ed with the compositions of their precursor sediments, because up to half of the rock consists of siliceous cement added postdepositionally. This is a major factor in the often noted compositional differences between arenaceous (cherty) and lutitic (slaty) rocks in iron formations.

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19. Well-sorted medium sands of epiclastic quartz average 40 to 45 percent initial porosity [D. C. Beard and P. K. Weyl, *Bull. Am. Assoc. Pet. Geol.* **57**, 351 (1973)]; well-sorted, medium sand-size calcareous ooids from the Bahamas average 43 percent initial porosity [R. B. Halley and P. M. Harris, *J. Sediment Petrol.* **49**, 980 (1973)]. Iron formation sands of comparable grain size described by G. A. Gross [*Sediment Geol.* **7**, 244 (1972)] show 40 percent intergranular material, interpreted here as cement.
20. We thank I. S. Zajac for the loan of fossiliferous material and H. J. Hofmann for helpful criticism. Fieldwork by B.S. was supported by grants in aid of research from Sigma Xi, the Geological Society of America, and Johns Hopkins University, as well as NSF grant EAR 77-08137 (L. A. Hardie, principal investigator). Financial support was also provided by NSF grant DEB 80-04290 (A.H.K.) Illustrated specimens will be deposited in the Canadian National Type Collection at the Geological Survey of Canada, Ottawa.

15 October 1980

Effects of Trace Gases and Water Vapor on the Diffusion Coefficient of Polonium-218

Abstract. *The two-filter method described by Thomas and LeClare was used to investigate the effects of trace concentrations of nitric oxide and nitrogen dioxide in dry nitrogen on the diffusion coefficient of radium A (polonium-218). Charged radium A was neutralized in 10 parts per million (ppm) nitrogen dioxide in dry nitrogen, in 8.3 ppm nitric oxide in dry 92 percent nitrogen and 8 percent oxygen, and in nitrogen with 20 and 80 percent relative humidity. No neutralization was seen in dry nitrogen, dry oxygen, dry air, or 10 ppm nitric oxide in dry nitrogen. The diffusion coefficient of the neutral radium A species was found to be 0.079 square centimeters per second, regardless of the relative humidity of the nitrogen gas atmosphere. Lower values were observed for charged species.*

There are conflicting reports of values for the molecular diffusion coefficient (D) of several of the radon daughter isotopes. Chamberlain and Dyson (*1*) first estimated the D value of radium A (^{218}Po) in dry air to be 0.054 cm^2/sec on the basis of their experimental evidence for ^{220}Rn decay products. Porstendorfer (*2*) found a 33 percent reduction in the D value of charged ^{212}Pb (thorium B), a decay product in the series of radionuclides formed from ^{220}Rn, relative to that of the neutral species. Porstendorfer and Mercer (*3*) later determined that the charged thoron daughters appear 88 percent of the time, and the neutral species appear the remaining 12 percent of the time. Raabe (*4*) reported a reduction in the value of D for radium A as a function of increasing humidity. At 16 percent relative humidity (RH), D was 0.047 cm^2/sec; at 35 percent RH, D was reduced to 0.034 cm^2/sec.

Porstendorfer and Mercer (*3*) have clearly distinguished the properties of neutral and charged ^{212}Pb. For the neutral species, D was found to be 0.068 cm^2/sec independent of RH. The D value of the charged species could not be determined unequivocally but was estimated as 0.024 cm^2/sec, and was found to be affected by increasing humidity. In air with RH between 30 and 90 percent D for the positively charged species was found to be 0.068 cm^2/sec, the same as for the neutral species. Thomas and LeClare (*5*) also observed a change in the D value of radium A at RH up to 20 percent but a constant value with increasing RH above 20 percent. Kotrappa *et al.* (*6*) observed no change in the D value of thoron decay products between 5 and 90 percent RH but did see a decrease in D with increasing residence time in their diffusion carboy. More recently, Kotrappa and Raghunath (*7*) compared the D values of radon and thoron daughters in air to the values in pure argon. Although small decreases in D were seen with increasing RH in the argon atmosphere, substantial decreases were observed in air.

Table 1. Diffusion coefficients (D) of radium A in different experimental gases.

Experimental gas in N_2	D (cm^2/sec)
N_2 (dry)	0.044 ± 0.003
97 percent O_2 (dry)	0.031 ± 0.003
10 ppm NO (dry)	0.031 ± 0.003
10 ppm NO_2 (dry)	0.072 ± 0.004
8.3 ppm NO (dry) with 8 percent O_2	0.079 ± 0.004
Dry pump air	0.027 ± 0.003
20 percent RH N_2	0.052 ± 0.003
80 percent RH N_2	0.079 ± 0.004

Busigin *et al.* (*8*) have examined D in a manner similar to that used by Raabe. They observed complex transmission curves for the passage of ^{218}Po through a diffusion tube. They suggested that complex reactions are occurring that lead to the presence of several different diffusing species.

The variation and conflicting nature of these earlier results indicate that complex mechanisms govern the behavior of D. More than one mechanism may be in operation. Possibly Kotrappa and Raghunath saw the formation of heavier, more slowly diffusing molecules in air. Porstendorfer and Mercer, as well as Thomas and LeClare, observed the neutralization of charged species of these radioelements. Busigin *et al.* suggested the importance of trace gases with low ionization potentials in the neutralization process. We have reexamined the D value of ^{218}Po in an effort to resolve these questions.

The two-filter method described by Thomas and LeClare (*5*) was used to measure the D values of radium A in N_2 with 0, 20 and 80 percent RH; in 97 percent dry O_2; in 10 parts per million (ppm) NO_2 and 8 percent O_2; and in dry pump air. Three stainless steel tubes (50, 75, and 100 cm in length and 3.17 cm in diameter) were used with a glass-fiber filter placed at the inlet to remove radon daughters from the gas stream. The radon daughters formed in the tube as a result of decay and not diffusing to the tube wall were sampled with a glass-fiber filter at the tube outlet. We supplied radon gas to the sampling system by bubbling gas through an acid solution of radium chloride and diluting into a larger stream of the experimental gas ahead of the tube inlet. Samples were taken at various flow rates ranging from 0.354 to 3.54 liter/min.

The measured values of D are given in Table 1. Within the experimental errors, it is difficult to distinguish among the possible chemical species that may be present. There does appear to be some decrease in the D value of the species formed in 97 percent O_2 with 3 percent N_2, in N_2 with NO present, and in dry pump air, an indication of some addition of mass to the polonium ion. It does seem possible to distinguish clearly between charged and uncharged species. The lower values indicate that the majority of the polonium atoms present are charged. The higher value is attributed to the neutral species.

The ability of radium A to be neutralized by the direct removal of electrons from gas molecules in the atmosphere depends on either the presence of negatively charged ions in the atmosphere or the ability of the radioactive ion to ionize the gas molecules. The ionization potential of polonium metal is 8.43 eV. The ionization potentials for the gases used in this study are given in Table 2. If the ionization potential of polonium is greater than that of a particular gas molecule, the polonium can obtain an electron from the gas. Since the ionization potentials of N_2 and O_2 are 15.58 and 12.10 eV, respectively, neutralization in these gases was not expected. The similarity of the D values in dry O_2 and in the atmospheres consisting of NO in dry N_2 to the D value of radium A in the dry N_2 atmosphere (Table 1) indicates no neutralization in either case. Although neutralization was not expected in the experiment with NO_2 in dry N_2, the substantially higher D value indicates the presence of a neutral species. Since the charged polonium ion is not capable of directly removing an electron from a NO_2 molecule, it must attack the relatively labile oxygen of the NO_2 molecule to form a polonium oxide species. This oxide ion can then be neutralized by removing an electron along with the oxygen or by removing an electron from another NO_2 molecule.

The $^{218}Po^+$ ions react quickly with O_2 to form polonium dioxide ions. By analogy to the ionization potentials of metallic lead and its oxides, Busigin *et al.* (*8*) have suggested that polonium oxide would have ionization potential of the order of 10 eV. The ionization potentials of NO and NO_2 are 9.25 and 9.79 eV, respectively. If polonium dioxide is indeed formed, then it should be neutralized by the presence of NO. Therefore, we measured the D value of radium A in 8.3 ppm NO in dry 92 percent N_2 and 8 percent O_2, and neutralization was observed. Neutralization was not observed in dry air, indicating the absence of sufficient quantities of gases with low ionization potentials to permit neutralization.

Table 2. Ionization potentials of experimental gases.

Gas	Ionization potential* (eV)
N_2	15.58
O_2	12.10
NO	9.25
NO_2	9.79
H_2O	12.56
Po	8.43†

*Values obtained from (*8*). †Value obtained from (*10*).

We also examined humidity effects. Neutralization of $^{218}Po^+$ was observed at 80 percent RH in pure N_2 and partial neutralization apparently occurs at 20 percent RH, leading to an intermediate value for D. Water cannot be a direct source of electrons for polonium since its ionization potential is 12.56 eV. However, water can trap electrons from the electron-ion path created by the emitted alpha particle. Under dry conditions in N_2, the electron-ion pairs created by the emitted alpha particle rapidly recombine. The extent of neutralization depends on the number of electrons caught for a long enough period by water such that $^{218}Po^+$ can use them for neutralization. Thus, there is an increasing rate of neutralization with increasing water vapor concentration up to RH greater than 20 percent where all the ions are neutralized. This mechanism is therefore consistent with the observations of Thomas and LeClare (*5*) and Porstendorfer and Mercer (*3*). Porstendorfer and Mercer used a charged central wire mounted in a diffusion tube, similar to those used in our experiments. They examined the number of daughter atoms collected on the central wire as a function of humidity and found greater collection for moist air than for dry. They interpreted this result to mean that the ions have a higher D in moist air. We suggest that the potential field draws some of the concentration of daughter ions closer to the central wire. The ions are then neutralized by the water molecules, and the faster diffusion of the neutral molecules results in the higher apparent collection efficiency. This interpretation requires that the neutralization is sufficiently slow (millisecond) that there is some movement of the ions toward the electrode before neutralization occurs. Miller (*9*) has observed such a slow neutralization effect by water molecules in his studies of the collection of charged radon daughters.

The D value of the neutral ^{218}Po species is 0.079 cm^2/sec. This value is in agreement with the value of 0.0725 cm^2/sec reported by Kotrappa and Raghunath and the value of 0.085 cm^2/sec reported by Thomas and LeClare. We may estimate the D value of $^{218}Po^+$ from the N_2 experiment, using the percentage of occurrences of the charged and neutral species; the D value of $^{218}Po^+$ is estimated to be 0.039 cm^2/sec. The D value of the charged polonium oxide species is 0.026 cm^2/sec. This result is comparable to the value Porstendorfer and Mercer (*3*) reported for the charged oxide of ^{212}Pb.

The apparent molecular D value for ^{218}Po depends on its chemical form and the fraction of the initially formed ions that have been neutralized. There are apparently two mechanisms by which charged radium A can be neutralized: (i) the charged species accepts electrons made available by trace gases of lower ionization potential present in the environment, or (ii) the charged species neutralizes by scavenging electrons from the electron-ion path created as a result of the emission of the alpha particle by the parent nuclei when rapid recombination is inhibited because of the presence of water vapor.

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12 August 1980; revised 6 November 1980