STUDIES ON DIFFUSION

Part I. Diffusion of Radioactive Matter Through Plates

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ABSTRACT

As a part of an attempt to develop new methods of measurement of diffusion constant of metals and non-metals into minerals using radioactivity techniques, expressions are derived to relate the processes of diffusion and the emission of alpha-particles under three sets of possible experimental conditions. The first case pertains to a plate whose thickness is equal to the characteristic range of the alpha-rays of the radionuclide used. Under the second case is considered a plate thicker than the characteristic range of the alpha-rays in the plate. The third deals with cases wherein the radioactive matter used emits alpha-rays with different ranges less than the thickness of the plate.

INTRODUCTION

THE problem of diffusion of metals in the solid state has been extensively studied during the last three decades (Mehl, 1936), because of its importance in the industrial operations like the preparation of special alloys and the development of nitriding, chromizing and calorizing, sherardizing and siliconizing and the formation of bi-metal strip and veneering metals. Diffusion data are also essential for studies on crystal physics and the elucidation of phenomena such as annealing, age hardening, plasticity and order-disorder transformations in alloys. Our present state of knowledge on the subject of solid diffusion has recently been reviewed by Barrer (1951).

The voluminous literature on metal diffusion stands in marked contrast to the virtual absence of any systematic work on the measurement of diffusion constants of metals and non-metals in minerals and rocks. The discovery of radioactive isotopes gave a great fillip to self-diffusion studies on metals (Hevesy *et al.*, 1913, 1925, 1929 *a*, *b*, 1932, 1933; Groh and Hevesy, 1920; McKay, 1938; Steigman *et al.*, 1939). The pioneering work of Jensen (1952) on the solid diffusion of radioactive sodium (Na^{22}) in perthite shows tremendous potentialities for further researches on the subject.

The principles of diffusion have been discussed in detail in an earlier communication (Aswathanarayana, 1954). As a part of the long-range programme of theoretical and laboratory investigations on the measurement of diffusion constants, an attempt is made now to derive expressions relating the processes of diffusion and emission of alpha-particles.

GENERAL STATEMENT OF THE PROBLEM

Let A B C D-A′ B′ C′ D′ with dimensions a, b, c (a being the thickness) represent a thin, uniform section of a mineral. A uniform, thin coating of an alpha-emitting radio-nuclide is given to the face A B C D. Let v be the concentration of radioactive matter on that surface. Simultaneously with the above operation, an α-particle detecting device—nuclear emulsion plate suitable for alpha-track radiography—is kept in contact with the face A′ B′ C′ D′ of the thin section. Each radioactive atom undergoing decay acts as a centre of a sphere of radius equal to the range of alpha radiation in the medium. When the sphere intersects the plane of the emulsion, it leaves behind its imprint in the form of an image (Yagoda, 1949). The number of alpha-particles emerging from the lower face A′ B′ C′ D′ and getting recorded on the nuclear emulsion plate is a function of (1) the concentration (v) of radioactive matter on the face A B C D, which determines the rate of production of alpha-particles, (2) the characteristic range (ρ) of the alpha-particles of the radioactive nuclide in the mineral and (3) the thickness of the plate (a). With time, a part of the material of the radioactive nuclide diffuses into the mineral and this results in a larger number of alpha-rays emerging from the face A′ B′ C′ D′ of the mineral section and getting recorded on the plate. Thus from a knowledge of (1) the range (ρ) of alpha-particles of the radioactive nuclide, (2) the concentration (v) of radioactive matter on the plate and (3) the number of alpha-particles (N) recorded on the plate after a time T, it is possible to determine the diffusion constant (D).

MATHEMATICAL TREATMENT

The following assumptions are made in this context:—

(i) The diffusion of radioactive matter in a mineral is governed by laws analogous to laws of conduction of heat in a solid and (ii) the mineral exhibits diffusion isotropy.

The concentration $v\ (x, y, z, t)$ is governed by

$$\frac{\partial v}{\partial t} = D\nabla^2 v\,.$$

The experimental conditions may be taken as equivalent to the diffusion of matter in a rectangular slab of thicknesses a, b, c, where a is the distance between the plane on which radioactive matter has been spread and the opposite plane (which is in contact with the nuclear emulsion plate).

The boundary conditions are:

$$\begin{array}{lll} \text{at} & x = 0 & v = v_1 \\ & \left.\begin{array}{l} x = a \\ y = 0 \\ y = b \\ z = 0 \\ z = c \end{array}\right\} & v = 0 \\ \text{at} & t = 0 & v = 0. \end{array}$$

Then v can be written as the sum of two terms U and W (say)—U being a function of x, y, z alone and W, a function of x, y, z, t (Carslaw and Jaeger, 1947, pp. 18–19, 165).

Concentration $v = v\ (x, y, z, t)$

$$v\ (x, y, z, t) = \frac{16 v_1}{\pi^2} \sum_{p=0}^{\infty} \sum_{q=0}^{\infty} \frac{\sinh \alpha\,(a - x) \sin \frac{(2p+1)\,\pi y}{b} \sin \frac{(2q+1)\,\pi z}{c}}{(2p+1)\,(2q+1) \sinh \alpha a}$$

$$- \frac{32 v_1}{\pi} \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} \sum_{l=1}^{\infty}{}' \frac{l e^{-D\alpha^2_{l,m,n} t} \sin \frac{l\pi x}{a} \sin \frac{m\pi y}{b} \sin \frac{n\pi z}{c}}{mn \left\{\pi^2 l^2 + a^2 \left(\frac{m^2\pi^2}{b^2} + \frac{n^2\pi^2}{c^2}\right)\right\} \sinh a \sqrt{\frac{m^2\pi^2}{b^2} + \frac{n^2\pi^2}{c^2}}}$$

where

$$\alpha^2 = \frac{(2p+1)\,\pi^2}{b^2} + \frac{(2q+1)\,\pi^2}{c^2}$$

$$\alpha^2_{l,m,n} = \pi^2 \left(\frac{l^2}{a^2} + \frac{m^2}{b^2} + \frac{n^2}{c^2}\right).$$

Case I.—The following experimental conditions are assumed in this case: (i) the thickness (a) of the slab is equal to the range (ρ) of the α-particles of the radio-nuclide, (ii) the α-rays are mono-energetic or in other words, they all have a characteristic specific range (ρ) in the mineral, (iii) the radioactive matter has been spread on the mineral surface uniformly and (iv) the nuclear emulsion plate is in contact with the lower side of the mineral plate simultaneously with the spreading of radioactive matter on the upper surface.

The number of α-particles emitted between the time instants 0 and T is given by

$$N = \int_0^T dt \int_0^c dz \int_0^b dy \int_0^a v\lambda dx$$

where λ is the fraction of radioactive matter that disintegrates per second.

Putting

$$A = \frac{64\, v_1 bc}{\pi^4 \alpha_0 \sinh(\alpha_0 a)} [\cosh(\alpha_0 a) - 1],$$

$$B = \frac{256\, v_1 a b^3 c^3}{\pi^6 \{b^2c^2 + a^2(b^2 + c^2)\} \sinh \frac{\pi a}{bc}\sqrt{b^2 + c^2}},$$

$$\alpha_0 = \pi\sqrt{\frac{1}{b^2} + \frac{1}{c^2}},$$

$$\alpha = \pi\sqrt{\frac{1}{a^2} + \frac{1}{b^2} + \frac{1}{c^2}},$$

we get

$$\frac{N}{\lambda} = (A - B)\,T + \frac{B\alpha^2 T^2}{2} D \qquad \text{(approximately)},$$

taking into account the terms in v with $p = q = 0$, and $l = m = n = 1$. From this equation D can be calculated.

Case II.—In the second case, the following experimental conditions are assumed: (i) The thickness (a) of the plate is larger than the range (ρ) of the alpha-particles of the radio-nuclide in the mineral, (ii) the alpha-particles are mono-energetic, (iii) spreading of the radioactive matter on the mineral face A B C D is uniform and (iv) the nuclear emulsion plate is in position by the time radioactive matter reached $x = a - \rho$ plane.

$$\frac{N}{\lambda} = \int_{\tau}^{T} dt \int_0^c dz \int_0^b dy \int_{a-\rho}^{a} v dx$$

where τ is the time taken by the readioactive matter to reach the plane $x = a - \rho$. It is given by

$$D\tau = \frac{1}{a^2}\log_e \frac{\pi\left\{1 + a^2\left(\frac{1}{b^2} + \frac{1}{c^2}\right)\right\} \sinh \pi a \sqrt{\frac{1}{b^2} + \frac{1}{c^2}} \sinh (\alpha_0 \rho)}{2 \sinh (\alpha_0 a) \sin \pi \left(1 - \frac{\rho}{a}\right)}$$

approximately.

Putting

$$A' = \frac{64 v_1 bc}{\pi^4 \sinh (\alpha_0 a)} \left[\cosh (\alpha_0 \rho) - 1\right],$$

$$\beta = \frac{B}{4}\left\{1 + \cos \pi\left(1 - \frac{\rho}{a}\right)\right\} \alpha^2 (T^2 - \tau^2),$$

$$\gamma = A' - \frac{B}{2}\left\{1 + \cos \pi\left(1 - \frac{\rho}{a}\right)\right\}$$

(β, α, α_0 the same as above in Case I), we get

$$\frac{N}{\lambda} = \gamma (T - \tau) + \beta D \qquad \text{(approximately)},$$

whence D can be calculated.

Case III.—This case is similar to the second case but for the assumption that the alpha-particles emitted by the radioactive matter are not mono-energetic, *i.e.*, they have got several ranges as different from a single range considered under the second case.

Let $\rho, \rho', \rho'' \ldots . \rho^{(n)}$ be the ranges of α-particles emitted by the radioactive matter. Let $\rho < \rho' < \rho'' \ldots . \rho^{(n)} < a$. Let $\tau^{(r)}$ be the time taken by the radioactive matter to penetrate to such depth that radiations from it of range $\rho^{(r)}$ can influence the photographic plate. Let T be the time that has elapsed upto the removal of the photographic plate.

Then

$$\frac{N}{\lambda} = \int_{\tau}^{T} dt \int_0^c dz \int_0^b dy \int_{a-\rho}^{a} v dx$$

$$+ \int_{\tau'}^{T} dt \int_0^c dz \int_0^b dy \int_{a-\rho'}^{a} v dx$$

$$+ \ldots\ldots\ldots\ldots\ldots\ldots$$

$$+ \int_{\tau^{(n)}}^{T} dt \int_0^c dz \int_0^b dy \int_{a-\rho^{(n)}}^{a} v dx,$$

Let the values of β and γ in Case II be termed as $\beta^{(s)}$ and $\gamma^{(s)}$ when $\rho = \rho^{(S)}$ and $\tau = \tau^{(S)}$. Then

$$\frac{N}{\lambda} = \sum_s \gamma_s (T - \tau_s) + D \sum_s \beta_s \quad \text{(approximately).}$$

From this formula D can be calculated.

Further investigations are proceeding.

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