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Driving Force of Diffusion of Coexisting Isotopes

Takao Yamamoto

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Migration of a chemical species in a material in which other isotope(s) of the species coexists and influences its state was investigated on the basis of the diffusion theory. It is shown that a driving force term is obtained by differentiating the chemical potential of the species by taking account of its dependence on the other isotopes. The formulation obtained was applied to interpreting tritium uphill migration observed in a neutron generating target bombarded by a deuterium beam.

This paper treats migration of a chemical species in a material in which another isotope of the species coexists influencing its state. Such migration occurs in a neutron generator target containing tritium bombarded by a deuterium ion beam to stimulate nuclear reactions for neutron production\(^1\)\(^-\)\(^3\). It was found that tritium atoms, initially distributed homogeneously in a titanium target migrate out of a region around the projected range of the incident ions causing the decrease of neutron yield and the shift of the energy spectrum of neutrons with increasing the beam fluence. A similar migration out of the region was studied as a component of fuel recycle processes anticipated to occur on the first wall of a thermonuclear fusion device, by investigating transport, across the surface and in a near-surface region, of hydrogen isotopes induced by the irradiation of the energetic particles\(^4\). Amounts of migration losses in the region, in both the examples, were far larger than either nuclear reactions or kinetic processes like collisions can account for. The author and his coworkers reported experimental data on the tritium targets and pointed out that this “uphill” migration could be ascribed to the diffusion induced by deuterium accumulation\(^5\)\(^-\)\(^6\). In this paper, the author derives, on the basis of the diffusion theory, a general formulation including a term driving the migration of a species out of the region when another isotopes pile up.

On the basis of the diffusion theory, the diffusion flux \(J_i\) of a species \(i\) is given by

\[
J_i = -N_i B_i \nabla \mu_i,
\]

where \(\nabla \mu_i\) is the gradient of the chemical potential, \(B_i\) the mobility and \(N_i\) the concentration of the species \(i\). The chemical potential \(\mu_i\) is expressed as

\[
\mu_i = \mu_i^0 + kT (\ln N_i + \ln \gamma_i),
\]

where \(\mu_i^0\) is the value of \(\mu_i\) at a standard state, \(\gamma_i\) the activity coefficient, \(k\) and \(T\) the Boltzmann constant and temperature, respectively. The term with \(\ln N_i\) is involved exclusively with the configurational mixing entropy, which does not take account of any interaction of the species \(i\) with other species. Actual interactions with the host lattice and other isotope species are all reflected in the other term through the value of \(\gamma_i\). These interactions mainly originate chemically and appear as caloric and volumetric changes, which generally cause dependencies of \(\gamma_i\) not only on \(N_i\) but also on \(N_j\) (\(j \neq i\)). In evaluating the gradient on the right-hand side of Eq. (1), therefore, the dependencies should be taken into account. The Eq. (1) should then be developed as

\[
J_i = -N_i B_i \sum_{j \neq i} \left( \frac{\partial \mu_j}{\partial N_j} \right) \nabla N_j,
\]

This is the general formulation for diffusion flux of species \(i\) driven by the concentration gradient of all species influencing its chemical potential. When this summation is neglected, in other words, when the
dependencies of $\mu_i$ on $N_i$ are not taken into account, this equation directly gives an expression of Fick's first law postulating a diffusion flux along the concentration gradient merely of itself and never accounting for the uphill migration now considered. The additional terms express the influence of coexisting species.

Assuming chemical equivalence amongst isotopes, all activity coefficients should be equal and can be substituted by that of a species, for instance, species $i$. Equation (4) is then modified as

$$J_i = -D_i^* \nabla N_i - \sum_{j=1}^s \frac{N_j B_i}{N_i B_j} (D_j^* - D_i) \nabla N_j.$$  (5)

Note $(D_i^* - D_i) = D_i (\partial \ln \gamma_i / \partial \ln N_i)$, indicating that only a term in $F_{\alpha i}$ due to chemical interactions of species $j$ works in driving the diffusion of species $i$.

Now this equation is applied to interpreting the tritium uphill migration in a neutron generating target bombarded by a deuteron beam. Subscripts $t$ and $d$ are assigned to tritium and deuterium, respectively, and one-dimensional coordinate $z$ along the depth are adopted. Tritium diffusion flux $J_t$ given by Eq. (4) is

$$J_t = -D_t^* \frac{dN_t}{dz} - D_t \frac{N_t}{N_d} \left( \frac{\partial \ln \gamma_t}{\partial \ln N_d} \right) \frac{dN_d}{dz},$$  (6)

where $\gamma$ is an activity coefficient assumed to be universal for tritium and deuterium, $\gamma = \gamma_t (N_t, N_d) = \gamma_d (N_t, N_d)$. Two major factors in the second terms are separately discussed. The factor $(dN_d/dz)$ is estimated from the range distribution of implanted deuterium atoms around the projected range of the beam, which should have been broadened because of deuterium diffusion. It is illustrated in Fig. 1. Another factor $(\partial \ln \gamma / \partial \ln N_d)N_t$ is estimated by referring to $F_{\alpha t}N_t$ equilibrium isotherms, where $F_{\alpha t}$ is the equilibrium hydrogen pressure over a phase with hydrogen content of $N_t$. The ideal gas approximation applied to the gas-solid equilibrium leads to a derivative corresponding to the factor now considered,

$$\frac{\partial \ln \gamma_t}{\partial \ln N_t} = \frac{1}{2} \frac{\partial \ln F_{\alpha t}}{\partial \ln N_t} - 1,$$  (7)

which generally increases with $N_t$ and jumps up in the vicinity of an ultimate hydrogen content achieved in the phase, i.e., $H/\text{Ti}=1.89$. Ion bombardment easily attains an extraordinarily high hydrogen concentration near and sometimes over the terminal hydrogen content to make a transient non-equilibrium state. The uphill migration is due to the second term of the Eq. (6) overcoming the first term, since $(dN_d/dz)$ given by the ion implantation is far larger than $(dN_t/dz)$ and their signs are opposite each other once tritium migration has occurred.

In summary, a formulation for the diffusion driving force of a chemical species induced by the concentration gradient not only of itself but also of other coexisting isotopes has been derived by starting from a principal equation of the diffusion theory, $J_i = -N_i B_i \nabla \mu_i$, and an assumption of chemical equivalence amongst isotopes by referring neither to any microscopic diffusion nor interaction mechanisms.

References