

NON-MASS DEPENDENT OXYGEN ISOTOPIC FRACTIONATION DURING THE  
CONDENSATION OF SIMPLE OXIDE SMOKES; Robert N. Nelson  
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A previous study has shown that non-mass dependent oxygen isotopic fractionation can occur during the condensation of refractory oxide smokes(1). Results reported in this paper support the conclusions of the previous study but raise the possibility that the specific isotopic composition of a particular smoke sample is the end product of several competing kinetic and thermodynamic factors, only some of which may have been important in the primitive solar nebula. Amorphous iron oxide and silicon oxide smokes were prepared in the Condensation Flow Apparatus(2) at GSFC by flowing either iron carbonyl or silane, respectively, plus either oxygen or nitrous oxide diluted in hydrogen through a furnace(725K<T<1000K). Products of the combustion reactions condensed both in the furnace itself and in a cooler region nominally at room temperature. Smoke samples were collected both from the furnace and from a copper collection surface. After a run the system was allowed to cool to room temperature, opened to the atmosphere, samples collected and these samples shipped to UCSD for analysis. We note that no special precautions were taken to eliminate possible atmospheric contamination of these samples prior to analysis.

Samples were analyzed using the BrF<sub>5</sub> extraction technique(3). Prior to fluorination the samples were heated to 450-500K while under vacuum for periods between 8-10 hours to remove any adsorbed water and oxygen. The oxygen yields were consistent with a stoichiometry of SiO<sub>2</sub> and FeO/Fe<sub>3</sub>O<sub>4</sub>. The results of the analysis are shown below:

SAMPLE	§170(per mil)	§180(per mil)	Δ(per mil)
SiO <sub>2</sub> (O <sub>2</sub> ) collector	-0.60	-1.53	+0.17
SiO <sub>2</sub> (O <sub>2</sub> ) "	0.15	0.43	-0.07
SiO <sub>2</sub> (O <sub>2</sub> ) "	-0.45	-0.63	-0.13
SiO <sub>2</sub> (O <sub>2</sub> ) "	-0.90	-1.57	-0.11
SiO <sub>2</sub> (O <sub>2</sub> ) furnace	-2.95	-5.66	-0.09
SiO <sub>2</sub> (O <sub>2</sub> ) "	-2.06	-4.03	-0.02
SiO <sub>2</sub> (N <sub>2</sub> O) collector	-3.52	-7.75	+0.39
SiO <sub>2</sub> (N <sub>2</sub> O) "	-3.97	-8.49	+0.32
SiO <sub>2</sub> (N <sub>2</sub> O) "	-3.28	-8.01	+0.77
SiO <sub>2</sub> (N <sub>2</sub> O) furnace	-1.55	-3.52	+0.23
SiO <sub>2</sub> (N <sub>2</sub> O) "	-0.15	-0.91	+0.31
SiO <sub>2</sub> (N <sub>2</sub> O) "	-1.20	-2.80	+0.23
SiO <sub>2</sub> (N <sub>2</sub> O) "	-0.47	-1.39	+0.23
SiO <sub>2</sub> (N <sub>2</sub> O) "	-1.62	-3.66	+0.23
FeO (O <sub>2</sub> ) collector	-5.95	-11.68	-0.05
FeO (O <sub>2</sub> ) "	-5.61	-11.40	+0.15
FeO (N <sub>2</sub> O) collector	-6.36	-13.21	+0.31
FeO (N <sub>2</sub> O) "	-6.45	-13.24	+0.24
FeO (N <sub>2</sub> O) "	-6.25	-13.11	+0.37

## OXYGEN ISOTOPIC FRACTIONATION: Nelson R. N. et al.

The data in the above table are normalized to the O<sub>2</sub> flow gas composition ( $\delta^{180}=24.97$ ,  $\delta^{170}=12.63$ ).  $\Delta$  is given by  $\delta^{170} - 0.505\delta^{180}$ , absolute value.

There are several notable features in the results. First, for the SiO<sub>2</sub> samples there is very little fractionation between the molecular O<sub>2</sub> flow gas and the SiO<sub>2</sub> product. The largest fractionation is 5.66 per mil in 180 from a furnace sample. The actual collection surface has less than 2 per mil fractionation. For the SiO<sub>2</sub> samples produced from O<sub>2</sub>, the fractionation is essentially mass dependent, as defined by its departure from the mass fractionation line,  $\Delta$ . When SiO<sub>2</sub> is prepared from N<sub>2</sub>O (which dissociates to yield free O atoms) the  $\delta^{180}$  from the collector increases to near 7 per mil as compared to near 0 for O<sub>2</sub> produced smokes under similar P,T conditions. The deviation from the mass dependent fractionation line tentatively indicates the presence of a mass independent component. Thiemens(4) has previously suggested that the reaction of O + SiO should be a mass independent process, based on symmetry considerations whereas the reaction of O<sub>2</sub> + SiO may not be.

The FeO oxidation reaction has similar features, with the product possessing a significantly larger observed fractionation than SiO<sub>2</sub>, e.g. the  $\delta^{180}$  is 6 per mil lighter for the FeO (N<sub>2</sub>O) experiments than is its SiO<sub>2</sub> counterpart. The FeO (O<sub>2</sub>) samples are nearly 10 per mil lighter than the comparable SiO<sub>2</sub> samples. Once again the N<sub>2</sub>O experiments have deviations from the mass fractionation line of approx. 0.3 per mil compared to approx. 0.1 per mil for the O<sub>2</sub> runs. Further work is required to resolve the source of the variable deviations from a simple mass fractionation which we observe in these experiments. Although it is clear that the results are reproducible, the magnitude of the effect is probably intimately connected to the details of the specific chemical pathway which produces the individual smoke as well as the possible dilution which may have occurred if the initial smoke samples were oxidized by reaction with atmospheric O<sub>2</sub> or H<sub>2</sub>O prior to isotopic analysis. We note that the oxygen isotopic composition of the N<sub>2</sub>O is currently unknown and that the thermal decomposition of the N<sub>2</sub>O may itself be somewhat anomalous(5). However there is no reason to suspect that the thermal decomposition of N<sub>2</sub>O in the presence of iron should be significantly different from the decomposition in the presence of Silane, as we observe.

References: (1) Nelson et al. Proc. LPSC XIX p559. (2) Nuth et al. in Experimental Studies of Cosmic Dust Analogs, Bussoletti et al, eds. (Kluwer, New York, 1988). (3) Clayton and Meyeda (1963) GCA 27, 43-57. (4) Thiemens, M.H. in Meteorites and the Early Solar System, Kerridge and Matthews, eds. (Univ. Ariz. Press, Tucson, 1988).