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Metals, Ceramics, and Materials (UC-25)
TID 4500, December 15, 1960

UNITED STATES ATOMIC ENERGY COMMISSION

Research and Development Report

DIFFUSION IN CERAMIC SYSTEMS; A
SELECTED BIBLIOGRAPHY

by

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at
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DIFFUSION IN CERAMIC SYSTEMS; A SELECTED BIBLIOGRAPHY

Michael F. Berard

INTRODUCTION

This bibliography and set of tables are the result of an effort to bring together much of the literature pertaining to diffusion in ceramic systems.

In the arrangement of the bibliography, primary classification is by the type of diffusion system, i. e., Oxides, Silicates and Glasses, or Borides, Carbides and Graphite. Subclassification in each case is by the diffusion species. Many quotations have been taken directly from Ceramic Abstracts, Chemical Abstracts, Nuclear Science Abstracts, or from the original work. When this was not satisfactory, and an abstract was deemed necessary, one was supplied by the author. The Roman numerals that appear at the end of certain citations refer to the tables in which data from these sources have been tabulated.

Whenever possible, the data have been presented in tabular form according to the Arrhenius temperature dependence equation:

$$D = D_0 \exp(-Q/RT)$$

where the frequency factor, D_0 , is given in square centimeters per second, and the activation energy, Q , is given in kilogram-calories

per mole. In some cases only the diffusion coefficient, D , for a given centigrade temperature is tabulated. The numbers listed in the final column of the tables refer to the bibliography.

An alphabetical author index keyed to the bibliography is included at the end of the work to further facilitate its use.

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3. Crank, J., Mathematics of Diffusion, Clarendon Press, Oxford, 1956.
4. Fical, C., Structural defects and diffusion in solids. Ceramica 8, 49 (1953). Ceram. Abs. May 100j, 1954.

"The mechanism of diffusion and chemical reaction taking place in solids are discussed. The principal factors on which the chemical reactions depend are (1) the energy factor, (2) the type of moving particles (ions, molecules), (3) their mobility (diffusion), and (4) the system in which such particles move. Formulas are given for calculating the energy and velocity of diffusion with diagrams to explain the phenomena. "

5. Frechette, V. D., Solid-state reactions and ceramic technology. Proc. Intern. Symposium Reactivity of Solids, Gothenburg, 1952, Pt. 2, 683 (Publ. 1954). Chem. Abs. 48:11748g, 1954.

"Ceramic industrial experience offers a multitude of practical observations which might prove valuable suggestions to the scientific investigator of solid-state reactions, of diffusion, and of ion exchange. With this in mind, the various techniques are reviewed. "

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9. Gray, T. J., James, B. D. and Giess, E. A., Diffusion in ionic compounds. PB Rept. 127266. Ceram. Abs. Sept. 224g, 1961.

"Theories and methods of studying diffusion are discussed. A bibliography of 237 references is included. "

10. Houseman, D. H., Radioactive tracer techniques in refractories research. Refractories J. 34, 540 (1958). Ceram. Abs. April 102h, 1959.

"The two main sources of radioactive tracers are pile-produced isotopes and fission-produced isotopes. The properties of radioactive isotopes are discussed, and the technique of radioactivation is explained. The use of isotopes in the refractories research laboratory is reviewed under (1) diffusion studies; (2) determination of porosity of granular materials; (3) radiometric titration; and (4) study of mixing efficiency. . . "

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12. Jander, W. and Stamm, W., Internal structure of solid oxygen-containing compounds at high temperatures. III. Electrical conductivity, diffusibility and reactivity of spinels in the solid state. Z. anorg. allgem. Chem. 207, 289 (1932).
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14. Jost, W., Diffusion in Solids, Liquids, Gases, Academic Press Inc., New York, 1952.
15. Lommel, J., Literature survey on the diffusion of metals into oxides. Atomic Energy Commission Publication No. AD-75227, 1953 and AD-75225, 1954.

"A literature survey on oxidation and diffusion covering the period 1946 to early 1954. There are 494 references included.

16. Moore, W. J., Diffusion mechanisms and gaseous corrosion, in 3^c Colloque de Métallurgie sur la Corrosion (Seche et Aguese). Organise à Saclay les 29-30 juin et 1^{er} juillet 1959, Centre d'Etude Nucléaires (Saclay) and North Holland Publishing Company (Amsterdam), 1960. NSA 14:12866, 1960.

"A general survey is given of the various types of defect mechanisms that may be applicable in diffusion in solid oxides and sulfides at high temperatures. . . "

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18. Shockley, W., Hollomon, J. H., Maurer, R. and Seitz, F. (editors), Imperfections in Nearly Perfect Crystals, John Wiley and Sons, Inc., New York, 1952.
19. Wilder, D. R., Sintering mechanism as applied to refractory oxides. A bibliography, 1937-1960. Atomic Energy Commission Publication No. ISC-288, December, 1952, ISC-288 (Suppl.), January, 1956, ISC-288 (Suppl. II), January, 1960.

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Aluminum

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"Experimental measurements of the rate of shrinkage of pressed Al_2O_3 compacts, the neck growth between single-crystal Al_2O_3 spheres and plates, and the effect of particle size on neck growth between single-crystal Al_2O_3 spheres and plates are mutually consistent with the bulk diffusion and sintering model. The temperature dependence of the rate of shrinkage and neck growth in Al_2O_3 is characterized by an activation energy of 165 kcal. per mole. Apparent diffusion coefficients and temperature dependence calculated from the shrinkage of pressed compacts of Fe_2O_3 agree with measured diffusion coefficients for the diffusion of Fe in Fe_2O_3 ."

21. Fischer, W. A. and Hoffmann, A., Volume changes in the diffusion zone of oxide systems. Naturwissenschaften 41, 162 (1954). Ceram. Abs. Sept. 222a, 1960.

"It was found that the Kirkendall effect also exists with metal oxides. Pure Al_2O_3 in contact with $\text{FeO} \cdot \text{Al}_2\text{O}_3$ shows a contraction in the Al_2O_3 part of the diffusion zone after sintering at 1500°C . for 50 to 100 hours. The dark-colored part of this diffusion zone contained 1% Fe. The diffusion of the components takes place principally as ions. Evidently Al ions migrate faster than Fe ions."

22. Kuczynski, G. C., Abernethy, L. and Allan, J., Sintering mechanisms of aluminum oxide, in Kinetics of High Temperature Processes (No. 11, this bibliography).

Diffusion of aluminum in Al_2O_3 is studied by observing the growth of the neck occurring where single crystal spheres of Al_2O_3 are in contact. Samples annealed in oxygen, helium and wet hydrogen gave similar results, while those annealed in dry hydrogen gave much higher activation energies. I.

(See also references 34, 56.)

Barium

23. Cowan, G. A. and Orth, C. J., Diffusion of fission products at high temperatures from refractory matrices. Atomic Energy Commission Publication No. A/Conf. 15/P/613, June, 1958.

Measurements on the diffusion of arsenic, bromine, strontium, zirconium, ruthenium, rhodium, palladium, silver, cadmium, tin, antimony, tellurium, iodine, cesium, barium, cerium, neodymium and europium from graphite as well as the diffusion of arsenic, strontium, zirconium, molybdenum, palladium, silver, cadmium, antimony, tellurium, iodine and barium from MgO matrices.

It is concluded that where stable carbides can be formed, the fission products do not diffuse from graphite. Fission products with refractory oxides generally diffuse less rapidly through magnesia than through graphite.

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"... The ionic transfer exclusively concerns the Ba^{++} ion. During and after the exposure of the material in an electrostatic field, the mobility of the Ba^{++} ions is distinctly increased. . . Evidently, the electrostatic field orients the particles of BaTiO_3 and thus increases the diffusion constant. . . "

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"Diffusion coefficients were studied with radiotracers. The diffusion coefficient of barium at 1150° is of the order of 10^{-14} sq. cm./sec. and increases after exposure to a direct electric field. The transport number for barium is of the order of 10^{-4} between 850 and 1040°..." III.

26. Lindner, R., Self-diffusion in oxide systems. Z. Naturforsch. 10a, 1027 (1955).

A compilation of frequency factors and activation energies for self-diffusion in oxide systems. III, XV, XVII, XX, XXVIII, XXXII.

27. Redington, R. W., Diffusion of barium in barium oxide. Phys. Rev. 87, 1066 (1952).

A sectioning technique is used to measure the diffusion coefficient for barium diffusing into single crystals of barium oxide. Two different diffusional processes are indicated by the data. The ionic mobility and surface diffusion are also considered. III.

Beryllium

28. Austerman, S. B., Diffusion of beryllium in beryllium oxide. Atomic Energy Commission Publication No. NAA-SR-3170, December 15, 1958.

Diffusion of beryllium in polycrystalline BeO annealed in an argon atmosphere is measured by a sectioning technique. IV.

29. Austerman, S. B., Diffusion of beryllium in beryllium oxide. Part. II. Atomic Energy Commission Publication No. NAA-SR-5893, May 1, 1961.

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30. Chang, R., Creep in polycrystalline beryllium oxide at high temperatures and low stresses. Atomic Energy Commission Publication No. NAA-SR-2458, 1958. Chem. Abs. 53:7544i, 1959.

"Creep data for stresses of 54, 108, and 216 lb./sq. in. show a linear variation of creep rate with applied stress consistent with the stress-directed vacancy-diffusion theory. The activation energy for creep was calculated as 114,000 cal./mole on the basis of the vacancy diffusion concept. The activation energy for the diffusion of Be-7 in polycrystalline BeO appears to be equal to the activation energy for creep according to the vacancy diffusion concept..." IV.

Calcium

31. Hedvall, J. A., Brisi, C. and Lindner, R., Self-diffusion in monocalcium ferrite. Arkiv Kemi 4, 377 (1952). VI, XV.
32. Lindner, R., Use of radioisotopes for the study of self-diffusion in oxide systems. Atomic Energy Commission Publication No. A/Conf. 15/P/167, May 23, 1958.

Data is tabulated for cation self-diffusion in simple oxides, aluminates, ferrites, chromites, stannates, calcium silicates and lead silicates. Tabulation is also given for diffusion of rare gases in oxide systems. VI, XXIX.

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"... The self-diffusion constants, determined from partial ionic mobilities, agree fairly well with those determined by radioactive measurements and indicate exclusively calcium ion diffusion in CaO." VI, XXXII.

(See also references 105, 106.)

Chromium

34. Klyucharov, Ya. V. and Zubakov, S. M., Reactions between chromite and magnesite at high temperatures. Trudy Leningrad. Tekhnol. Inst. im. Lensovet 1955, No. 34, 32. Chem. Abs. 50:13393d, 1956.

"The oxide diffusion on contacts between grains... of chromite ore... and magnesite... at 1600° and higher brings about a disintegration of the chromite. The ore is impoverished in Fe_2O_3 and SiO_2 , which are exchanged by MgO . On the other hand, magnesite takes up Fe_2O_3 and SiO_2 , and Cr_2O_3 and Al_2O_3 migrate very little. Repeated thermal exposure to 1600° shows that the first diffusion of Fe_2O_3 is the most efficient; it is slowed down rather rapidly. ... The diffusion process was studied by chemical analysis, microscopic inspections in the reflected light, and by x-ray diffraction methods."

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Measurement is made of the rate of formation of spinel when pellets of MgO and Cr_2O_3 are heated in contact. The rate of the process is determined by the diffusion of chromium oxide particles through the spinel layer.

(See also references 39, 46, 49, 53, 58.)

Cobalt

37. Borisenko, A. I. and Morozov, E. I., The cobalt-60 diffusion in cobalt ferrites. Doklady Akad. Nauk S.S.S.R. 105, 1274 (1955). Chem. Abs. 50:10360i, 1956.

"The diffusion coefficient in the ceramic-type materials, Co ferrites, was studied and the effect of a uniform phase composition on the diffusion of one of its components. The samples were synthesized in the solid phase by heating the required proportions of Fe_2O_3 and CoO to produce $3\text{CoO} \cdot 2\text{Fe}_2\text{O}_3$, $\text{CoO} \cdot \text{Fe}_2\text{O}_3$, $\text{CoO} \cdot 2\text{Fe}_2\text{O}_3$, $2\text{CoO} \cdot 3\text{Fe}_2\text{O}_3$, and $\text{CoO} \cdot 3\text{Fe}_2\text{O}_3$ A maximum diffusion activation energy was found in $\text{CoO} \cdot \text{Fe}_2\text{O}_3$ which contained the least amount of voids. The diffusion activation energy dropped sharply in ferrites with Fe_2O_3 : CoO less than one."

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39. Sun, R., Diffusion of cobalt and chromium in chromite spinel. J. Chem. Phys. 28, 290 (1958).

Polycrystalline samples of CoCr_2O_4 are annealed in air and analyzed by a sectioning technique. VIII, IX.

(See also reference 50.)

Copper

40. Andrievskii, A. I. and Mischenko, M. T., The diffusion of copper and oxygen atoms during formation of a cuprous oxide layer. Soviet Phys., Tech. Phys. 1, 419 (1956). XV.
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Indium

43. Thomas, D. G., The diffusion and precipitation of indium in zinc oxide. Phys. and Chem. Solids 9, No. 1, 31 (1959). XIII.

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Diffusion of iodine in polycrystalline UO_2 and uranium in UC is studied. XIV.

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A study is made of the diffusion of iron in magnesium-chromite refractories utilizing an autoradiographic technique.

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Diffusion of iron in polycrystalline FeO and Fe_2O_3 and in monocrystalline Fe_3O_4 is studied using a surface counting technique. The data are correlated with the Wagner theory of oxidation. XV.

49. Ignatov, D. V., Belokurova, I. N. and Belyanin, I. N., An investigation of the diffusion processes of iron and chromium in the oxides $\alpha\text{-Al}_2\text{O}_3$, $\alpha\text{-Cr}_2\text{O}_3$, NiCr_2O_4 and NiAl_2O_4 . Atomic Energy Commission Publication No. NP-tr-448, 1958. NSA 14:19351, 1960.

"Diffusion coefficients, activation energies, and frequency factors of iron and chromium were determined in $\alpha\text{-Al}_2\text{O}_3$, $\alpha\text{-Cr}_2\text{O}_3$, NiCr_2O_4 and NiAl_2O_4 at 900 to 1200°C." XV.

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51. Lindner, R., Diffusion of radioactive iron in iron(III) oxide and zinc-iron spinel. Arkiv Kemi 4, 381 (1952). XV, XVII.

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"The diffusion coefficients of iron oxides in magnesite, chromite, and magnesite + chromite were determined at 1500°, 1600° and 1700°C. The diffusion coefficients were generally proportional to porosity, increasing with increase in pore size."

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"... Laboratory experiments have shown that when one face of a pellet of magnetite is held against the face of a magnesiochromite pellet at 1350°, unequal diffusion takes place, Fe ions from the magnetite diffusing more rapidly into the magnesiochromite than Cr ions from the magnesiochromite into the magnetite..."

(See also references 20, 21, 26, 31, 34, 38, 56, 58.)

Lead

54. Lindner, R., Self-diffusion in lead oxide. Arkiv Kemi 4, 385 (1952). Chem. Abs. 47:3072b, 1953.

"Th B (Pb-212) is used as an indicator in the study of the self-diffusion of lead in PbO... No difference in the value of D is found for various crystallographic directions." XVII.

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"The rate of oxidation of Pb is determined between 300° and 600°, and the rate of self-diffusion of Pb-212 in the resulting lead oxide layers is measured between 200° and 590°... The oxidation rate is of the order of 1000 times the diffusion rate, so diffusion cannot be rate-determining in the oxidation process." XVII.

(See also references 26, 51.)

Magnesium

56. Carter, R. E., Mechanism of solid-state reaction between magnesium oxide and aluminum oxide and between magnesium oxide and ferric oxide. J. Am. Ceram. Soc. 44, 116 (1961).
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"Sintering is explained as the result of interdiffusion rather than plastic flow..."

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"The mechanism of the formation of spinels by reaction of the respective oxides in solid state was investigated by aid of self-diffusion measurements in NiCr_2O_4 , NiAl_2O_4 , and ZnAl_2O_4 . . . The diffusion of the cations in Ni and Cr oxide also was measured. . . The reaction mechanism of cation diffusion in opposite directions proposed by Wagner seems probable for the formation of chromites but improbable for the formation of aluminates." VIII, XV, XIX, XX, XXXII.

59. Lindner, R. and Parfitt, G. D., Diffusion of radioactive magnesium in magnesium oxide crystals. J. Chem. Phys. 26, 182 (1957).

Single crystals of MgO are annealed in air and sectioned. Evidence is found of a change in diffusion mechanism around 1400°C. XIX.

(See also reference 36.)

Nickel

60. Gorbunov, N. S. and Izvekov, V. I., The study of diffusion in metal oxides with the aid of radioactive isotopes. Uspekhi Fiz. Nauk 72, 273 (1960). NSA 15:7817, 1961.

"Various experimental arrangements for measuring the diffusion coefficients of elements in metal oxides during sintering at high temperatures in controlled atmospheres are described in detail. Methods of producing radioactive films

by vacuum evaporation are presented. Generally, the diffusion coefficient D is a function of the change in concentration of the diffusing element with depth of penetration x and the time of diffusion t at the sintering temperature. The mathematical equations for the diffusion coefficient D are presented, and graphical solutions are illustrated. The activation energies and diffusion coefficients for the following systems are summarized in tabular form and discussed in detail in the text for the indicated range of temperatures: Cu^{64} in Cu_2O (800 to 1000°C), Zn^{65} in ZnO (800 to 1300°C), Ca in CaO (850 to 1600°C), Fe^{59} in Al_2O_3 (900 to 1200°C), Fe^{59} in TiO_2 (800 to 1000°C), Sn in SnO_2 (1000 to 1260°C), Pb^{212} in PbO (400 to 600°C), Cr^{51} in Cr_2O_3 (1000 to 1350°C), Fe^{59} in Cr_2O_3 , Fe^{55} in FeO , Fe_3O_4 , and Fe_2O_3 (700 to 1200°C), Co^{60} in CoO (800 to 1350°C), and Ni^{63} in NiO (1140 to 1400°C). In a similar fashion the available diffusion data on the movement of various tracers in a number of spinels and silicates are summarized and tabulated. (83 references). " XV, XX, XXXII.

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A study of the exchange of position between tungstates and molybdates of the alkaline earths as well as of Zn, Cd, Ni and Mn.

62. Lindner, R. and Akerstrom, A., Diffusion of Ni-63 in nickel oxide (NiO). Disc. Faraday Soc. 23, 133 (1957). XX.
63. Shim, M. T. and Moore, W. J., Diffusion of nickel in nickel oxide. J. Chem. Phys. 26, 802 (1957).

Both polycrystalline and monocrystalline samples are used with a surface counting technique to measure the diffusion rate of nickel in nickel oxide. It is concluded that the oxidation of nickel proceeds nearly exclusively by diffusion of nickel through a nickel oxide layer. XX.

(See also references 26, 58, 75.)

Oxygen

64. Aronson, S., Roof, R. B., Jr. and Belle, J., Kinetic study of the oxidation of uranium dioxide. J. Chem. Phys. 27, 137 (1957). XXI.
65. Auskern, A. B. and Belle, J., Self-diffusion of oxygen in uranium dioxide. J. Chem. Phys. 28, 171 (1958). XXI.
66. Austerman, S. B., Meyer, R. A. and Swarthout, D. G., Oxygen diffusion in beryllium oxide. Atomic Energy Commission Publication No. NAA-SR-6427, September 15, 1961.

Polycrystalline samples of BeO are annealed in an O-18 atmosphere. The data show two and possibly three regions of diffusion present indicating that no simple mechanism is predominating. XXI.

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Diffusion of oxygen in stoichiometric and in oxygen-rich polycrystalline uranium dioxide is studied. XXI.

68. Fritzsche, H., Effect of dissolved oxygen on the electrical conductivity of zinc oxide. Z. Phys. 133, 422 (1952). Chem. Abs. 47:7851f, 1953.

"The electrical conductivity of evaporated ZnO layers was measured. . . Comparison of the measurements with the theoretical model gives the diffusion constant of oxygen in ZnO layers as about 10^{-14} cm.²/sec. at 460°." XXI.

69. Garino-Canina, V., Diffusion of oxygen in vitreous germanium oxide. Compt. rend. 248, 1319 (1959).
70. Haul, R. and Just, D., Measurement of diffusion in oxide crystals by isotopic exchange with gaseous oxygen. Naturwissenschaften 45, 435 (1958). XXI.
71. Kingery, W. D., Hill, D. C. and Nelson, R. P., Oxygen mobility in polycrystalline NiCr_2O_4 and $\alpha\text{-Fe}_2\text{O}_3$. J. Am. Ceram. Soc. 43, 473 (1960). XXI.

72. Kingery, W. D., Pappis, J., Doty, M. E. and Hill, D. C., Oxygen ion mobility in cubic $\text{Zr}_{0.85}\text{Ca}_{0.15}\text{O}_{1.85}$. J. Am. Ceram. Soc. 42, 393 (1959). XXI.
73. Kuntz, U. E. and Eyring, L., Diffusion of oxygen in rare-earth oxides, in Kinetics of High Temperature Processes (No. 11, this bibliography).

Activation energies for oxygen ion diffusion in the oxides of praseodymium and terbium are determined by loss in weight under vacuum.

74. Moore, W. J., Ebisuzaki, Y. and Sluss, J. A., Exchange and diffusion of oxygen in crystalline cuprous oxide. J. Phys. Chem. 62, 1438 (1958). XXI.
75. Moore, W. J., Lander, J. J., Logan, S. R., O'Keeffe, M., Choi, J. S., Ebisuzaki, Y., Brown, S. and Mitchell, D., Physical chemistry of the solid state. Atomic Energy Commission Publication No. TID-11020, 1960.

Measurement of the diffusion of O-18 in uncompacted NiO crystals and in polycrystalline samples of Cu_2O , as well as the diffusion of nickel in NiO polycrystalline samples is studied. A sectioning technique is used for the latter study. A surface counting technique is used as a check, but gives activation energies about 10 kcal/mole below the tabulated value. The effect of an electric field on grain boundary diffusion is studied, as well as the pressure sensitivity of oxygen diffusion in Cu_2O . XX, XXI.

76. Oishi, Y. and Kingery, W. D., Self-diffusion of oxygen in single crystal and polycrystalline aluminum oxide. J. Chem. Phys. 33, 480 (1960).

Polycrystalline samples give diffusion coefficients about two orders of magnitude greater than monocrystals, and also give lower activation energies. XXI.

77. O'Keeffe, M. and Moore, W. J., Diffusion of oxygen in single crystals of nickel oxide. J. Phys. Chem. 65, 1438 (1961).

The diffusion coefficient increases with increasing oxygen pressure under which the anneal takes place. The mechanism of transport is probably by means of interstitials. XXI.

78. Schnizlein, J. G., Woods, J. D., Bingle, J. D. and Vogel, R. C., Identification of the diffusing species in uranium oxidation. J. Electrochem. Soc. 107, 783 (1960). Ceram. Abs. June 153c, 1961.

"As part of a study of the kinetics of uranium oxidation, experiments were designed to establish whether uranium or oxygen diffuses through the oxide film during the oxidation reaction at 200°C. in 200 mm of oxygen ... Results show that there was an absence of uranium diffusion through the oxide layer ... It was definitely established that oxygen is the diffusing species."

79. Shurmovskaya, N. A., Bruns, B. P. and Mel'nikova, Z. Ya., Reaction between carbon monoxide and manganese dioxide. Z. Fiz. Khim. 25, 1306 (1951). Chem. Abs. 47:7931c, 1953.

"... the activation energy of the diffusion of oxygen in MnO_2 can be estimated to be 9 kcal/mole, although the value of the diffusion constant cannot be calculated (by the method used)..."

(See also references 40, 86, 87, 89.)

Samarium

80. Trofimov, A. K., Studies of luminescence spectra in rare-earth diffusion into various crystals. Izvest. Akad. Nauk S.S.S.R. Ser. Fiz. 21, 757 (1957).

The diffusion of europium and samarium in various crystalline phosphors is studied.

Tantalum

81. Yu-Liang Ho, Diffusion of foreign impurities in cuprous oxide. Wu Li Hsueh Pao 14, 442 (1958).

Measurements of the diffusion of thallium and tantalum in polycrystalline samples of Cu_2O are performed. It is found that below 760°C., diffusion proceeds along grain boundaries, and above 760°C., diffusion proceeds through crystal vacancies.

Tin

82. Lindner, R. and Engvist, O., Self-diffusion in zinc stannate and stannic oxide. Arkiv Kemi 9, 471 (1956). XXVIII.

83. Sinclair, W. R. and Loomis, T. C., Measurements of diffusion in the system $\text{TiO}_2\text{-SnO}_2$, in Kinetics of High Temperature Processes (No. 11, this bibliography).

The diffusion of tin in single crystals of TiO_2 is studied by a sectioning technique. Each section is analyzed by x-ray emissions. XXVIII.

(See also reference 26.)

Uranium

84. Anderson, J. S., Harper, E. A., Moorbath, S. and Roberts, L. E. J., The properties and microstructure of uranium dioxide; Their dependence upon the mode of preparation. Atomic Energy Commission Publication No. AERE C/R 886, August 19, 1952. XXIII.

85. Auskern, A. B. and Belle, J., Uranium ion self-diffusion in UO_2 . J. Nuclear Materials 3, 311 (1961).

A surface counting method is used to measure the diffusion coefficient of uranium in stoichiometric uranium dioxide. XXIX.

86. Belle, J., Properties of uranium dioxide. Atomic Energy Commission Publication No. A/Conf. 15/P/2404, January, 1958.

Measurement of the diffusion of oxygen, uranium, helium, krypton and xenon in polycrystalline UO_2 . A sectioning technique is used for the uranium study. XI, XVI, XXI, XXIX, XXX, XXXII.

87. Belle, J. and Lustman, B., Properties of UO_2 . Atomic Energy Commission Publication No. WAPD-184, September, 1957.

Diffusion of oxygen and uranium in polycrystalline UO_2 . A sectioning technique is used in the uranium study. XXI, XXIX.

88. Kneppel, D. S., Isotopic interchange in dispersion fuels. Atomic Energy Commission Publication No. NMI-1232, October 20, 1960.

Couples of sintered UO_2 in contact with enriched uranium metal annealed in argon are used to study the diffusion of uranium through UO_2 . A sectioning technique is employed. The diffusion coefficient increases with increasing distance from the original interface. XXIX.

89. Rothman, S. J., Diffusion in uranium, its alloys and compounds. V. Diffusion in UO_2 . Atomic Energy Commission Publication No. ANL-5700, Pt. C., May, 1961.

A discussion is given of the work of others in studying the diffusion of uranium, oxygen and rare gases in UO_2 . The data obtained in this study yield no definite results.

(See also references 32, 78.)

Zinc

90. Ferrandis, V. A. and Garcia-Verduch, A., The formation of zinc orthostannate by reaction in the solid state. Anales real soc. españ. fis. y quim. 51B, 189 (1955). Chem. Abs. 49:11477h, 1955.

"Samples of pure ZnO ... and pure SnO_2 ... were intimately mixed in a molar ratio of 2:1 and heated at various temperatures... The rate studies showed that two processes are involved: the initial rate is determined by self-diffusion of Zn ions in ZnO ; in the later stages the rate is determined by the diffusion of Zn ions through the product, Zn_2SnO_4 . The energy of activation of the reaction is 74,700 cal./mole, which is about the same as the activation energy for diffusion of Zn ions in ZnO ..."

91. Lindner, R., Campbell, D. and Akerstrom, A., The diffusion of radioactive zinc in $\text{ZnO} \cdot \text{Fe}_2\text{O}_3$ (spinel) and zinc oxide. Acta Chem. Scand. 6, 457 (1952). XXXII.
92. Munnich, F., Diffusion of zinc in zinc oxide. Naturwissenschaften 42, 340 (1955). Ceram. Abs. September 219b, 1960.

" ZnO crystals... were treated with Zn-65 vapor in quartz glass bombs at 850° to 1100°C . for different times. The activity of the crystals was then determined and also the distribution of the Zn-65 in the ZnO by gradual etching away of the surface The diffusion constants for different temperatures are given."

93. Pohl, R., The diffusion of surplus zinc into single crystals of zinc oxide (Investigations of electrical conductivity). Z. Phys. 155, 120 (1959).

94. Roberts, J. P., Hutchings, J. and Wheeler, C., Sintering and diffusion studies of zinc oxide. Trans. Brit. Ceram. Soc. 55, 75 (1956).

"Brief interim reports are given of studies of (1) the influence of oxygen on the sintering rate of zinc oxide, and (2) the factors affecting the rate of zinc self-diffusion in polycrystalline zinc oxide." XXXII.

95. Roberts, J. P. and Wheeler, C., Diffusion of zinc in polycrystalline zinc oxide. Trans. Faraday Soc. 56, 570 (1960).

"The diffusion of zinc-65 in dense sintered zinc oxide heated at temperatures between 800° and 1400°C. in oxygen, argon, and oxygen-argon mixtures was measured by a sectioning technique. Autoradiography and the shape of the diffusion profile showed that the diffusion was predominately along grain boundaries..." XXXII.

96. Secco, W. A. and Moore, W. J., Diffusion of zinc in crystalline zinc oxide. J. Chem. Phys. 23, 1170 (1955).

Samples consisting of uncompacted crystallites are annealed in a zinc atmosphere. The diffusion coefficient varies as the 0.65 power of the zinc pressure. XXXII.

(See also references 26, 33, 60, 82.)

Inert Gases

97. Booth, A. H., Diffusion of fission product gases from compacted UO_2 . Atomic Energy Commission Publication No. UK/C-5/117, 1956. XXX.
98. Booth, A. H. and Rymer, G. T., Determination of the diffusion constant of fission xenon in UO_2 crystals and sintered compacts. Atomic Energy Commission Publication No. CRDC-720, August, 1958.

Various types of cast and sintered UO_2 samples are studied. The diffusion coefficient is calculated from the fraction of the total xenon that diffuses from the sample. Tabulation of the data shows widely varying values of diffusivity.

99. Gregory, J. N. and Moorbath, S., The diffusion of thoron in solids. Part I. Investigations on hydrated and anhydrous alumina at elevated temperatures by means of the Hahn emanation technique. Trans. Faraday Soc. 47, 844 (1951). XXIII.
100. Lindner, R. and Matzke, H., Diffusion of radon in oxides by recoil indication. Z. Naturforsch. 15a, 1082 (1960).
- The diffusion of radon in Al_2O_3 , TiO_2 , Fe_2O_3 , ThO_2 and UO_2 is studied.
101. Moody, W. E., Whitehead, W. D. and Kriegel, W. W., Activation energy for diffusion of Xe-133 through MgO-Ni cermet compacts. J. Am. Ceram. Soc., 634 (1960). XXX.
102. Research into the diffusion of inert gases in solid bodies. Quarterly report number 2, October 1 to December 31, 1960. Atomic Energy Commission Publication No. TID-12573.

The diffusion of xenon into UO_2 , UC and ThO_2 is studied.

(See also references 84, 86, 89.)

SILICATES AND GLASSES

Aluminum

103. Henderson, J., Ling Yang and Derge, G., Self-diffusion of aluminum in $\text{CaO-SiO}_2\text{-Al}_2\text{O}_3$ melts. Trans. Met. Soc. AIME 221, 56 (1961). Ceram. Abs. September 225a, 1961.

"Self-diffusion coefficients of aluminum were measured by the capillary reservoir technique in liquids of the $\text{CaO-SiO}_2\text{-Al}_2\text{O}_3$ system containing equimolar portions of CaO and SiO_2 in the range 1400° to $1520^\circ\text{C}.$"

Barium

(See reference 26.)

Boron

104. Sakano, T., Nakamura, K. and Moriya, T., The reaction of the system $\text{B}_2\text{O}_3\text{-SiO}_2$. J. Ceram. Assoc. Japan 62, 454 (1954). Chem. Abs. 49:3495g, 1955.

"The reaction rate was determined between B_2O_3 and SiO_2 in the solid state by determining

the silica chemically. The apparent activation energy of the diffusion of B_2O_3 , which is rate determining, is estimated to be 27.6 kcal. "

Calcium

105. Esin, O. A., Vorontsov, E. S. and Chukmarev, S. K., The diffusion of phosphorous and calcium in $CaO-Al_2O_3-SiO_2$ and $CaO-P_2O_5$ melts. Z. Fiz. Khim. 31, 2322 (1957).

106. de Keyser, W. L. and Cypres, R., Utilization of calcium-45 in the study of the reactivity of solids. Inst. Interuniv. Sci. Nucléaires, Monographic Rappt. ann. 1954, 80. Chem. Abs. 52:11545e, 1958.

"In the systems $CaO-Al_2O_3$ and $CaO-SiO_2$ the diffusion of calcium was determined by using Ca-45. The rate of the reaction in the solid state depended on the rate of diffusion. "

107. Lindner, R. and Obermayer, A. S., Diffusion of radioactive calcium in $Ca_3Si_2O_7$. J. Chem. Phys. 23, 188 (1955).

Diffusion is studied using sectioning, surface counting, and conductivity methods. The latter method did not give results in agreement with the former two methods. A large break occurs in the temperature dependence curve which may be due to the freezing-in of high temperature modifications: VI.

108. Lindner, R. and Spicar, E., Kontinuierliche Messung der Selbstdiffusion im Kalziumorthosilikat. Arkiv Kemi 7, No. 62 (1954). VI.

109. Towers, H. and Chipman, J., Diffusion of calcium and silicon in a lime alumina silica slag. J. Metals 9, Trans. AIME 209, 769 (1957).

A capillary method is used to measure the rate of diffusion of calcium and silicon in a liquid slag of the composition $CaO-39$, SiO_2-40 , $Al_2O_3-21\%$. VI, XXIV.

110. Volkonskii, B. V. and Sudakas, L. G., Processes of the isotope exchange and diffusion in calcium silicates. Tsement 23, No. 3, 17 (1957). VI.

(See also reference 119.)

Cobalt

111. Borisenko, A. I., Diffusion migration of cobalt in protective coating on molybdenum. Z. Priklad. Khim. 34, 2175 (1961). NSA 16:5663, 1961.

"The diffusion of cobalt in molybdenum frit coating as a high-temperature corrosion inhibitor is analyzed. The concentration of cobalt increases from the surface coating down to the boundary layer reaching the maximum at the interface layer over molybdenum. The cobalt migration continues from the outer surface of the coating toward molybdenum. The cobalt concentration gradient depends on the composition of frit coating and the vitreous bond. The cobalt diffusion into molybdenum increases with temperature increases."

Copper

112. Ivanova, E. A. and Vargin, V. V., Effect of composition on the process of diffusion of copper ions in glass. Z. Priklad. Khim. 32, 1432 (1959).

Gold

113. Moore, D. G. and Thorton, H. R., Effect of oxygen on the bonding of gold to fused silicon. J. Research Natl. Bur. Standards 62, 127 (1959). Chem. Abs. 53:15702h, 1959.

"... Tests using Au-198 as a tracer indicated that gold diffused into the SiO_2 lattice at oxygen pressures but no diffusion occurred during vacuum heating..."

Hydrogen

114. Kondrat'ev, V., Diffusion of hydrogen in quartz. J. Exptl. Theoret. Phys. U.S.S.R. 13, 59 (1943).

(See also reference 140.)

Iron

115. Ling Yang, Chien, C. and Derge, G., Self-diffusion of iron in iron silicate melts. J. Chem. Phys. 30, 1627 (1959).

A capillary-reservoir method is used to measure the diffusion of Fe-55 and Fe-59 in a $\text{FeO} \cdot \text{SiO}_2$ melt. XV.

116. Pawlowski, S., Qualitative assessment of diffusion and migration of ferric oxide in quartzites and silica products. Prace Inst. Ministerstwa Hutnictwa 9, 253 (1957).
117. Szymborski, W. and Pawlowski, S., Qualitative evaluation of the diffusion of Fe_2O_3 in quartzites and siliceous products by means of the radioactive tracer element Fe-59. Bull. soc. franc. ceram. 1957, 3. Ceram. Abs. May 127c, 1960.

"The depth of penetration of Fe_2O_3 increased with the porosity of the specimen. The diffusion was greater in products manufactured with the addition of calamine than in those containing only CaO. A reducing atmosphere also gave deeper penetration."

Lead

118. Lindner, R., Self-diffusion in lead silicate. Acta Chem. Scand. 5, 735 (1950). XVII.
119. Lindner, R., Silicate formation in the solid state. Z. Phys. Chem. (N. F.) 6, 129 (1956). Chem. Abs. 50:6237g, 1956.

"... Measurement of the self-diffusion constants of the Pb and Ca ions in different silicates over considerable temperature ranges indicate that with increasing interlinkage of the SiO_4 tetrahedra the activation energy for the dislocation and motion of the metal ions increases, and that Wagner's hypothesis of a reaction mechanism based upon cation diffusion in opposite directions through the silicate layer is improbable."

VI.

120. Lindner, R., Hassenteufel, W. and Kotera, Y., Diffusion of radioactive lead in lead metasilicate glass. Z. Phys. Chem. (N. F.) 23, 408 (1960).

The diffusion of Pb-210 and Pb-212 in lead metasilicate glass is considered. Discussion of previous values for cation diffusion in crystalline, glassy and molten silicates of lead and sodium is given. XVII.

(See also references 26, 32, 125.)

Lithium

121. Verhoogen, J., Ionic diffusion and electrical conductivity in quartz. Am. Mineralogist 37, 637 (1952). Chem. Abs. 48:7378g, 1954.

"The diffusion rates of lithium, sodium, and potassium along the [00.1] direction of a quartz crystal in an electric field were determined by measurements of the quantity of material transported, and of electrical conductivity. . . Diffusion of Mg^{++} was too low to be measurable (i. e. $< 10^{-12}$ at 500°), and could not be detected at all for Ca^{++} , Fe^{++} , and Al^{+++} . . . The measurements are used as a basis of discussion of diffusion mechanisms, involving the production and migration of Frenkel and Schottky defects with the diffusion of O^{--} and Si^{++++} , as well as foreign ions taking place. Entropy calculations indicate that lithium and sodium move freely through the undisturbed structure, but potassium moves only by distorting the oxygen framework. . . " XVIII, XXVI.

Magnesium

122. Degueldre, L., The formation of forsterite. Bull. soc. chim. Belges 62, 347 (1953). Chem. Abs. 48:4294e, 1954.

"... The reaction is thought to occur by diffusion of MgO into SiO_2 , $MgSiO_3$ being the first product formed at the interface, and later transformed to Mg_2SiO_4 as the $MgSiO_3$ layer diffuses further into the SiO_2 until reaction is complete. "

(See also reference 121.)

Oxygen

123. Kingery, W. D. and Lecron, L. A., Oxygen mobility in two silicate glasses. Phys. and Chem. Glasses 1, 87 (1960). Ceram. Abs. May 114c, 1961.

"Oxygen diffusion coefficients were determined for two glasses by means of isotope exchange measurements using the stable isotope $O-18$ and mass spectrometric analysis. . . "

124. Sawai, I., Tashiro, M. and Ago, S., Factors affecting diffusion velocity of oxygen through heat-resistant enamel coatings. Bull. Inst. Chem. Research Kyoto Univ. 28, 61 (1952). Chem. Abs. 47:8335h, 1953.

"... The diffusion velocity of oxygen is controlled mainly by the fusibility of enamel and the relationship is linearly inversely proportional. The composition of the enamel does not affect the diffusion velocity when the fusibilities of the enamels are the same."

(See also reference 140.)

Radium

125. Rosenqvist, I. T., Some investigations in the crystal chemistry of silicates. I. Diffusion of Pb and Ra in feldspars. Acta Chem. Scand. 3, 569 (1949).

"A method of studying diffusion processes in crystalline silicates is outlined. The diffusion coefficients of Pb and Ra in potash and soda-feldspars are measured at different temperatures. It is shown that the diffusibility depends on the crystallographic directions. The diffusion anisotropy seems to increase with increasing temperature." XVII, XXII.

Silicon

126. Borisov, A. F. and Dertev, N. K., The measurements of diffusion coefficients of silica in glass melts of the system sodium oxide-silicon oxide. Trudy Gor'kovsk. Politekh. Inst. 13, No. 5, 21 (1957). Chem. Abs. 53:16494g, 1959.

"A method for continuous determination of the diffusion coefficients of oxides in glass melts was developed. This method is based on e. m. f. measurements close to a quartz crystal that is dissolving in a homogeneous melt... A number of diffusion coefficients of SiO_2 in different glass melts (system $\text{Na}_2\text{O-SiO}_2$) were determined..."

127. Lungu, S. N., The diffusion in glass and its mechanism. Acad. rep. populare Romane, Bul. Stiint., Sect. Stiint. teh. si chim. 5, 85 (1953). Chem. Abs. 50:14193d, 1956.

"The diffusion coefficients of silicon were measured in combinations of four soda-lime glasses... at 1360-70°..."

(See also reference 119.)

Silver

128. Kinumaki, J. and Ito, T., The diffusion of silver into glass. Science Repts. Research Insts. Tohoku Univ., Ser. A, 8, 60 (1956). XXV.

Sodium

129. Hemano, K., Diffusion layers formed around quartz grains dissolving in feldspathic melts. Yogyo Kyokai Shi 64, 217 (1956). Ceram. Abs. February 57a, 1958.

"The glassy diffusion layer formed around the quartz grains was studied microscopically, and its refractive index was determined. In the case of potash feldspar the diffusion layer was continuous and had a linear gradient of refractive index, while in the case of soda feldspar a double diffusion layer appeared temporarily; after prolonged soaking this double layer disappeared. "

130. Johnson, J. R., Photo-density method of determining the relative penetration of diffused sodium-24 tracer into glass. J. Appl. Phys. 20, 129 (1949).
131. Kamel, R., Measurement of the heat of activation for sodium-ion diffusion in glass. J. Appl. Phys. 24, 1308 (1953). Chem. Abs. 48:1097a, 1954.

"Internal friction peaks resulting from Na-ion diffusion in three kinds of soda glasses were observed in the temperature range 100° to 200° for transverse and torsional vibration frequencies between 20 to 400 cycles/sec. The heat of activation for Na-ion diffusion was 17,000-21,000 cal./mole and probably might depend slightly on the sodium content. The results were confirmed by electrical resistivity measurements. . . "

132. Leavitt, W. Z., Geochemical studies, in The Adaptation of New Research Techniques to Mineral Engineering Problems, Atomic Energy Commission Publication No. NYO-3670, 1952.

Diffusion of sodium in single crystals of albite feldspar is studied. XXVI.

133. Leavitt, W. Z., Geochemical studies, in The Adaptation of New Research Techniques to Mineral Engineering Problems, Atomic Energy Commission Publication No. NYO-3670, October, 1952.

Diffusion coefficient of sodium in a single crystal of oligoclase feldspar is measured perpendicular to the "c" axis. XXVI.

134. Petzold, A., Use of radioactive isotopes in silicate investigation. Silikattech. 6, 321 (1955).

(See also reference 121.)

Strontium

135. Walker, G. F., Diffusion of exchangeable cations in vermiculite. Nature 184, 1392 (1959).

The exchange of magnesium and strontium ions in vermiculite single crystals is measured optically by noting the change in the "c" dimension of the crystal. Strontium replacing magnesium shows an activation energy of about 12 kcal/gram ion.

Inert Gases

136. Braaten, E. O. and Clark, G. F., The diffusion of helium through fused silica. J. Am. Chem. Soc. 57, 2714 (1935).

137. Brandt, W. W., Diffusion in zeolites and glasses. Atomic Energy Commission Publication No. TID-11840, February, 1961.

Study of the diffusion of argon, methane and carbon dioxide in zeolite using a radioactive tracer technique. II.

138. Burton, E. F., Braaten, E. O. and Wilhelm, J. O., Diffusion of helium through quartz; Relation to temperature. Can. J. Research 8, 463 (1933).

139. Jaquerod, A. and Perrot, F. L., Diffusion of helium through silica. Compt. rend. 139, 789 (1904).

140. McAfee, K. B., Stress-enhanced diffusion in glass: I. Glass under tension and compression. J. Chem. Phys. 28, 218 (1958). Ceram. Abs. August 1958d, 1958.

"... The diffusion constant of helium was found to be a true constant with dilation of the glass until the stress became about one-half the breaking stress of the glass. Beyond this point the diffusion constant increased and under very high stress was a factor of ten larger than its original value... It was further determined that equal and even larger compressional stresses on the same glass specimen had little or no effect on the diffusion constant..."

II. ibid., p. 226... It was found that shear strain, γ , up to 4×10^{-3} does not alter the diffusion constant appreciably in the glass..."

141. Norton, F. J., Helium diffusion through glass. J. Am. Ceram. Soc. 36, 90 (1953). Chem. Abs. 47:4567i, 1953.

"A mass spectrometer... was used both as an analytic tool and to determine the rate of diffusion of helium, at -80 to 600°C ., through nine types of glass, including fused SiO_2 , borosilicate, phosphate, soda lime, combustion tubing, x-ray shield, Pb borate, and chemical glassware. The highest rates of diffusion were for fused SiO_2 and Corning Vicor glass (SiO_2 96, B_2O_3 3, Al_2O_3 1%); the lowest were for x-ray shield and Pb borate glasses. The rate of diffusion decreased enormously with the decrease in the percentage of glassformers (SiO_2 , B_2O_3 , P_2O_5) in the glass. A few determinations were made of the rate of diffusion of hydrogen, deuterium and neon through fused SiO_2 ..."

142. Pellas, P., The mechanism of diffusion of helium in zircons. C. R. Acad. Sci. (Paris) 235, 1134 (1952).

143. Reynolds, M. B., Diffusion of argon in a potassium-lime-silica glass. J. Am. Ceram. Soc. 40, 395 (1957).

"Diffusion coefficients for bulk diffusion of argon in a potassium-lime-silica glass have been determined at temperatures in the range 308° to 470° by a radioactive counting technique. Argon diffusion was found to follow classical bulk diffusion theory within the limits of experimental accuracy..."

General

144. Beattie, I. R., Reaction between water and vitreous silicates. Trans. Faraday Soc. 49, 1059 (1953). Chem. Abs. 48:5587e, 1954.

"Several series of glasses, obtained by the substitution of various oxides in base glass compositions were examined; aluminum replacing silicon, boron replacing silicon, Na_2O replacing CaO , and Na_2O replacing SiO_2 . The products of reaction were removed as they were formed. The rate-controlling process is probably the migration of Na^+ through the glass. Calculated values for the internal diffusion coefficients and the apparent energies of activation agree well with the corresponding values found for ionic migration in similar systems. The diffusion coefficient decreased as silicon was replaced by boron or aluminum. The substitution of SiO_2 by soda is complex, as the two oxides do not act in a similar manner in the glass structure. Strong Si-O-Si bonds are broken and the concentration of the diffusion ion increased. The replacement of two relatively mobile sodium ions by a comparatively immobile calcium ion caused marked decrease in the measured diffusion coefficient. "

145. Charles, R. J., Polarization and diffusion in a silicate glass. J. Appl. Phys. 32, 1115 (1961). Ceram. Abs. October 238d, 1961.

"The self-diffusion of alkali ions in alkali silicate glasses is discussed in terms of the formation, migration, and ultimate annihilation of defects. . . Relations between self-diffusion, a. c. and d. c. conductivity, and orientation polarization were found. "

146. Eitel, W., The Physical Chemistry of the Silicates, The University of Chicago Press, Chicago, 1954.

An extensive compilation of work on solid state reactions in the silicates drawing heavily on European literature. The book covers such subjects as diffusion anisotropy in zeolites, diffusion of ions in silicates and glasses, diffusion of gases through silica and soda-lime glass, inner diffusion of albite and anorthite, and others. XII.

147. Johnson, J. R., Bristow, R. H. and Blau, H. H., Diffusion of ions in some simple glasses. J. Am. Ceram. Soc. 34, 165 (1951).

"An investigation of the diffusion of ions in several glasses was made using radioactive tracer techniques. The resulting diffusion curves were analyzed to obtain the thermodynamic properties of the diffusion process. These properties are indicative of energy and structure changes in the glass networks. Interpretations of the results are advanced. . . "

148. Munakata, M., Kuwamura, H., Iwanoto, M. and Niitsu, H., Alkali migration in glasses. I. Bull. Electrochem. Lab., Japan 18, 846 (1954). Chem. Abs. 49:15196b, 1955.

"The diffusion constant of an alkali ion which is calculated from the rate of water solubility of 8 kinds of soda-lime glass in which Al_2O_3 is substituted for SiO_2 was compared with that calculated from the electrical conductivity. . . The water solubility of glass is regulated by some factors other than the mobility of alkali ion in bulk glass. "

149. Muth, H., Radioactive tracer methods in glass technology. Glastech. Ber. 27, 248 (1954). Chem. Abs. 48:13188d, 1954.

"... The special field in which radioactive tracer methods are useful in glass technology is the study of complex physical-chemical reactions during the manufacturing and working of glass, with special emphasis given to diffusion and softening phenomena, the surface changes, the problems of corrosion by chemical agents, etc. . . "

150. Nazarenko, M. F. and Razumova, V. L., The reaction of kaolin with feldspar melts in the presence of quartz. Izvest. Akad. Nauk Kazakh. S. S. R. 1957, No. 3, 78.

151. Nitkitin, O. A., Esin, O. A. and Vorontsov, E. S., The determination of the diffusion coefficients in fused oxides. Z. Fiz. Khim. 32, 1420 (1958). Chem. Abs. 53:2722b, 1959.

"... The method used for the diffusion measurements of samples of $\text{CaO-Al}_2\text{O}_3\text{-SiO}_2$ system at 1500° involved the use of radioactive tracers, and polarization of electrodes by an alternating current. . . "

BORIDES, CARBIDES AND GRAPHITE

Barium

152. Young, C. T. and Smith, C. A., Preliminary experiments on fission product diffusion from uranium-impregnated graphite in the range 1800-2200°C. Atomic Energy Commission Publication No. NAA-SR-232, June, 1953.

Measurement of the diffusion of barium, strontium and zirconium in uranium-impregnated graphite. Zirconium shows much slower diffusion than barium and strontium.

(See also reference 164.)

Boron

153. Kislii, P. S. and Samsonov, G. V., The diffusion of boron in carbon. Fiz. Tverdogo Tela. 2, 1729 (1960). NSA 15:1467, 1961.

"Preliminary experiments are performed on the diffusion of boron in graphite and investigations of the properties of the boron carbide obtained in this way. It is found that by the diffusion of boron in graphite alloys are obtained which show greater solidity and lower brittleness than boron carbide obtained by compression under heat. These alloys have semiconductor properties, and can be utilized for the preparation of high temperature thermocouples. By the diffusion of boron into the surface of graphite samples, their corrosion resistance becomes noticeably higher, particularly at higher temperatures. The purpose of this work is to investigate the mechanism of diffusion and to determine its parameters. The object investigated is a cylindrical sample of spectroscopically pure graphite onto whose surface a 2 mm thick layer of a paste of amorphous boron is applied. After the samples are dried at 150°C, they are enclosed in a graphite shell and preheated in an atmosphere of hydrogen at 700 to 800°C, for 60 to 80 min. After this treatment the samples are subjected to metallographic, chemical, and x-ray analyses. Further, the reverse process of diffusion of carbon in boron is investigated. For this purpose, boron samples of a porosity of 36% are employed. They are pre-

pared by compression of boron powder and sintering at 1900°C. In this case there results a saturation of the carbon samples with carbon in 30 minutes in a vacuum oven at 1940°C. Experiments show that in similar conditions the boron penetrates deeper in carbon (1.4 to 1.6 mm) than carbon does in boron (0.6 to 0.8 mm). This indicates a remarkably higher mobility of boron atoms. The diffusion coefficients are calculated to be $6.2 \cdot 10^{-6} \text{ cm}^2/\text{sec}$ ($B \rightarrow C$) and $1.8 \cdot 10^{-6} \text{ cm}^2/\text{sec}$ ($C \rightarrow B$). Numerical data for two samples showing boron content at different depths of the carbon sample (chemical analysis) are given. The boron concentration diminishes exponentially with depth. That a solid solution is formed due to diffusion, is shown by an x-ray analysis. Here the interplanar spacings of graphite lattice are measured as function of boron concentration. Further, the temperature dependence of diffusion of boron in graphite is investigated; $D = 3.02 \exp(-28625/T)$ is found to hold. " V.

154. Samsonov, G. V. and Neshpor, V. S., Investigation of mutual diffusion of borides of titanium and niobium. Doklady Akad. Nauk S.S.S.R. 101, 899 (1955). Chem. Abs. 50:1400d, 1956.

"The diffusion of equimolar mixtures of powdered TiB_2 and NbB_2 was studied by heating them after compression in a vacuum electric furnace at 1400, 1600, and 1800°. . . An x-ray examination of the samples disclosed a substantially single-phase solid solution of the components, with NbB_2 acting as the solvent. . . The activation energy was 25,000 cal./mole, or about $\frac{1}{2}$ as large as the diffusion activation energy in TiB_2 - ZnB_2 mixtures; this shows that the metal atoms play the principal role in the diffusion in boride systems. "

Carbon

155. Dayton, R. W. and Tipton, C. R., Jr., Progress relating to civilian applications. Atomic Energy Commission Publication No. BMI-1504, March, 1961. VII.
156. Dienes, G. J., Mechanism for self-diffusion in graphite. J. Appl. Phys. 23, 1194 (1953).

157. Feldman, M. H., Goeddel, W. V., Dienes, G. J. and Gossen, W., Studies of self-diffusion in graphite using C-14 tracers. J. Appl. Phys. 23, 1200 (1952).
158. Kanter, M. A., The mechanism for atom motion in graphite crystals, in Kinetics of High Temperature Processes (No. 11, this bibliography).

Self-diffusion of carbon in graphite is studied by two methods; (1) diffusion of C-14 plated on naturally occurring graphite crystals (poor results); (2) uptake of C-14 from a C-14O₂ atmosphere. The mechanisms of atom motion are considered. VII.

159. Kuczynski, G. C. and Landauer, R., Measurement of carbon diffusion in metallic carbides. J. Appl. Phys. 22, 152 (1951).

"A method for measuring the diffusion coefficient of carbon in metallic carbides is proposed. The rate at which excess metallic inclusions are consumed by carbon diffusing in from a surface is used to indicate the extent to which diffusion has taken place. Conditions are derived under which the results are susceptible to simple analytic treatment."

160. Tomonari, T., Carborundum. II. Reaction mechanism of SiC formation. Electrochem. Soc. Japan 24, 27 (1956). Chem. Abs. 50:10993c, 1956.

"... five mechanisms are considered as the synthetic reaction of SiC... in all of these the rate of reaction is closely related to the diffusion coefficient of C in SiC."

(See also reference 153, 163.)

Cobalt

161. Turnbull, R. C., Diffusion of iron, nickel and cobalt into hot-pressed titanium carbide. Atomic Energy Commission Publication No. WADC-TR-53-1. January, 1953.

Surface counting and sectioning techniques show that grain boundary diffusion predominates in this system. Diffusion coefficients at different temperatures are tabulated.

Iodine

(See reference 44.)

Iron

(See reference 161.)

Nickel

(See reference 161.)

Strontium

(See references 164, 152.)

Uranium

162. Loch, L. D., Gambino, J. R. and Duckworth, W. H., Diffusion of uranium through graphite. A. I. Chem. E. Journal 2, 195 (1956).
163. Rough, F. A. and Chubb, W., ed., Progress on the development of uranium carbide-type fuels. Atomic Energy Commission Publication No. BMI-1488, December 27, 1960.

The interdiffusion of uranium and carbon and the diffusion of uranium in cast UC is studied by a sectioning technique. VII, XXIX.

(See also reference 44.)

Yttrium

164. Saunders, A. R., A study of fission product transport mechanisms in high temperature gas-cooled reactor fuel elements. Atomic Energy Commission Publication No. ORNL-3145, July 13, 1961.

"The experimental work on the diffusion of fission products in ceramic matrices at high temperatures is reviewed and the possible mechanisms of mass transport are discussed. Preliminary data are given on diffusion in these systems..." III, XXVII, XXXI, XXXIII.

Zirconium

(See references 164, 152.)

Inert Gases

165. von der Decken, C. B., Investigation of fuel elements in a high-temperature loop test. Tech. Mitt. Krupp 19, 173 (1961). NSA 16:5548, 1961.

"Fission products are classified according to volatility. Service conditions of spherical fuel elements of uranium carbide and graphite in a high-temperature reactor are discussed. Experimental facilities are described. Results of diffusion measurements for Xe^{133} , Xe^{135} , Kr^{85m} , and Kr^{87} from uranium carbide cartridges are given. Diffusion of inert gases in graphite is discussed. Results obtained in measuring diffusion from the fuel elements, using different grades of graphite are presented and discussed. The importance of inert gases of long half life in cooling system contamination is also discussed."

(See also reference 164.)

TABLES

I. Aluminum

System	D_o	Q	T	Reference
$Al_2O_3^*$	2.5×10^4	135		22

II. Argon

System	D_o	Q	T	Reference
Zeolite	4.8×10^{-5}	2.2	100	137

III. Barium

System	D_o	Q	T	Reference
BaO^*		10.2	<1117	27
BaO^*		254	>1117	27
$BaTiO_3^{(1)}$	1.8	58	884-1180	24, 25
$BaTiO_3$	0.8	89	884-1180	24, 26
$BaSiO_3$	5×10^{-2}	80		26
Graphite		16.4	1225-1500	164

(1) After exposure to an electric field.

IV. Beryllium

System	D_o	Q	T	Reference
BeO	5.56×10^3	111.6	1570-1730	28
BeO	6.14×10^{-2}	66.1	1730-1934	28
BeO	1.37	91.9	1550-1725	29
BeO	1.10×10^{-6}	36.15	1725-2000	29
BeO		114		30

* Single crystal

V. Boron

System	D_o	Q	T	Reference
C	3.02	28.63		153

VI. Calcium

System	D_o	Q	T	Reference
CaO	0.4	81	850-1600	33
CaCO ₃		60		32
α -CaSiO ₃	7×10^{-4}	112	1130-1400	119
β -CaSiO ₃	0.2	78		119
α -Ca ₂ SiO ₄	2×10^{-2}	55		108
α' -Ca ₂ SiO ₄	3.6×10^{-2}	65		108
Ca ₃ SiO ₅	5.5×10^{-3}	95		110
Ca ₃ Si ₂ O ₇	10^{-2}	73	<1260	107
CaFe ₂ O ₄	30	86	890-1140	31
Ca-Al-Si Slag ($D=1 \times 10^{-6}$)		70	1430	109

VII. Carbon

System	D_o	Q	T	Reference
Graphite	4-24	163	1995-2350	158
UC	$(D=5 \times 10^{-8})$		1600	155
U (Interdiffusion)	148	70	1200-2000	163

VIII. Chromium

System	D_o	Q	T	Reference
Cr_2O_3	4×10^{-3}	100	1000-1350	58
$ZnCr_2O_4$	9	81	1000-1400	58
$CoCr_2O_4$	2	70	1400-1600	39
$NiCr_2O_4$	0.75	73	950-1450	46
$NiAl_2O_4$	1.17×10^{-3}	50	900-1200	46

IX. Cobalt

System	D_o	Q	T	Reference
CoO	1.4×10^{-2}	34.5	800-1350	38
$CoCr_2O_4$	10^{-3}	51	1400-1600	39

X. Copper

System	D_o	Q	T	Reference
Cu_2O	4.4×10^{-2}	36.1	800-1000	42
Cu_2O	3.58×10^{-2}	37	800-1000	41

XI. Helium

System	D_o	Q	T	Reference
UO_2		46		86

XII. Hydrogen

System	D_o	Q	T	Reference
Silica Glass	$(D=10^{-5}-10^{-6})$		300-1000	146

XIII. Indium

System	D_o	Q	T	Reference
ZnO	2.5×10^2	3.2	800-1300	43

XIV. Iodine

System	D_o	Q	T	Reference
UO ₂	($D=1.8 \times 10^{-10}$)		1000	44
UO ₂	($D=4.64 \times 10^{-10}$)		1200	44

XV. Iron

System	D_o	Q	T	Reference
Al ₂ O ₃	9.18×10^{-8}	27.6	900-1100	60
CaFe ₂ O ₄	3.2	72	835-1092	31
CaFe ₂ O ₄	0.4	72		26
Cr ₂ O ₃	4.95×10^{-6}	44	900-1200	49
FeO	1.4×10^{-2}	30.2	700-1000	48
Fe ₂ O ₃	4×10^4	112	750-1300	51
Fe ₂ O ₃	4×10^5	112	1000-1217	48
Fe ₃ O ₄	0.25	53.9	770-1200	40
Fe ₃ O ₄ [*]	5.2	55	750-1000	48
FeSiO ₃ melt		40	1250	115
NiAl ₂ O ₄	1.33	82	900-1200	46, 49
NiFe ₂ O ₄	5×10^2	82		58
ZnFe ₂ O ₄	10^3	82	750-1300	51
ZnFe ₂ O ₄	8.5×10^2	82		26
TiO ₂	1.98×10^{-2}	55	800-1000	60
TiO ₂	1.92×10^{-1}	55.4	770-1000	60

* Single crystal

XVI. Krypton

System	D_o	Q	T	Reference
UO ₂		30	<1150	86

XVII. Lead

System	D_o	Q	T	Reference
PbO	10^6	69	400- 600	51
PbO	10^5	66		26, 54
PbO		13	< 460	55
PbO		66	> 460	55
PbSiO ₃	85	59.5	550- 700	118
PbSiO ₃		59.5	300- 600	120
PbSiO ₃ glass	10^{-4}	24.8	300- 600	120
PbSiO ₄	0.2	47	550- 700	118
PbSiO ₄	8.2	47		26
Microcline*	2.5×10^{-6}	21.3	603-1038	125
$\perp \{001\}$				
Albite* $\perp \{001\}$	2.5	43.2	873-1038	125

XVIII. Lithium

System	D_o	Q	T	Reference
Quartz* (1)	6.9×10^{-3}	20.5		121

(1) In an electric field.

* Single crystal

XIX. Magnesium

System	D_o	Q	T	Reference
MgO	0.23	78.7	1400-1600	59
MgAl ₂ O ₄	2×10^2	78		58

XX. Nickel

System	D_o	Q	T	Reference
NiO	4.4×10^{-4}	44	1000-1400	63
NiO	1.7×10^{-2}	56		62
NiO	8.01×10^{-4}	53.5	1300-1700	75
NiO*	3.9×10^{-4}	44.2	1000-1400	63
NiAl ₂ O ₄	2.9×10^{-5}	53.3	880-1388	60
NiCr ₂ O ₄	1.5×10^{-3}	61.4	1130-1450	58
NiCr ₂ O ₄	0.85	75		26

XXI. Oxygen

System	D_o	Q	T	Reference
Al ₂ O ₃ *	1.9×10^3	152	>1600	76
Al ₂ O ₃ *	6.3×10^{-8}	57.6	<1600	76
BeO	5.2×10^{-7}	42.9	1600-1900	66
CdO	($D=2.7 \times 10^{-17}$)		600	70
CdO	($D=7.04 \times 10^{-16}$)		649	70
CdO	($D=1.32 \times 10^{-15}$)		698	70
Cu ₂ O	6.5×10^{-8}	39.6	1030-1120	74, 75
α -Fe ₂ O ₃	1×10^{11}	146	1450-1520	71
NiCr ₂ O ₄	1.7×10^{-2}	65.4	1450-1800	71

* Single crystal

XXI. Oxygen (cont.)

System	D_o	Q	T	Reference
NiO*	1.0×10^{-5}	54	1100-1500	77
NiO	($D=3.9 \times 10^{-13}$)		1300	75
UO ₂	($D=1.0 \times 10^{-13}$)		500	86
UO ₂	2.6×10^{-5}	29.7	450- 600	65, 67, 87
UO ₂	5.5×10^{-3}	26.3	160- 350	64
UO ₂ (O ₂ -rich)	4.0×10^{-4}	29.7	317- 465	67
ZnO	($D=10^{-14}$)		460	68
Zr(Ca)O ₂	1.0×10^{-2}	1.22	700-1100	72

XXII. Radium

System	D_o	Q	T	Reference
Microcline* ⊥{001}	5.1×10^{-4}	24.3	823-1063	125
Microcline ⊥{010}	2.4×10^{-4}	24	823-1063	125
Microcline ≠ [100]	2×10^{-2}	29.2	823-1063	125
Albite* ⊥{001}	4.8×10^{-3}	31	848-1073	125
Albite ⊥{010}	1.6×10^{-3}	29.8	848-1073	125
Albite ≠ [100]	5×10^{-2}	32.2	848-1073	125

* Single crystal

XXIII. Radon

System	D_o	Q	T	Reference
Al_2O_3		60.6		99
Cr_2O_3		49.5		99
Fe_2O_3		42.2		99
TiO_2		120		99
UO_2		40-81	800-1350	84

XXIV. Silicon

System	D_o	Q	T	Reference
Ca-Al-Si Slag	($D = 1 \times 10^{-7}$)		1430	109

XXV. Silver

System	D_o	Q	T	Reference
Glass	1.91	27	500- 600	128

XXVI. Sodium

System	D_o	Q	T	Reference
Albite*	($D = 6 \times 10^{-10}$)		1000	132
Oligoclase*	($D = 3.3 \times 10^{-10}$)			133
Quartz*(1)	3.6×10^{-3}	31.7		121

(1) In an electric field.

* Single crystal

XXVII. Strontium

System	D_o	Q	T	Reference
Graphite		52.1	800-1150	164
Graphite		119.8	1500-1700	164

XXVIII. Tin

System	D_o	Q	T	Reference
SnO_2	10^6	119	1000-1260	82
SnO_2	10^7	126		26
SnZn_2O_4	2×10^5	109	1000-1260	82
TiO_2^*	0.16	80	900-1050	83

XXIX. Uranium

System	D_o	Q	T	Reference
UC	0.23	82	1600-2000	163
UO_2		90-120		32
UO_2		90-125	1410-1750	87
UO_2	4.3×10^{-4}	88	1450-1785	85
UO_2	$(D=5-45 \times 10^{-18})$		1000	88
UO_2	$(D=10^{-13})$		1410	86
UO_2	$(D=4.3 \times 10^{-12})$		1750	86
C (Interdiffusion)	148	70	1200-2000	163

* Single crystal

XXX. Xenon

System	D_o	Q	T	Reference
UO ₂		45		86
UO ₂ *	($D = 7 \times 10^{-14}$)		1400	97
MgO-Ni Cermet	1.85×10^7	101.5	800-1100	101

XXXI. Yttrium

System	D_o	Q	T	Reference
Graphite		6.3	1350-1700	164

XXXII. Zinc

System	D_o	Q	T	Reference
SnZn ₂ O ₄	37	76.3	1000-1250	86
ZnAl ₂ O ₄	2×10^2	78	1000-1400	60
ZnCr ₂ O ₄	60	85	1000-1400	58
ZnFe ₂ O ₄	10^3	86	900-1350	33, 91
ZnFe ₂ O ₄	8.8×10^{-2}	86		26
ZnO	4.8	73	900-1025	96
ZnO	1.3	74	800-1370	33, 91
ZnO	10^{-1}	89	800-1300	60
ZnO		24		95
ZnO ⁽¹⁾	($D = 0.6 \times 10^{-10}$)		1290	94
ZnO ⁽²⁾	($\bar{D} = 2.2 \times 10^{-10}$)		1290	94

(1) Annealed in oxygen.

(2) Annealed in argon.

* Single crystal

XXXIII. Zirconium

System	D_o	Q	T	Reference
Graphite		136.8	1500-1700	164

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