https://doi.org/10.46813/2026-161-023

UDC 54.058+621.039.341

ON EVAPORATION REFINING OF A SUBSTANCE CONTAINING ISOTOPES

A.I. Kravchenko
National Science Center "Kharkiv Institute of Physics and Technology", Kharkiv, Ukraine
E-mail: krwchnko@gmail.com

Attention is drawn to the fact that sublimation or distillation refining of a substance with monoisotopic base and with impurities in the form of isotopes of the same chemical element occurs with different efficiency for these impurities due to the dependence of not only the separation factor but also the Peclet diffusion number on the atomic mass of the isotope. The possibility of cases in which one of these isotopes is a low volatile impurity and the other is a highly volatile impurity. In the case where the base of a substance consists of isotopes and contains one monoisotopic impurity, then the refining of such a substance is characterized by the separation coefficient and the Peclet number, which depend on the atomic masses and concentrations of all components and change during the evaporation process. It is noted that the presence of isotopes in a substance should also be reflected in the results of its crystallization refining.

INTRODUCTION

Evaporative refining (sublimation and distillation) are among the main methods for obtaining high-purity substances [1–3], and recently, in connection with the interest in isotopic engineering, substances consisting of isotopes or containing isotopes have begun to be included in the list of substances being refined [4–9].

There is a method for calculating evaporative refining (primarily sublimation: in the absence of convective and/or mechanical mixing of the evaporating material) – with a separation factor β_0 and a Peclet diffusion number

$$Pe = X\frac{v}{D},$$

as two process parameters (v is the linear speed of evaporation surface; D is the diffusion coefficient of impurity; X is the dimensional factor of the evaporated material), with a strong dependence of Pe on the process temperature [10–13]. Examples of the dependence of the condensate purity on the degree of distillation g for different values of β_0 and Pe are shown in Figure [13] (C_0 is the initial concentration of impurity, and C is the average concentration of impurity in condensate; $Pe = 0$, if the mixing is ideal).

Using the Langmuir equation, it was shown that the separation factor (as the ratio of the concentrations of the impurity in the vapor and the evaporated material near the evaporation surface) can be represented as follows [2, 14, 15]:

$$\beta_0 = \frac{\gamma_i p_i}{\gamma p}\left(\frac{M}{M_i}\right)^{1/2},$$

where p_i and p are vapor pressures of pure components at process temperature; γ_i and γ are the activity coefficients of components and M_i and M are the atomic masses of the impurity and base, respectively. At the same time, a peculiarity of the evaporation of a substance consisting of two isotopes with atomic masses M_1 and M_2 was noted: the process occurs at $\gamma_i = \gamma = 1$ and $p_i = p$ with a temperature-independent separation

$$\beta = (M_2/M_1)^{1/2},$$

i.e., it was noted that there is a fundamental possibility a separation of isotopes in evaporative processes [14, 15]. (According to reference data on the values f atomic masses of isotopes, the ratio of the smaller mass to the larger has a value from 0.5 (for protium and deuterium; $\beta = 0.71$) and 0.85 (for lithium isotopes; $\beta = 0.92$) to 0.95 and even closer to 1 for isotopes of many other elements with $\beta = 0.97$ or closer to 1).

The aim of the work was to consider the evaporative refining of a material containing isotopes, taking into account the diffusion of impurities in the evaporated material.

ANALYSIS OF EVAPORATION OF MATERIAL CONTAINING ISOTOPES

If a substance contains two impurities in the form of isotopes with atomic masses M_1 and M_2 of the same chemical element, and the base of the substance has an atomic mass M, then the refining of such a substance is characterized by separation factors $\beta_{01} = (\gamma_i p_i/\gamma p)(M/M_1)^{1/2}$ and $\beta_{02} = (\gamma_i p_i/\gamma p)(M/M_2)^{1/2}$, where p_i and p are vapor pressures, and γ_i, and γ are activity coefficients of the impurity and base, respectively (with a simple ratio $\beta_{01}/\beta_{02} = (M_2/M_1)^{1/2}$), - and by the Peclet numbers $Pe_1 = Xv/D_1$ and $Pe_2 = Xv/D_2$, where D_1 and D_2 are the diffusion coefficients of impurities. In the general case, $D_1 \neq D_2$, since $M_1 \neq M_2$.

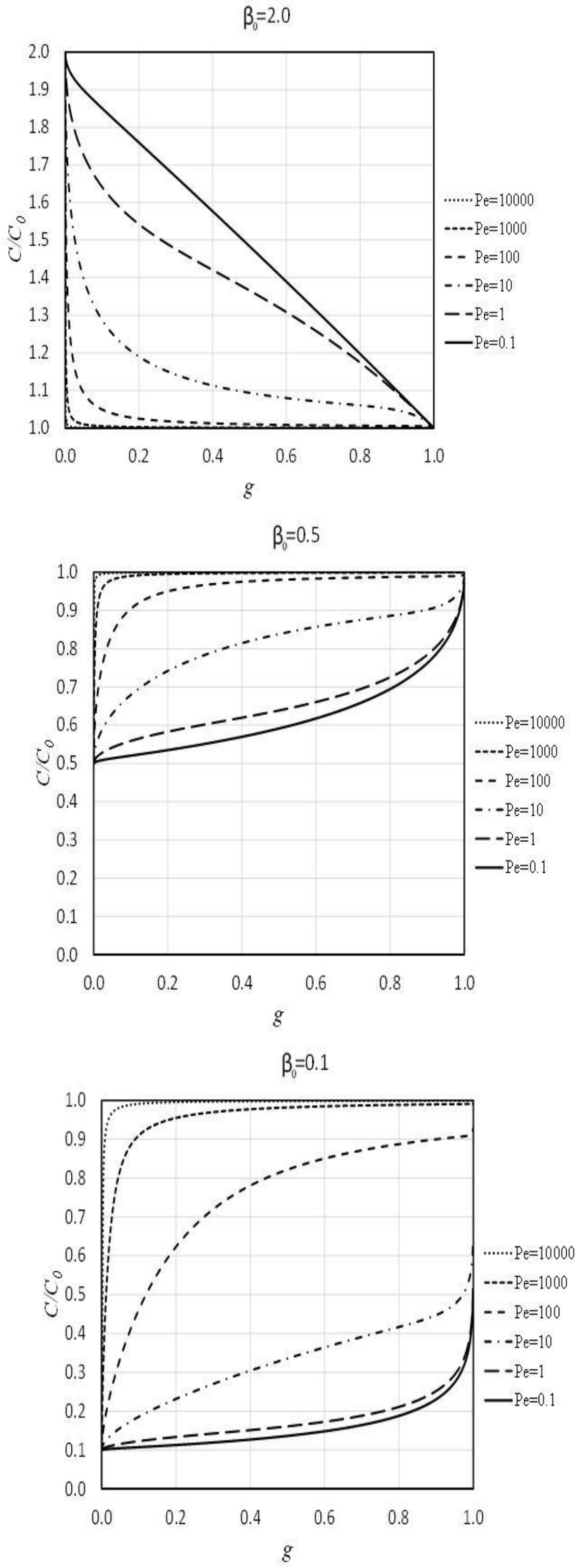
If a base of substance consists of isotopes with atomic masses M_1 and M_2 of the same chemical element (with concentrations C_1 and C_2 correspondently) and contains impurities with atomic mass M_i, then the substance has an average atomic mass

$$M^* = \frac{C_1 M_1 + C_2 M_2}{M_1 + M_2},$$

which determines the separation coefficient of the base and impurity

$$\beta_0 = (\gamma_i p_i/\gamma p)(M^*/M_i)^{1/2}$$

and Pe is determined by M_1, M_2, M_i, and by concentrations of all components.



Dependence of C/C_0 on g at different β_0 and Pe in a evaporation refining with a flat and constant evaporation surface

RESULTS AND DISCUSSION

Thus, due to the inequalities $\beta_{01} \neq \beta_{02}$ and $Pe_1 \neq Pe_2$, evaporation refining of a substance occurs with different efficiency for the impurities in question.

It is known that there are differences in the diffusion coefficients of isotopes of the same chemical element. Usually $D_1 < D_2$, if $M_1 > M_2$. Thus, at room temperature, for ^{12}C and ^{14}C in carbon materials, as well as for ^{28}Si and ^{29}Si in silicon materials, $D_1/D_2 = 1.2$ (according to data from ChatGPT). Then Pe_2/Pe_1 has the same value 1.2 for the same pairs of isotopes – while the modulus of deviation of $(M/M_1)^{1/2}$ or $(M/M_2)^{1/2}$ from 1 in many cases has a value ~ 0.01. The value of D_1/D_2 (and accordingly Pe_2/Pe_1) increases with increasing temperature [11, 12].

If $M_1 < M_2 < M$, then $\beta_{02} < \beta_{01} < 1$ and $Pe_2 < Pe_1$ (Figure. 1). If $M_1 < M < M_2$, then $\beta_{01} < 1$ and $\beta_{02} > 1$, i.e. the substance contains both a low-volatile impurity and a highly volatile impurity (see [16]).

If the base of a substance consists of isotopes and there is an impurity, then M^* and, as a consequence, β_0 (as well as Pe) change during the evaporation process.

A remark can also be made about crystallization refining. The effective separation coefficient in these processes is determined by the Burton-Prim-Slichter equation [1–3], which contains the diffusion coefficient D of impurity in the refined substance. And since D of an isotopic impurity depends on its atomic mass, the presence of isotopes in the refined substance should be reflected in the result of crystallization refining.

CONCLUSIONS

Taking into account two parameters (β_0 and the Peclet diffusion number), the evaporative refining of some substances containing isotopes is considered and the following features of their refining are revealed.

If a substance has a monoisotopic base and contains two impurities in the form of isotopes of the same chemical element, then the refining of such a substance occurs with different efficiency for these impurities due to the dependence of not only the separation coefficient, but also the Peclet diffusion number on the atomic mass of the isotope. It is possible that one of these isotopes is a volatile impurity, and the other is a non-volatile one.

If the base of the substance consists of isotopes and contains one monoisotopic impurity, then the refining of such a substance is characterized by the separation coefficient and the Peclet number, which depends on the atomic masses and concentrations of all components and change during evaporation.

REFERENCES

1. C.J. King. *Separation processes* / Second edition. N.Y.: “Dover Publication”, 2013.

2. G.G. Devyatykh, Yu.E. Yelliev. *Glubokaya ochistka veshchestv.* M.: “Vysshaya Shkola”, 1990 (in Russian).

3. Yu.I. Dytnerskii. *Protsessy i apparaty khemi-cheskoi tekhnologii:* Uchebnik dlya vuzov / Ed. 2. In two books. P. 2. Mass exchange processes and apparatus). M.: “Himiya”, 1995 (in Russian).

4. V.G. Plekhanov. Isotope engineering // *Phys. Usp.*, 2000, v. 43, N 11, p. 1147-1154.

5. A.D. Bulanov, A.N. Moiseev, O.Yu. Troshin, et al. Fine Purification of Monoisotopic Silanes $^{28}SiH_4$, $^{29}SiH_4$, and $^{30}SiH_4$ via Distillation // *Inorganic Materials.* 2004, v. 40, N 6, p. 555-560.

6. A.V. Adamchik, A.D. Bulanov, A.N. Sennikov, et al. Ultrapurification of ^{76}Ge-enriched GeH_4 by

distillation // *Inorganic Materials.* 2004, v. 47, N 7, p. 694-700.

7. R.S. Boiko, V.D. Virich, F.A. Danevich, et al. Ultrapurification of archaeological lead // *Inorganic Materials.* 2011, v. 47, N 6, p. 672-676.

8. G.P. Kovtun, A.P. Shcherban', D.A. Solopikhin, et al. Production of radiopure natural and isotopically enriched cadmium and zinc for low background scintillators // *Functional Materials.* 2011, v. 18, N 1, p. 121-127.

9. M.V. Sukhanov, T.I. Storozheva, I.I. Evdokimov, et al. Fine purification of monoisotopic ^{32}S and ^{34}S // *Inorganic Materials.* 2017, v. 53, N 2, p. 142-147.

10. A.I. Zhukov, A.I. Kravchenko. Calculation of sublimation with taking into account of diffusion of impurity // *Inorganic materials.* 2017, v. 53, N 6, p. 648-653; DOI: 10.1134/S0020168517060164

11. A.I. Kravchenko, A.I. Zhukov. Temperature dependence of the Peclet diffusion number in the processes of sublimation of some simple substances // *Inorganic materials.* 2021, v. 57, N 7, p. 753-759; DOI: 10.1134/S0020168521070104

12. A.I. Kravchenko, A.I. Zhukov, O.A. Datsenko. Temperature dependences of the Peclet number in sublimation processes of simple substances *// Problems of Atomic Science and Technology*. 2022, N 1, p. 13-16; https://doi.org/10.46813/2022-137-13

13. A.I. Zhukov, A.I. Kravchenko. Calculation of distribution of impurities in solidified condensate into account of Peclet number // *Problems of Atomic Science and Technology.* 2024, N 1, p. 25-28; https://doi.org/10.46813/2024-149-025

14. V.A. Pazukhin, A.Ya. Fisher. *Razdelenie i rafinirovanie metallov v vakuume.* M.: "Metallurgiya", 1969 (in Russian).

15. V.E. Ivanov, I.I. Papirov, G.F. Tikhinskii, V.V. Amonenko. *Chistye i sverkhchistye metally (poluchenie metodom distillyatsii v vakuume).* M.: "Metallurgiya", 1965 (in Russian).

16. A.I. Kravchenko. Calculation of distillation refining of a substance containing low and highly volatile impurities // *Inorganic materials*. 2018, v. 54, N 5, p. 501-503; DOI: 10.1134/S0020168518050087

Article received 16.10.2025

ПРО ВИПАРНЕ РАФІНУВАННЯ РЕЧОВИНИ, ЯКА МІСТИТЬ ІЗОТОПИ

О.І. Кравченко

Звернуто увагу на те, що сублімаційне або дистиляційне рафінування речовини з моноізотопною основою та з домішками у вигляді ізотопів одного і того ж хімічного елемента відбувається з різною ефективністю для цих домішок – внаслідок залежності не тільки коефіцієнта поділу, але і дифузійного числа Пекле від атомної маси ізотопу. Відзначено можливість випадків, у яких один із цих ізотопів є легколетючою домішкою, а інший – важколетючою. У разі, коли основа речовини складається з ізотопів і містить одну моноізотопну домішку, рафінування такої речовини характеризується коефіцієнтом поділу та числом Пекле, які залежать від атомних мас та концентрацій усіх компонентів та змінюються у випарному процесі. Зазначено, що присутність ізотопів у речовині має відбиватися також і на результатах його кристалізаційного рафінування.