Reports

Oxygen Isotopes: Experimental Vapor Fractionation and Variations in Tektites

Abstract. *High-temperature (about 2800°C) vapor fractionation of a silicate glass that originally contained 82 percent by weight of SiO_2 resulted in a decrease of the silica content to 45 percent. Oxygen isotope analyses show that the O^{18}/O^{16} ratio increased from 13.80 per mil in the starting material to 14.47 and 15.03 per mil in the residuum. This suggests that bediasites, which also exhibit an increase in this ratio with decreasing content of silica, have been subjected to a process of vapor fractionation.*

Taylor and Epstein (*1*) concluded that the range of oxygen isotope ratios in tektites is similar to that in granite, but that it is too low to permit the derivation of tektites from terrestrial soil. Later these authors (*2*) determined the oxygen isotope compositions of additional tektites and other materials. They found that the O^{18}/O^{16} ratio of bediasites was directly correlated with the refractive index of the glass. (In tektites, the refractive index decreases as the content of silica increases.) They suggested that the correlation between the oxygen isotope ratio and the index of refraction was due either to variations in the tektite parent material or to the high-temperature melting of the glass. These authors also performed experimental work in which they showed that the heating of soils resulted in a slight increase of the O^{18}/O^{16} ratio. The heating was carried out at 1700°C for 5 minutes, and the change in the oxygen isotope ratio was considered to be due to the loss of water which is relatively rich in O^{16}. They did not suggest, however, that such a process could account for the relations in the bediasites.

Table 1. Results of oxygen and oxygen isotope analyses. SMOW, standard mean ocean water.

Sample	Oxygen (% by wt.)	δO^{18} relative to SMOW (‰)
Indochinite	49.5	10.19
Muong Nong indochinite	49.5	10.33
Unfractionated glass	51.4	13.80
Fractionated glass, opaque	48.7	14.47
Fractionated glass, clear	49.0	15.03

More recently, Walter and Giutronich (*3*) have reported the results on a study of vapor fractionation. In this investigation, an initially "dry" synthetic glass of tektite composition was heated to approximately 2800°C for 45 minutes. The starting material contained 82 percent by weight of SiO_2, and this was reduced to 45 percent in the residuum after the material was heated and fractionated. Microprobe analysis showed that the fractionated material had a fairly uniform chemical composition, probably because of convection in that portion of the sample that was strongly heated in the solar furnace.

This fractionated portion of the material was separated from the unfractionated portion by sawing along the interface that was readily apparent between the two zones. The fractionated material was crushed and from these grains several "opaque" and several "clear" fragments were separated by hand-picking. These, together with some of the starting material, were analyzed for oxygen isotopes. In addition, duplicate analyses were made of one indochinite and one indochinite of the Muong Nong (layered) type (Table 1). The oxygen content is probably correct to within ±1 percent (relative), and the oxygen isotope ratios are probably correct to within 0.1 per mil.

Since the starting material is anhydrous, any change in the oxygen isotope ratio cannot be due to the loss of water. Therefore, fractionation of the isotopes would be due either to equilibration with atmospheric oxygen (O^{18}/O^{16} = 21 per mil) or to a greater intrinsic volatility of species containing O^{16}. The first alternative is inconsistent only with the direct-entry hypothesis of tektite origin, since, in this case, tektites could undergo thorough heating only under vacuum conditions near the moon's surface. The second alternative is possible under all proposed hypotheses of tektite origin.

Values for the two indochinites are similar to those published by Taylor and Epstein (*2*). The analytical results for the experimental material indicate a significant increase in the O^{18}/O^{16} ratio. Unfortunately, since the starting material was initially compounded by the use of materials of sedimentary origin, the initial ratio is quite high. The difference between the fractionation of synthetic material and bediasites is shown in Fig. 1. Values for tektites are those presented by Taylor and Epstein (*2*), while the SiO_2 content is derived from values for the index of refraction, presented in that paper, with the use of the relationships given by Chao (*4*). The fractionation of oxygen isotopes in the experimental materials is not as great as in the bediasites for a similar range of SiO_2 (see Fig. 1). The reason for this may be that the temperature or p_{O_2} con-

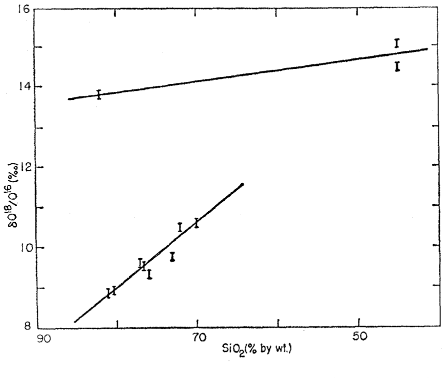

Fig. 1. Relation between the O^{18}/O^{16} ratio and the content of silica for experimental materials (upper curve) and bediasites (lower curve). Oxygen isotope data for bediasites were obtained from Taylor and Epstein (*2*). Refractive index measurements given in that paper have been converted to SiO_2 values with the use of Chao's curve (*4*).

ditions of the experiment may not duplicate exactly those at which the bediasites formed. There is no evidence in the work concerning the linearity of the fractionation trend. It is possible that the O^{18}/O^{16} ratio reaches a maximum at intermediate values for SiO_2.

However, these data show that the vapor fractionation of "dry" melts can result in an increase of the O^{18}/O^{16} ratio. This provides an explanation of the correlation of this ratio with the content of silica in bediasites and suggests that the compositional trend in these tektites is due to a process of vapor fractionation, which is in accord with conclusions based on the compositional trends of the major oxides (*5*).

Note added in proof: Recently, one of us (R.N.C.) performed oxygen-isotope analyses on soda-lime glasses that had been heated for 16 hours at 1600°C. Since this material did not exhibit any measurable change in the O^{18}/O^{16} ratio, the possibility, mentioned in this report, that the O^{18}/O^{16} increase in the bediasites is due to equilibration with the atmosphere, is unlikely. Thus, the other alternative of enhanced volatility of O^{16}-bearing species seems more likely.

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References and Notes

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6. Helpful discussion on the problem of oxygen isotope fractionation with S. Epstein of the California Institute of Technology is gratefully acknowledged. Work was supported, in part, by NSF grant GA-514.

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Climate Modification by Atmospheric Aerosols

Abstract. *Theoretical considerations and empirical evidence indicate that atmospheric turbidity, a function of aerosol loading, is an important factor in the heat balance of the earth-atmosphere system. Turbidity increase over the past few decades may be primarily responsible for the decrease in worldwide air temperatures since the 1940's.*

The possibility of deliberate or inadvertent modification of weather and climate from the emission of man-made pollutants into the atmosphere is receiving increased attention by the scientific community (*1, 2*). Of those constituents whose concentration and distribution have a bearing on the heat balance of the earth-atmosphere system, particular attention was drawn to CO_2 in a report (*3*) which concluded that the atmospheric CO_2 has been increasing over the past century and will continue to do so because of the continuing increase in combustion of fossil fuels. However, the worldwide cooling of mean annual air temperature, reported (*4, 5*) to have started between 1940 and 1950, discouraged the conclusion that there has been a direct effect on climate, that is, atmospheric warming. This enigma led to the further conclusion that " . . . climatic 'noise' from other processes has at least partially masked any effects on climate due to past increases in atmospheric CO_2 content" (*3*). Nevertheless, it was suggested that the possibility of inducing "countervailing climatic changes" should be explored by such means as raising the earth's albedo through the spreading of small buoyant reflective particles over large oceanic areas.

We may already be achieving this increased planetary reflectivity by the emission of fine particulate and aerosol pollutants into the atmosphere. The climatic significance of variations in the solar "energy albedo" of the earth by changes in the atmospheric turbidity was studied in some detail by Angstrom (*6*). Turbidity was expressed in terms of the Angstrom "turbidity coefficient" β, derived some years earlier (*7*) and defined by the empirical formula,

$$P_\lambda = \exp - \beta/\lambda^\alpha \qquad (1)$$

where P_λ is the solar transmission factor whose values range from 0 to 1 in accordance with the degree of scattering and absorption of solar radiation by atmospheric aerosol, α is called the wavelength factor (apparently related to a particle size), the reference wavelength λ is taken to be 1.0 μ, and β with limiting values lying between ∞ and 0 is related to the "dust" loading of the atmosphere. The quantity β is little affected by selective absorption of solar radiation by permanent gaseous components of the atmosphere, for example, water vapor, CO_2, and O_3, because of the techniques of determination (*7–9*).

While recognizing the dominating influence of the amount and albedo of clouds on the variations of the planetary albedo, Angstrom presented arguments to the effect that if the cloud amount remains constant, " . . . a change of 10 percent in the turbidity produces a change of about 1.5 percent in the albedo value, or about 0.8 percent in the energy available to warm the earth." Independent empirical evidence from data on solar transmission for the United States supports this conclusion (*10*). From 1962 through 1965, on days with 100 percent possible sunshine and no clouds reported (daylight hours) over Washington, D.C., and Cincinnati, Ohio, a 100 percent increase in turbidity produced a 5 percent reduction, on the average, in the transmission of solar radiation to the ground. This loss of transmission was not just that of the direct solar beam, but also that of the total hemispherical solar radiation incident on a horizontal surface, as measured by a 180-degree pyrheliometer.

The direct effect of the above process in lowering the temperature of the earth-atmosphere system cannot be determined within known limits of precision. Angstrom made a "rough estimate" (*6*) that a " . . . change of 1 percent in the albedo (from 0.40 to 0.41) corresponds to a change . . . (in the mean temperature of the earth) . . . of close to 1°C." Humphreys made similar calculations (*11*) with about the same results and also showed that the interception of outgoing radiation by fine atmospheric dusts " . . . is wholly negligible in comparison with the interception of solar radiation." Temporal and spatial changes in the atmospheric turbidity of 100 percent, that is, albedo change of 10 to 15 percent, from one day to the next or from one locality to another are very commonplace. Even though these figures may well overestimate the actual changes brought about in atmospheric temperatures, the course of atmospheric turbidity over the earth is an important climatic factor.