

laboratory values even though the samples have different carbon dioxide contents. However, the carbon dioxide content must be taken into consideration if it is desired to determine how many moles of calcium carbonate must dissolve or precipitate to saturate a sample of seawater. This can be done by an acid titration of the sample and the calculation procedures of Weyl (10).

From the data obtained during the Meteor expedition and the solubility of calcium carbonate at 1 atm, Wattenberg concluded that the deep waters of the Atlantic Ocean are nearly saturated with respect to calcite (11). Because calcite rather than aragonite is present in most sinking calcareous organisms and our results apply only to aragonite, we are limited to a tentative evaluation of Wattenberg's conclusion.

Revelle and Fairbridge estimated that the pressure coefficients of the apparent solubility products of calcite and aragonite differ by 12 percent at 1000 atm. If this is the case, the difference is within the precision of our results and the data in Table 1 represent the effect of pressure on the solubility of calcite. Because Wattenberg used the solubility at 1 atm and this increases with depth, our results suggest that the deep Atlantic waters are undersaturated rather than saturated. In this case sinking calcareous organisms should partly dissolve and the alkalinity of deep oceanic waters would increase with time. If, on the other hand, future data on the solubility of calcite at high pressures should indicate saturation at great depths, the solution of aragonitic shells would be accompanied by precipitation of calcite. Then a steady-state alkalinity is possible.

RICARDO M. PYTKOWICZ
DONALD N. CONNERS

Department of Oceanography,
Oregon State University,
Corvallis 97331

References and Notes

1. H. Wattenberg and E. Timmerman, *Ann. Hydrogr. Maritimen Meteorol.* **64**, 23 (1936).
2. R. Revelle and R. Fairbridge, *Geol. Soc. Am. Mem.* **67**, 239 (1957).
3. E-an Zen, *Geochim. Cosmochim. Acta* **12**, 103 (1957).
4. J. Lyman and R. H. Fleming, *J. Marine Res. Sears Found. Marine Res.* **3**, 134 (1940).
5. C. E. ZoBell and C. H. Oppenheimer, *J. Bacteriol.* **60**, 771 (1950). We acknowledge the use of the pressure apparatus obtained through USPHS grant AM06752-B10 awarded to Richard Y. Morita, Oregon State University.
6. R. M. Pytkowicz, in preparation.
7. K. Buch and S. Gripenberg, *J. Conseil, Conseil Perm. Intern. Exploration Mer* **7**, 233 (1932).

8. R. M. Pytkowicz, *Deep-Sea Res.* **10**, 633 (1964).
9. J. Lyman, thesis, Univ. of California, Los Angeles (1956).
10. P. K. Weyl, *J. Geol.* **69**, 43 (1961).
11. H. Wattenberg, *Wiss. Ergebn. Deut. Atlant. Exped. 'Meteor' 1925-1929*, **8**, Tl. 2, 122 (1933).
12. Supported in part by ONR contract Nonr 1286 (10), project NR 083-102, and by NSF grant GP-2566. P. K. Weyl offered valuable comments, and Marilyn George helped with the measurements.

23 March 1964

Uranium Contents of Ancient Man-Made Glass

Abstract. *The concentrations of uranium impurities in a variety of ancient man-made glasses have been determined by counting the sites of neutron-induced fissions. There appears to be a chronological trend from low to high and back to low uranium contents over the past three millennia. Crude estimates of ages by fission-track counting should be possible with considerable labor for the group of glasses which was found to have the highest concentration of uranium.*

Sayre and Smith (1) suggested that the grouping of ancient glasses according to their composition might be useful for indicating their times and places of origin. By sorting approximately 200 glasses from Europe, Africa, and eastern Asia according to their major constituents, they proposed five major compositional categories, each of which they found to be prevalent over a wide geographical area for a period of several centuries or more. In this report we consider the questions of whether the quantity of a trace impurity such as uranium follows any similar, systematic regional or chronological pattern and whether knowledge of the uranium content will make possible further subdivision of the larger groups proposed by Sayre and Smith.

Uranium is of special interest because very minute quantities ($< 10^{-18}$ weight fraction) may be readily detected by means of neutron-induced fission (2). The sites of fissions are then displayed as conical pits (such as are shown in Fig. 1) (3). The concentration, c , of uranium by weight may be found from the relation

$$c = \rho_1 u / (\sigma \phi R \delta N_0 \cos^2 \theta)$$

Where ρ_1 is the number of fission track

etch pits per square centimeter, u is the molecular weight of uranium, σ is the cross section for thermal neutron-induced fission, ϕ is the total flux of thermal neutrons, R is the range over which a fission fragment creates a track that may be etched, δ is the glass density, N_0 is Avogadro's number, and the factor $\cos^2 \theta$ results from the fact (4) that tracks are not revealed if their inclination to the etched surface is less than an angle θ . To find c , ρ_1 was measured, θ was specified by the irradiation, and $\cos^2 \theta$ was interpolated from the following experimental results: $\cos^2 \theta = 0.915, 0.75$, and 0.50 for $\text{Na}_2\text{O} + \text{K}_2\text{O}$ contents of 0 percent, 4 percent, and 14 percent. The values $R = 10^{-8}$ cm and $\delta = 2.4$ g/cm³ were assumed. Although both these quantities depend on the composition of the glass, one increases while the other decreases, so that little error is introduced by assuming their product to be constant. Although absolute values found by this procedure are probably good to no better than ± 40 percent, the relative values are considerably more precise; and our estimated error (± 15 percent) is primarily the statistical standard deviation on the fission track count (at least 100 tracks usually being counted).

A group of 34 ancient glasses was selected by Sayre and Smith from samples whose major constituents they had analyzed earlier (1, 5). Our results for uranium content as a function of age and compositional category are presented in Fig. 2 and Table 1. A list giving descriptions of the glasses and their uranium contents as measured by counts of the fission tracks is available on request. No results are given for four of the glasses, three because of the presence of crystalline phases, and one because of an excessive bubble content,

Table 1. Uranium content of ancient glasses from various categories.

Glass group*	No. of samples	Uranium (ppm by wt.)	
		Av.	Range
2nd millennium B.C.	3	0.46	0.42-0.50
High antimony	7	3.0	1.5-6.6
Roman	6	1.3	1.0-1.7
Early Islamic	10	0.7	0.2-1.7
High lead content	1	0.4	
Others	3	1.1	0.5-1.8

* Classifications are those of Sayre and Smith (see 1).

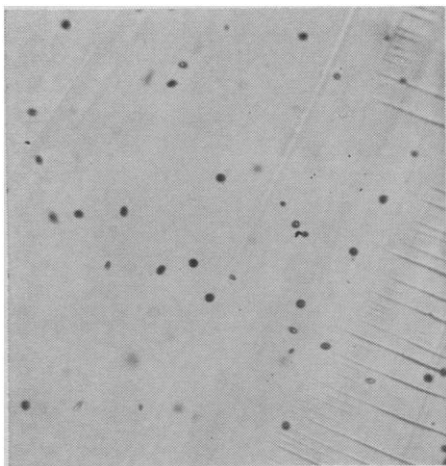


Fig. 1. Fission-track pits in a 4th to 2nd century B.C. glass from the Achæmaenid Palace at Persepolis. Irradiated with 1.3×10^{15} thermal neutrons per square centimeter; etched in hydrogen fluoride.

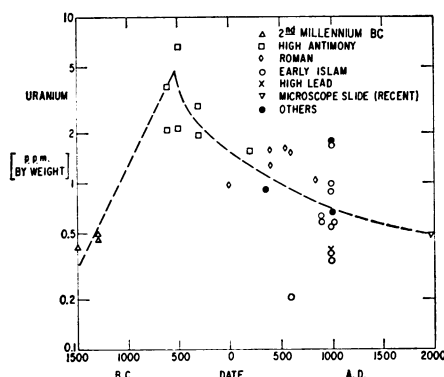


Fig. 2. Uranium contents of glasses as a function of the probable dates of origin.

both of which precluded track counting. As the figure shows, there appears to be a historical trend from initially low (2nd millennium B.C.) to high (group with high antimony content), then to progressively lower but more widely scattered values of uranium concentration. There is only one result for a lead glass, since it was found that the others contained crystalline phases. It might, at first thought, be suggested that this variation with time is associated with a similar variation of one of the major glass constituents. However, comparison of the uranium content with those of Sb, Mn, Mg, Pb, and K revealed no such correlation.

The results in Fig. 2 show that knowledge of the uranium content of glasses aids in distinguishing between some of the groups recognized by Sayre and Smith. For example, the 2nd millennium B.C. group is distinct from the high

antimony group, which in turn is distinct (except for one sample) from the other more recent groups. Whether uranium can be used to subdivide the major groups is unclear from this work. Unfortunately, the number of samples in any one group is too small to allow a statistically meaningful trend to be detected. A second difficulty arises from the fact that the true age of many of the samples is only known within ± 200 years, so that trends within groups are likely to be obscured.

In part, this study was undertaken so that we might learn whether ancient glasses commonly contain enough uranium to allow dating by counting the tracks caused by spontaneous fission (6), a procedure that has made possible the dating of natural glasses (4, 7) and certain man-made glasses to which uranium has been intentionally added (8). On the basis of the present measurements we conclude that only the group of glasses rich in antimony can be dated with a reasonable amount of work, and then rather imprecisely. For a typical such glass (3 parts per million of uranium and 2000 years old) it would be necessary to survey nearly 10 cm^2 of surface to count 16 tracks caused by spontaneous fission. Since the likely error on such a count is ± 25 percent, it should be apparent that this procedure may be a proper method of authenticating the great age of a glass but that it is not a convenient method of establishing a precise age. There are, however, exceptional glasses with a high uranium content, such as that analyzed from the 1st century villa at Cape Posilipo near Naples (9). Such material could be readily dated by the fission track method.

R. L. FLEISCHER
P. B. PRICE

General Electric Research Laboratory,
Schenectady, New York

References and Notes

1. E. V. Sayre and R. W. Smith, *Science* **133**, 1824 (1961).
2. P. B. Price and R. M. Walker, *Appl. Phys. Letters* **2**, 23 (1963).
3. R. L. Fleischer and P. B. Price, *J. Appl. Phys.* **34**, 2903 (1963).
4. ———, *J. Geophys. Res.* **69**, 331 (1964).
5. We are greatly indebted to E. V. Sayre and R. W. Smith for their generous contribution of samples and of constructive advice.
6. P. B. Price and R. M. Walker, *J. Geophys. Res.* **68**, 4847 (1963).
7. R. L. Fleischer and P. B. Price, *Geochim. Cosmochim. Acta*, in press.
8. R. H. Brill, R. L. Fleischer, P. B. Price, R. M. Walker, *J. Glass Studies*, in press.
9. J. J. Manley and E. G. Laws, *Archaeologia* **63**, 106 (1912).

23 March 1964

Radar Observations of Jupiter

The recent opposition of Jupiter has provided an opportunity for a group of engineers and scientists at the Jet Propulsion Laboratory to study that planet by radar. Experiments were conducted almost nightly from 17 October to 23 November 1963, at the Goldstone Tracking Station, located in the Mojave desert of California (1).

The parameters of the radar system were as follows: antenna gain (two-way, including losses), 108.5 db; continuous transmitter power, 100 kw; wavelength, 12.5 cm; total system noise temperature, 28°K. A continuous wave signal was transmitted to Jupiter for intervals of 1 hour and 6 minutes (the time taken by a radar signal to get to Jupiter and back). At the end of each transmission, the antenna was switched to the receiver. After another similar interval, the transmitter was again switched on. There were, during the course of the experiments, 100 such runs.

Two signal processing devices were used simultaneously: (i) total echo power through a bandpass filter was measured, with the receiver in the configuration of a Dicke radiometer, and (ii) the signal spectrum was measured by the digital equivalent of a bank of 44 contiguous bandpass filters of adjustable bandwidth.

A wave of high spectral purity was transmitted to Jupiter, but the echo was both shifted and broadened in frequency by the Doppler effect. The shift was caused by the relative orbital velocity between Jupiter and the radar station, and was accounted for by the use of an ephemeris-tuned receiver (2); while the broadening was caused by the rotation of Jupiter, which imparts differing velocities along the line of sight to different parts of the surface. The spectrum analyzer was designed to measure this broadening.

Jupiter rotates at such a rapid rate that a broadening of up to 398 kcy/sec might be observed. However, if the surface were very smooth, echoes would not be returned from the limbs, and the observed Doppler broadening would be less. If Jupiter had the same reflecting properties as Mars (3), the echo bandwidth would be 24 kcy/sec. The bandwidth would be greater if Jupiter were similar to Venus (4).

Theory suggests that optimum detection of a weak signal which is masked