

A NOTE ON SELF-DIFFUSION IN DOUGH CONTAINING Br⁸²-LABELED BROMATE¹

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Diffusion of mass refers to the dissipation of a concentration gradient by molecular transfer with no over-all mass flow caused by external forces. Self-diffusion differs from ordinary diffusion in that its occurrence does not require the existence of an over-all chemical concentration gradient. The method used for studies of self-diffusion generally involves the observation of the net translational motion of a given amount of substance labeled with a known isotope of one of the constituent elements in an otherwise chemically uniform medium. The self-diffusion rate thus gives the nearest available approach to the observation of the magnitude of actual molecular motion in the gross sense (6). As an attempt to obtain some indications of the gross mobility of bromate and its decomposition product, bromide, in doughs, a study on self-diffusion was made with dough initially containing 30 p.p.m. potassium bromate.

Two pieces of dough were made up with 100 g. of flour and 60

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ml. of distilled water by mixing for 3 minutes in a Hobart mixer. One contained 30 p.p.m. ordinary potassium bromate and the other contained the same amount of Br⁸²-labeled potassium bromate (7) with initial specific activity of about 5×10^5 c.p.m. per mg. The doughs were joined together, being separated only by a moist sheet of Whatman No. 4 filter paper which made possible a clean division at the boundary at the end of the diffusion period. Diffusion was allowed to proceed for 4 hours and 20 minutes in a cabinet maintained at 30°C. and 80% relative humidity. The doughs were then frozen and separated at the boundary. Cores of dough, 23 mm. in diameter, were cut at right angles to the boundary from each of the two pieces and then sliced into samples of approximately 2-mm. thickness. The radioactivity of each sample was measured and the actual sample thickness was estimated from the sample weight with the assumption that dough density was 1.2. Duplicate experiments were carried out and the results are given in Table I.

TABLE I
Br⁸² ACTIVITY FROM SELF-DIFFUSION IN DOUGH

SAMPLE ^a	NET ACTIVITY		SAMPLE THICKNESS		CUMULATIVE DISTANCE FROM BOUNDARY	
	Trial 1	Trial 2	Trial 1	Trial 2	Trial 1	Trial 2
	<i>cpm</i>	<i>cpm</i>	<i>mm</i>	<i>mm</i>	<i>mm</i>	<i>mm</i>
1a	74.6	87.6	2.0	2.7	2.0	2.7
2a	95.4	99.8	1.8	0.9	3.8	3.6
3a	112.2	116.2	2.1	1.8	5.9	5.4
4a	119.9	113.1	2.0	1.4	7.9	6.8
5a	121.8	125.7	2.2	1.8	10.1	8.6
6a		119.8		2.0		35
1b	36.1	37.0	1.8	1.4	1.8	1.4
2b	21.7	19.7	1.9	1.7	3.7	3.1
3b	10.2	7.3	2.3	1.5	6.0	4.6
4b	0.9	1.7	2.3	2.6	8.3	7.2
5b	0	0	2.3	1.6	10.6	8.8
6b		0.1		2.2		35

^a The "b" samples were derived from dough which initially contained nonradioactive bromate.

When sample activities were plotted against distance from the boundary, typical diffusion curves were obtained. The self-diffusion coefficient, *D*, was calculated from a solution of Fick's law given by Barrer (1) for the case of diffusion across a sharp boundary in an "infinite solid" where no appreciable amount of material diffuses from the far extremities of the system. The solution (1) takes the form of equation [1], where *C*₀ is the initial concentration, *C* is the concentration at distance *x*, and *t* is the time. The second term of equation [1] is the Gaussian error function; hence [1] may be written as equation [2].

$$C = \frac{C_0}{2} \left[1 + \frac{2}{\sqrt{\pi}} \int_0^{\frac{x}{2\sqrt{Dt}}} e^{-y^2} dy \right] \quad [1]$$

or

$$C = \frac{C_0}{2} [1 + \operatorname{erf}(y)] \quad [2]$$

Using the curves derived from the two sets of experimental data given in Table I, an average activity value, C , corresponding to a given distance from the boundary, x , can be obtained graphically. With these values of C and x , equations [1] and [2], and mathematical tables of error functions (4,5), the diffusion coefficient D can be evaluated. In this way, a number of D values corresponding to various distances x were calculated, giving rise to a mean value of $0.24 \text{ cm.}^2 \text{ day}^{-1}$ for D .

As a check on the mean value of $0.24 \text{ cm.}^2 \text{ day}^{-1}$ as a valid self-diffusion coefficient for the present data, the theoretical curve for this D value was calculated using equation [3] derived from expanding equation [1] as a series (1).

$$C = \frac{C_0}{2} \left[1 + \frac{2}{\sqrt{\pi}} \left\{ \frac{x}{2\sqrt{Dt}} - \frac{x^3}{3 \cdot 1! (2\sqrt{Dt})^3} + \frac{x^5}{5 \cdot 2! (2\sqrt{Dt})^5} - \frac{x^7}{7 \cdot 3! (2\sqrt{Dt})^7} + \dots \right\} \right] \quad [3]$$

This theoretical curve is shown in Fig. 1, together with the data from Table I being given as observed experimental points. It is evident from Fig. 1 that in the water-flour dough system, the move-

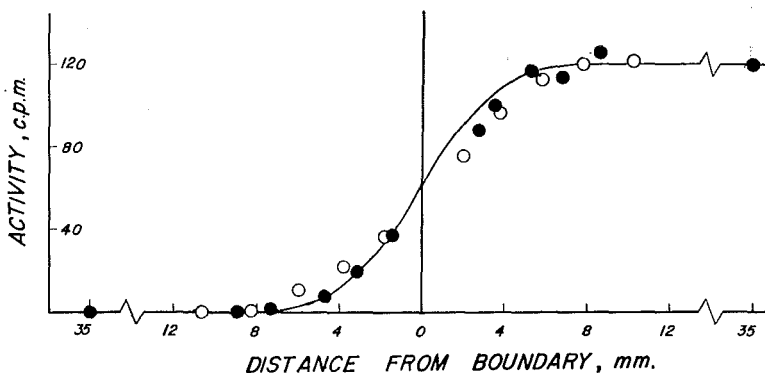


Fig. 1. Self-diffusion of Br^{82} in dough. The theoretical curve for D equals $0.24 \text{ cm.}^2 \text{ day}^{-1}$ is shown together with data from Table I as observed experimental points. Open circles are data from trial 1; solid circles from trial 2.

ment of Br^{82} -labeled ions, which would include both undecomposed bromate and the decomposition product, bromide, follows the laws of diffusion and fits reasonably well with a self-diffusion coefficient of $0.24 \text{ cm.}^2 \text{ day}^{-1}$.

The self-diffusion coefficients of bromate and bromide in the anion exchanger, Dowex-2, have been found to be, respectively, 4.55 and $3.87 \times 10^{-7} \text{ cm.}^2 \text{ sec.}^{-1}$ at 25°C . (8). The self-diffusion coefficient of bromide in 0.05 molar sodium chloride at 25°C . was reported as $1.62 \text{ cm.}^2 \text{ day}^{-1}$ (2). In cereal chemistry, from the properties of the resulting doughs when fermented under carbon dioxide, oxygen, or nitrogen, Sullivan and Richards (9) have postulated that the sponge is permeable to the gas above it. In an attempt to substantiate this postulate, Burtle and Sullivan (3) were able to show that radioactive carbon dioxide can diffuse back into the sponge even against the developing internal pressure of the dough. The present study on self-diffusion clearly demonstrated that, under the conditions used in these experiments, net movements of bromate and bromide in the dough are detectable within about 1 cm. on either side of the boundary. This finding points to a dynamic nature for the soluble constituents of dough which may merit further investigation.

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