

Table 1. Ionium-thorium ratio and rate of deposition of cores from the western North Pacific Ocean.

Depth interval (cm)	Io/Th	Rate of sedimentation (mm/10 ³ yr)
<i>Sample JEDS-1-R; 30°24.6'N, 124°18.7'E; 8450 m (Ramapo Deep)</i>		
0-3	3.4 ± 0.2	0.5 ± 0.04
3-7	2.0 ± 0.1	.3 ± .04
12-15	0.2 ± 0.1	
<i>Sample JEDS-2-C₂; 42°48.0'N, 147°59.0'E; 8005 m</i>		
0-4	4.0 ± 0.2	.8 ± .05
4-8	2.6 ± 0.1	
<i>Sample JEDS-3-D₁; 29°50.0'N, 147°28.3'E; 6215 m</i>		
0-5	6.2 ± 0.5	.5 ± .04
5-10	2.8 ± 0.2	.3 ± .04
10-15	0.4 ± 0.1	

energies of 4.6 to 4.7 and 3.98 Mev, respectively, they can be measured separately with a pulse-height analyzer. Thus the abundance ratio of Th²³⁰ to Th²³² was obtained as the ratio of alpha-counts at each energy level.

The results of analyses of Io and Th in three cores of deep-sea sediments collected from the depth of 6215 to 8450 m in the western North Pacific are given in Table 1. Exponential decrease in the Io/Th ratio with the depth of burial is observed in each sample. Thus, it may be said that, during the time period of the last 100,000 yr, the sedimentation environment in this region has not differed largely from that of the present.

From these analytical results the rate of sedimentation of approximately 0.5 to 0.8 mm/10³ yr was obtained in the upper 10-cm layer below the sea bottom. These values are of the same order of magnitude that Goldberg and Koide (2) gave to the deposits of the Eastern Pacific and that Pettersson (5) gave to the sediments from near Tahiti Island in the South Pacific.

Baranov and Kuzmina (6) gave a much more rapid rate of deposition of 10 to 30 mm/10³ yr in the western region of the North Pacific. However, there are some doubts about their analytical results and the estimation of the rate of sedimentation.

According to Goldberg and Koide, the Io/Th surface ratios in the Eastern Pacific clays can be classified into two groups. One is found in the region between the Aleutian Islands and Hawaii with the value of the Io/Th ratio of about 15; the other group has the value of about 35, and is found from the region between 120°W and 140°W longitude and between 40°N and 40°S latitude. However, our results regarding the western North Pacific cores show that the Io/Th surface ratio ranges from 3 to 6, which is considerably smaller than that in other areas of the

Eastern Pacific (7). This difference is difficult to explain at present, but it may be noted that Goldberg, Patterson, and Chow (8) suggested the similarity between the distribution patterns of isotopes of lead (Pb²⁰⁰) in manganese nodules and the Io/Th ratio in the deposits in the Pacific. In this respect, they gave the extraordinarily smaller value of the lead isotope for the sea area near the Japanese islands (9, 10).

YASUO MIYAKE

YUKIO SUGIMURA

*Geochemical Laboratory,
Meteorological Research Institute,
Suginami, Tokyo, Japan*

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7. As will be reported later elsewhere, the thorium content in these samples ranges from 10 to 30 µg/g. It was also confirmed that these samples contained more than 50 percent of clayey materials with a size smaller than 2 µ. If we assume that the ratio of clayey and detritals is 1:1, and that the thorium content in detrital part is 10 µg/g which is the average value of terrestrial rocks, the ratio of thorium content in clayey and detrital minerals would be about 4:1. Therefore, even if some leaching of thorium from detrital materials would have occurred, the ratio of ionium and thorium would not have varied so much.
8. E. D. Goldberg, C. Patterson, T. Chow, *Proc. Second U.N. Intern. Conf. on Peaceful Uses of Atomic Energy* **18**, 347 (1958).
9. T. Chow and C. Patterson, *Geochim. et Cosmochim. Acta* **17**, 21 (1959).
10. This study is contribution No. 15 of the Japanese Expedition to the Deep Sea (JEDS).

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Compositional Categories of Ancient Glass

Abstract. From chemical analyses of ancient glasses found in Europe, Western Asia, and Africa from roughly the 15th century B.C. until the 12th century A.D., five main compositional categories have become apparent. With the possible exception of lead glasses which are only occasionally encountered in finds from this period, each of the main categories was prevalent over a wide geographic area for a period of at least several centuries. The categories are described in terms of the expected ranges of concentrations of the five elements, magnesium, potassium, manganese, antimony, and lead, which most clearly differentiate the groups.

A series of approximately 200 fragments and powdered samples of ancient glass has been collected through the agency of the International Committee on Ancient Glass and analyzed chemically at Brookhaven National Laboratory. The specimens were selected to

represent a cross section of glass produced in the Middle East, Africa, and Europe after the inception of hollow glass manufacture during the second millennium B.C. Our terminal date in Europe coincides with the introduction of postassium-rich compositions in about the 10th century A.D. We follow Islamic glass to the development of its splendid enameled bottles and lamps about two centuries later. The specimens were generously provided from the collections of 20 museums, universities, and related institutions, 12 active sites of archeological excavations, and several individuals. Most of these specimens were excavated under scientific supervision; others were of known provenance to the degree, at least, that the locations of their discovery were recorded. Occasionally the data on certain groups of specimens were supplemented by analysis of objects of unknown origin but showing a high degree of conformity in style. Otherwise, specimens of unknown origins have been investigated only where equally valid justification was present.

The purpose of this research has been to reveal any systematic compositional differentiations in ancient glass, whether based on regional or chronological classifications. The work represents an initial approach to a systematic technological study of ancient glass (1). We have placed the principal emphasis on the common types of clear glass, to avoid undue bias of color chemistry or the distractions of highly atypical specialties in ancient glass. Only occasionally have deliberately colored specimens been examined.

The glasses were analyzed for 26 elements by a combination of three techniques: arc spectrography, flame photometry, and colorimetry, the spectrographic plates being read quantitatively by means of a photoelectric densitometer. The resulting analyses were found to be reproducible with standard deviations within 20 percent of the reported values.

On the basis of these analyses, five main categories of ancient glass have become apparent, not considering ancient production in the Far East. Each is characterized by a chemical composition distinctly different from that of the others. A preliminary report of these groupings is presented here (2).

The most significant chemical differences between these five groups of ancient glass are presented in Table 1 and Fig. 1. No finality is claimed for these groups. Further research, for example, will surely reveal transitional and marginal groups. Within a group, a plot of the occurrence of specimens against the logarithm of concentration closely approximates a chance (Gaussian) distribution. The significance of

Table 1. Mean concentrations of the oxides that best characterize ancient glass.

Glass group	Specimens (No.)	Mean percent concentrations and standard deviation ranges				
		Magnesium MgO	Potassium K ₂ O	Manganese* MnO	Antimony Sb ₂ O ₃	Lead PbO
Second millennium B.C.	15	3.6 (4.6-2.9)	1.13 (1.89-0.69)	0.032 (0.046-0.021)	0.058 (0.32-0.011)	0.0068 (0.048-0.0010)
Antimony-rich	34	0.86 (1.24-0.60)	0.29 (0.47-0.17)	0.022 (0.035-0.014)	1.01 (1.93-0.53)	0.019 (0.077-0.0047)
Roman	73	1.04 (1.47-0.73)	0.38 (0.63-0.22)	0.41 (1.60-0.10)	0.040 (0.089-0.018)	0.014 (0.057-0.0033)
Early Islamic	66	4.9 (6.5-3.6)	1.45 (2.2-0.94)	0.47 (1.07-0.21)	0.021 (0.035-0.012)	0.0088 (0.047-0.0016)
Islamic lead	6	0.33 (0.47-0.24)	0.026 (0.051-0.013)	0.022 (0.031-0.016)	0.081 (0.19-0.035)	36 (40-33)

* Some glasses with a characteristic dark blue or violet color contain considerably more manganese than colorless glasses of the same type. It has been assumed that manganese was added deliberately to such glasses as a colorant, and they have been excluded in calculating the average manganese values.

this is that the chance deviations from the most probable concentrations are of a fractional nature. In light of this observation it is reasonable to regard the average and the standard deviation of the logarithms of concentrations as most correctly describing the probability of the occurrence of concentrations within a group. The mean concentrations so obtained are in a sense geometric means of the concentrations. The means and deviation ranges in Table 1 and Fig. 1 were obtained in this way. The lengths of the diamonds in Fig. 1 indicate the limits of the logarithmic standard deviation ranges.

Glass of the second millennium B.C. group (15th century B.C. to about 7th century B.C.) is a typical soda-lime glass with a high magnesium content, examples of which, predominantly from Egyptian finds, have been reported in earlier studies (3). It is now indicated that this composition was characteristic of glass produced or used throughout the Mediterranean area during the second millennium B.C., as our specimens come from Egypt, Mesopotamia, Greece (Mycenae), and Persia (Elam). This early group is not quite as compositionally consistent as later ones. For example, occasionally sizable concentrations of lead or antimony oxide appear to have been added to the basic glass mixtures.

Glass of the antimony-rich group (about 6th century B.C. to about 4th century A.D.) is characterized by a lower potassium and magnesium content and by the consistent appearance of antimony in high concentration. Glass of this over-all composition has not been reported previously. If we judge by the areas of the finds, it represents the composition used in Greece (Olympia), Asia Minor (Gordion), and Persia (Persepolis) during the 5th and 4th centuries B.C., and the type apparently continued to be popular in areas from the Euphrates eastward (Dura Europos and Begram in particular) during the ascendancy of Rome. Within areas dominated by

Rome this glass ceased to be prevalent, being encountered only occasionally.

The Roman group (about 4th century B.C. to 9th century A.D.) appears to represent the normal composition of "Roman glass" and was probably typical of the Syrian coastal cities, Egypt,

Italy, and the western Provinces, remaining with little change through the Frankish and Saxon periods. The composition is similar to that of the antimony-rich group except that the antimony content is much lower and in most cases the manganese content cor-

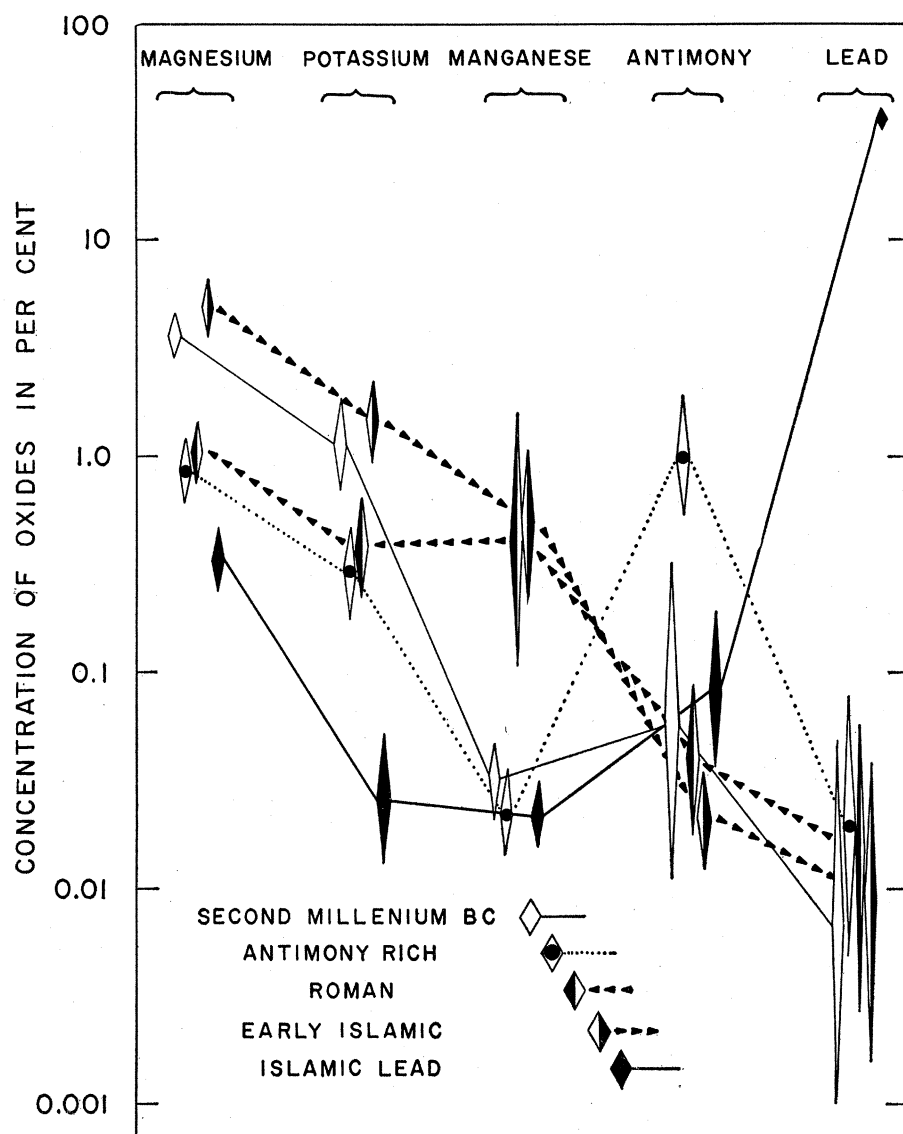


Fig. 1. Standard deviation ranges for concentrations of various metal oxides in the five main categories of Western ancient glass.

respondingly higher. This suggests the possibility that the distinction represents simply the use of one decolorant instead of another. Certainly coloration due to iron is often effectively suppressed in glasses of both compositions.

Glasses of this "Roman" composition have been reported in earlier analyses. Matson (4) pointed out the essential differences between this glass and early Egyptian glass, suggesting the possibility that the change from one composition to the other was occasioned by the advent of glassblowing. Now, however, it appears that at least in eastern areas the early glass was superseded by the antimony-rich composition several centuries before the appearance of mass-produced blown glass.

The early-Islamic group (introduced 8th–10th century A.D.) is the one comprising what might be called "Islamic soda-lime glass." It is marked by a return to the higher magnesium and potassium concentrations of the second millennium B.C. group but, in general, without showing the low manganese content of the early glass. The principal sources of these specimens included Nishapur, Susa, Quadrisia, Kish, Raqqa, Fostat, Sabrah, and various Iraqi sites represented in the collection of the Baghdad Museum.

This glass is so similar to that of the second millennium B.C. group that the assumption has been voiced that it represents an uninterrupted continuation of the same glassmaking tradition (5). The present analyses, however, would suggest that a lapse of many centuries occurred between the production of these two categories of glass. Certainly glasses of both the antimony-rich and Roman groups were produced in some of the areas destined to become Islamic. The possibility, however, that the early formulation was used without interruption in certain regions (Mesopotamia, Parthia, or Central Asia) merits investigation.

The four preceding categories represent only relatively minor variations in the basic soda-lime glass of Western antiquity. The distributions of the major components, silicon, sodium, and calcium oxide, essentially overlap for the different categories with standard deviations of the order of 25 percent of the mean values.

The identification of the Islamic lead-glass group (introduced 8th–10th century A.D.?) is based upon only six analyses, but the six glasses came from a variety of sources and yet possess remarkably similar compositions. Glass of this composition has not been singled out previously. Islamic lead-glass contains considerably more lead and less alkali and lime than most New Kingdom high-lead glass. It is distinctly

different from the lead-barium Chinese glass reported by Seligman and Beck (6) and also from the two main types of 11th–13th century A.D. Russian lead glasses described by Bezborodov (3).

Out of the 26 elements determined, only the five that show the clearest differences between the major glass categories are listed in Table 1. Some of the other elements confirm these differentiations, others are present in the same range of concentration in all except the lead glasses, and some are present in small erratic concentrations that have not yet revealed a correlation with provenance. Indications of some regional correlations within the main categories have been noted and are being explored. For example, Islamic glass from Iraq and northwestern Iran appears to have significantly less manganese than other Islamic glass. Also, high-antimony glass found at Persepolis can be differentiated from fundamentally similar and contemporary glass excavated in Greece and Asia Minor upon the basis of its relatively low contents of titanium, zirconium, and lead. The possibility of determining the concentration of additional elements, which are present in concentrations below the limits of spectrographic sensitivity, by means of neutron activation analysis is being investigated. Also, it has been possible to determine without the consumption of sample the concentrations of the five elements of Table 1 by x-ray fluorescence analysis.

The initial and terminal dates of the main groups should eventually become more precise, and some of the main compositional changes in ancient glassmaking will probably be explainable in terms of the raw materials utilized, improved furnace designs, and other shop improvements. An ultimate objective will be to pin down individual deposits and to establish trade routes in raw materials (7).

E. V. SAYRE

*Department of Chemistry,
Brookhaven National Laboratory,
Upton, Long Island, New York*

R. W. SMITH

*International Committee on Ancient
Glass, Dublin, New Hampshire*

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5. For example, W. Geilmann [*Glastech. Ber.* 28, 146 (1955)] states (p. 148): "Die Abweichung der Zusammensetzung der frühen ägyptischen von den arabischen Gläser des 14. Jahrh. n. Chr. sind nicht so gross, dass der

Schluss nicht berechtigt wäre, die Rohstoffe der Glasherstellung dürften sich in dem Zeitraum von 3000 Jahren in diesem Gebiet kaum geändert haben."

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Zinc-65 in Marine Organisms along the Oregon and Washington Coasts

Abstract. The concentration of zinc-65 in marine animals and plants near the mouth of the Columbia River is presented. Amounts of radiozinc found in the biota diminished rapidly with the distance from the river mouth. The highest levels were found in plankton, algae, and mollusks. Of the human foods, oysters exhibited the highest levels.

Zinc-65, a nonfission product commonly formed by nuclear detonations and nuclear reactors, was found in pelagic fish collected in 1954 near the Pacific Proving Grounds after the Castle series of weapon tests (1). It was subsequently reported in tuna (2), marine plankton (3), mollusks (4), and reef fishes (5) of the western Pacific. In these marine organisms Zn^{65} was responsible for up to 40 percent of the total radioactivity and in most instances was more abundant than any of the fission products. Radiozinc has also been found in marine shellfish from the Atlantic coast of the United States (6).

This radionuclide is present in Hanford reactor effluents and is one of the dominant radioisotopes in aquatic organisms inhabiting the Columbia River downstream from reactors (7). It is also found in agricultural products which are irrigated with river water taken from the Columbia at a point approximately 45 miles down river from the reactors (8, 9).

The amounts of Zn^{65} which enter the Pacific Ocean through the Columbia River are dependent on several factors, including reactor-operating levels and river flow. Concentrations of 9×10^{-9} to $1.5 \times 10^{-7} \mu\text{C}$ of Zn^{65} per cubic centimeter of Columbia River water were measured at Vancouver, Wash., located about 90 miles from the river mouth, during the last half of 1959 and the first quarter of 1960 (9, 10). Total discharge of Zn^{65} based on these measurements ranged from 8 to 55 curies of Zn^{65} per day; this would result in an equilibrium value of about 7000 curies