and cost. The number of times of substitution processes n necessary to obtain a fraction of the first element, p_T , is described as a function of the amount of the compound, Δ , as

$$n(p_T, \Delta) = \log[(\Delta - p_T\varepsilon)/\Delta(1 - p_T)]/\log[(1 + \Delta)/(1 + \varepsilon)], \quad (\Delta \neq \varepsilon), \quad (10)$$

$$n(p_T, \Delta) = p_T(1 + \Delta)/\Delta(1 - p_T), \quad (\Delta = \varepsilon). \quad (11)$$

The total amount of the necessary compound, $\zeta = n \times \Delta$, is expressed as

$$\zeta(p_T, \Delta) = \Delta \log[(\Delta - p_T\varepsilon)/\Delta(1 - p_T)]/\log[(1 + \Delta)/(1 + \varepsilon)], \quad (\Delta \neq \varepsilon), \quad (12)$$

$$\zeta(p_T, \Delta) = p_T(1 + \Delta)/(1 - p_T), \quad (\Delta = \varepsilon). \quad (13)$$

The parameter ζ can be used as an indicator to evaluate the cost efficiency. The minimum amount of the compound, or minimum cost, can be found for a fixed fraction of the favorable alkali element. Note that Eqs. (10)-(13) are evaluated with Eqs. (8) and (9).

Figure 2 illustrates examples of the times and cost efficiencies for three cases with $p_T = 0.8$ for three different values of $\varepsilon = -0.2, 0.5$ and 1.5 . In all cases, as is expected, the necessary times of substitution decreases as the amount of the compound increases. In the first case, the total amount of the compound (or the cost) monotonically increases; a replacement process is enough if the parameter $\Delta > p_T(\varepsilon + 1)/(1 - p_T)$ is chosen. In the other cases, the maximum of the cost efficiency (or minimum cost) exists for the choice of the parameter Δ . In the second case ($\varepsilon = 0.5$), the cost minimum is achieved for the four times replacements $n=4$ at $\Delta \simeq 1$, while seven times replacements $n = 7$ are necessary for the third case ($\varepsilon = 1.5$) at the choice of $\Delta \simeq 3$.

A rough comparison between the model and an experiment is shown in Fig. 3. Here, the experiment was performed with the condition of $\Delta = 1$ using zeolite ($Na_{12}[(AlO_2)_{12}(SiO_2)_{12}] \cdot 27H_2O$) and cesium chloride (CsCl). Corresponding to the number of the sodium atom in the chemical formula, the zeolite is assumed to have holes of 12 times the Avogadro number per a mole. Hence, the amount of cesium corresponding to $\Delta = 1$ is 12 moles for a mole of the zeolite. The ratio of cesium to the total alkali ions (sodium and cesium) in zeolite increases obviously, as is shown in Fig. 3, according to the curve based on Eq. (8) when $\varepsilon = 1.22$ is assumed. In this rough comparison, the present model can well reproduce the real process of the replacement.

[1] A. Fujisawa, H. Iguchi, S. Lee, Y. Hamada, Rev. Sci. Instrum. **68** 3393(1997).

FIG. 1: Change of alkali fractions in zeolite through substitution reactions. Four cases are shown with difference in parameters ε and Δ ; i) $\varepsilon = -0.5$, $\Delta = 0.5$, ii) $\varepsilon = 0.5$, $\Delta = 1.0$, iii) $\varepsilon = 0.5$, $\Delta = 0.5$, and iv) $\varepsilon = 1.0$, $\Delta = 0.5$.

FIG. 2: Efficiency of alkali replacement in time and cost for three cases. The necessary times of replacement processes and the total amount of the alkali compound are shown as a function of the amount of compound for each process. (a) The case of $\varepsilon = -0.2$, (b) $\varepsilon = 0.5$ and (c) $\varepsilon = 1.5$. The lines with circles represent the necessary times of substitution.

FIG. 3: A comparison between the model and an experiment. The circles represent the fraction of cesium to the total alkali ions (sodium and cesium). The ratios are estimated from the measurements of atomic ingredients by using scanning electron microscopy. The line shows the curve based on Eq. (8) with the parameter $\varepsilon = 1.22$.

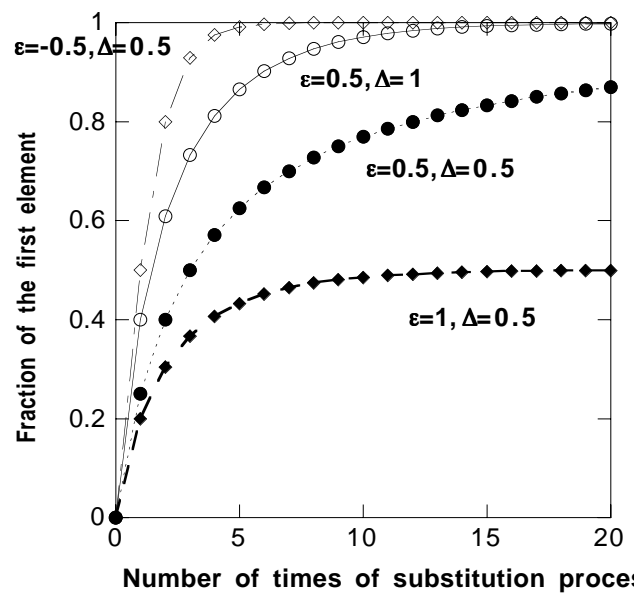


Figure 1 A. Fujisawa et al.

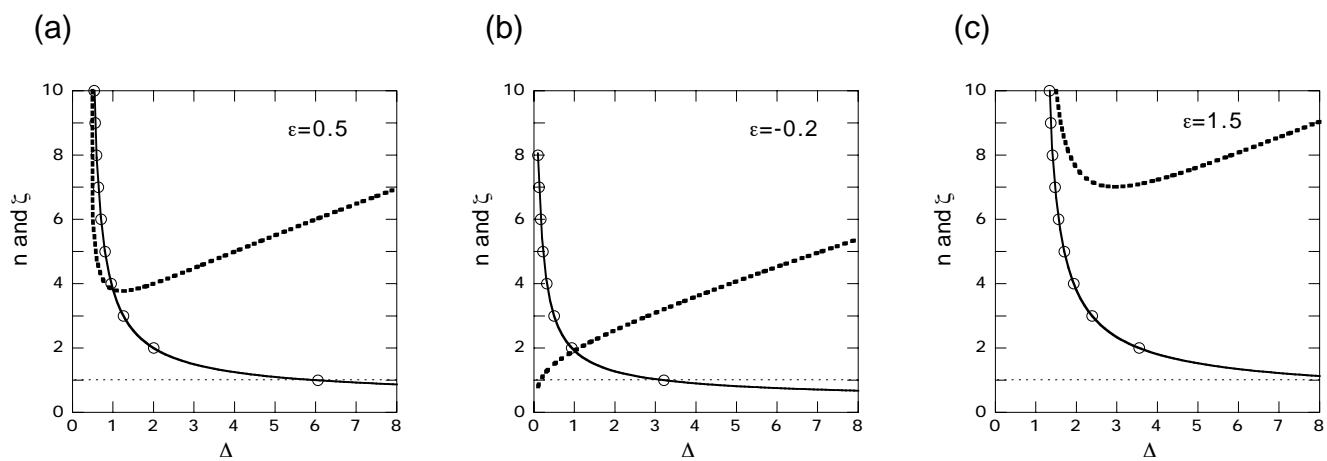


Figure 2 A. Fujisawa et al.

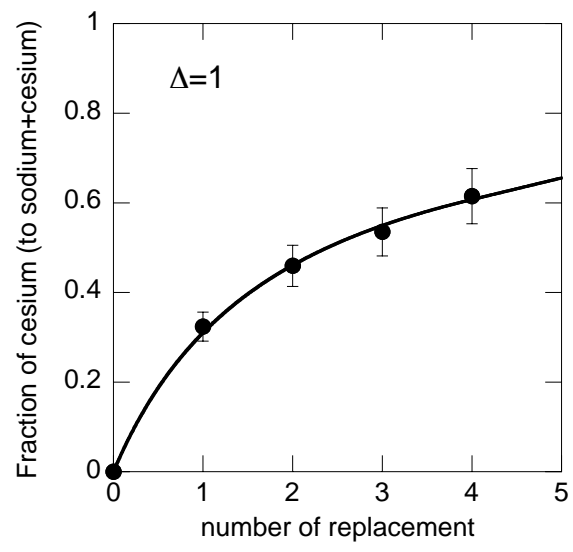


Figure 3 A. Fujisawa et al.