

whose rate of formation is very slow. Since there are very abrupt changes in the concentrations of many compounds where the asphalt, hydrogen, and oxygen thresholds of the general C—H—O ternary diagram (Fig. 1) are crossed, it should be possible to map out experimentally the main features over the entire range of this diagram. The effects of other elements on the final composition of the systems may be similarly investigated.

Initial results with a simple carbon-hydrogen system indicate the presence of the asphalt barrier. Excitation of gases in which the ratio of hydrogen-to-carbon is less than four-to-one leads to the formation of tars consisting primarily of polynuclear aromatic compounds. Among these, pyrene, coronene, and chrysene (Fig. 5) have been identified so far; fluoranthene (Fig. 5) is also apparently present. Only minute traces of elemental carbon have been found. With water cooling we get practically all nonaromatic material, although products are at least partially unsaturated. In other words, the system appears to act as a limited equilibrium where aromatics are slow to form. This result is interesting in light of the computed results in Table 2, where aromatics have been excluded in a simulated limited equilibrium. If the hydrogen-to-carbon ratio is somewhat greater than four to one, the system appears in the hydrogen-rich region of the ternary diagram and only gaseous products are obtained. A detailed report of this work will appear at a later date. These experiments seem to demonstrate the plausibility of our use of the concept of "limited thermodynamic equilibrium."

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References and Notes

1. M. H. Studier, R. Hayatsu, E. Anders, *Science* **149**, 1455 (1965).
2. E. Anders, *Ann. N.Y. Acad. Sci.* **108**, 514 (1963); A. P. Vinogradov and G. P. Vdovkin, *Geokhimiya* **1964**, 843 (1964); W. G. Meinschein, *Space Sci. Rev.* **2**, 653 (1963); B. Nagy and D. J. Hennessy, *Ann. N.Y. Acad. Sci.* **108**, 553 (1963); N. H. Briggs and G. Mamikunian, *Space Sci. Rev.* **1**, 647 (1962-63).
3. M. O. Dayhoff, E. R. Lippincott, R. V. Eck, *Science* **146**, 1461 (1964). Professor Urey has kindly pointed out a slight error in the standard free energies used in these computations, a total of 0.38 kcal per mole for H₂, O₂, and N₂. This leads to a change by a small factor in calculated equilibrium concentrations for a few compounds. However, none of the conclusions are affected.

4. H. C. Urey and J. Lewis, *Science* **151**, 102 (1966); H. C. Urey, *ibid.*, p. 157.
5. M. H. Studier, R. Hayatsu, E. Anders, "Origin of organic matter in early solar system. I. Hydrocarbons," Enrico Fermi Inst. preprint 65-115 (1965). This work was also presented at the autumn meeting of the National Academy of Sciences, Seattle, Washington, 12 October 1965.
6. W. B. White, S. M. Johnson, G. B. Dantzig, *J. Chem. Phys.* **28**, 751 (1958).
7. D. W. van Krevelen and H. A. G. Chermin, *Chem. Eng. Sci.* **1**, 66 (1951); J. L. Franklin, *Mod. Eng. Chem.* **41**, 1070 (1949).
8. M. O. Dayhoff, E. R. Lippincott, R. V. Eck, G. Nagarajan, NASA report, in preparation.

9. H. E. Suess, *J. Geophys. Res.* **67**, 2029 (1962); S. L. Miller and H. C. Urey, *Science* **130**, 245 (1959).
10. F. X. Powell, O. Fletcher, E. R. Lippincott, *Rev. Sci. Instr.* **34**, 36 (1963); E. R. Lippincott, F. X. Powell, J. A. Creighton, D. G. Jones, in *Developments in Applied Spectroscopy*, vol. 3 (Plenum Press, New York, 1963).
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Mass-Yield Distribution of the Fission Products in Fallout from the 14 May 1965 Nuclear Explosion

Abstract. *Twenty single particles, separated from a 20-liter sample of rain collected in Osaka, Japan, shortly after the 14 May 1965 test explosion of the Chinese nuclear device, were analyzed radiochemically. The abundance pattern of the fission products in these particles resembled the shape of the mass-yield curve for the thermal neutron-induced fission of uranium-235, except for the facts that cesium-137 and strontium-90 were markedly depleted and the yields near the symmetric fission region appeared to be somewhat enhanced.*

The fallout of highly radioactive single particles occurs in Japan, because of the geographical location with respect to the nuclear weapons testing sites in the Asian mainland (1). Fission products appear to be highly fractionated in these particles, but there has been no attempt made in the past to investigate radiochemically the abundance patterns of the fission products in the particles.

A 20-liter sample of rain was collected on 20 May 1965 on the roof of the Radiation Research Center of Osaka Prefecture, Shinkecho, Sakai, Osaka, Japan (34°N, 135°E). The rain was evaporated to dryness and the residue was treated with a small quantity of distilled water. The slurry was spread over the surface of a filter paper (Whatman No. 40), dried, and then exposed to type KK Kodak x-ray film for 6 days. The autoradiograph thus obtained is shown on the cover of this issue.

The single particles thus located on the filter paper were punched out in circles 5 mm in diameter and further isolated by monitoring. The radius of these particles was estimated to be in the range of 5 to 15 μ . Twenty single particles were combined and were treated as follows.

Ten to 20 mg of Sr, Zr, Ru, Sb, Sn, Te, Cs, Ce, and La carriers were added to the sample and treated with HNO₃ and HClO₄, and the ruthenium was separated by distillation and further purified according to the method described by Rao (2). The residual ma-

terial was treated with HNO₃, HF, and HClO₄, evaporated to a small volume, and treated with fuming nitric acid. The strontium fraction thus obtained was further purified radiochemically (3). The sulfides of Te, Sn, and Sb were precipitated from chloride medium, separated, and further purified (4). Zirconium and the rare earths were separated from Cs by hydroxide precipitation and the Cs was precipitated as Cs₃Bi₂I₉ (5). The Zr was separated from the rare earths and yttrium by anion exchange and further purified (6). The Ce was separated from Pm and Y by iodide precipitation (6) and the Pm and Y were separated from La by cation exchange (7).

The filter paper, from which the single particles were punched out, contained the bulk of the radionuclides

Table 1. Fission products in the combined sample of 20 single particles and in the bulk of rain concentrate collected in Osaka, Japan, on 20 May 1965.

Nuclide	Half-life (d, days; y, years)	Number <i>N</i> of atoms as of 14 May 1965 (10 ⁶ N)	
		Single particles	Rain concentrate
Sr ⁸⁹	50.4 d	N.D.*	2640 ± 500
Sr ⁹⁰	28 y	89 ± 30	47200 ± 5000
Y ⁹¹	59 d	138 ± 45	4500 ± 1000
Zr ⁹⁵	65 d	131 ± 30	3880 ± 400
Ru ¹⁰⁶	1.0 y	8 ± 4	590 ± 100
Sn ¹²³	125 d	N.D.*	160 ± 80
Sb ¹²⁵	2.7 y	5 ± 3	750 ± 200
Te ^{127m}	105 d	21 ± 10	600 ± 100
Cs ¹³⁷	30 y	48 ± 25	37300 ± 4000
Ce ¹⁴⁴	285 d	132 ± 25	2880 ± 400
Pm ¹⁴⁷	2.7 y	23 ± 7	3700 ± 800

* N.D., not detected.

from the rain water concentrate. One-half of the filter paper was treated with a mixture of HNO_3 , HCl , and HF , and was used for the analysis of Sr, Te, Sb, Sn, Zr, Ce, Pm, Y, and Cs by the method described above. In this sample the ruthenium was separated from one quarter of the filter paper which was fused by the $\text{NaOH} + \text{Na}_2\text{O}_2$. The two samples were correlated by using Ce as a reference element.

Tracerlab Low Background Beta

Counting System (CE-14SL) and the Pancake-type (Anton 1007-TA) low-level counting system were used for the radioactivity measurements. The results are shown in Table 1 and Fig. 1.

In Fig. 1, the solid and dashed curves represent the mass-yield curves for the thermal neutron-induced fission of U^{235} (see legend to Fig. 1). The experimental value for Cs^{137} from particles falls far below the mass-yield curve, and the value for Sr^{90} also falls

somewhat below the curve. It may be possible that cesium and strontium were leached from the particles by rainwater, but it appears more likely that the depletions of Cs^{137} and Sr^{90} in the particles can be explained as due to the fact that the half-lives of gaseous precursors of these fission products are fairly long (Xe^{137} , 3.9 minutes; Kr^{90} , 33 seconds). Either they are not incorporated as much in single particles which start their growth immediately after the nuclear reaction, or perhaps these large particles were formed by early attachment of vaporized bomb products to unvaporized surface debris thrown up by the explosion. It is interesting to recall that Sr^{89} and Ba^{140} were markedly enriched relative to other fission products in rain collected at Fayetteville, Arkansas, 10 days after the 16 October 1964 test explosion of the first Chinese nuclear device (8). This would seem to indicate that much smaller fallout particles were transported several thousand miles away to Fayetteville, and most of these particles had adsorbed Sr^{89} and Ba^{140} after the decay of Kr^{89} (half-life, 3.2 minutes) and Xe^{140} (half-life, 16 seconds).

The experimental values for Sb^{125} and Te^{127} are somewhat above the mass-yield curve for thermal neutron-induced fission of U^{235} . This, of course, can be explained as owing to the fact that the trough is expected to be shallower for the mass-yield curve for nuclear devices as compared with thermal neutron-induced fission. The fact that the experimental value for Ce^{144} lies slightly above the mass-yield curve is due to its 285-day half-life, and suggests that some of the Ce^{144} was remaining from prior atmospheric testing.

The open circles and open triangles in Fig. 1 show the values for the bulk of fission products not incorporated in single particles. The half-lives of the former are less than about 1 year and they were produced almost exclusively in the 14 May 1965 nuclear explosion. The half-lives of the nuclides denoted by open triangles are longer than about 1 year and they were considered to have been remaining from the U.S.S.R. and U.S. test series prior to 1963. The half-lives of Sr^{90} and Cs^{137} are the longest of them all, and the values for these two nuclides lie far above the second mass-yield curve for the bulk of fission products shown in Fig. 1 as the dashed curve.

The radiochemical analysis described

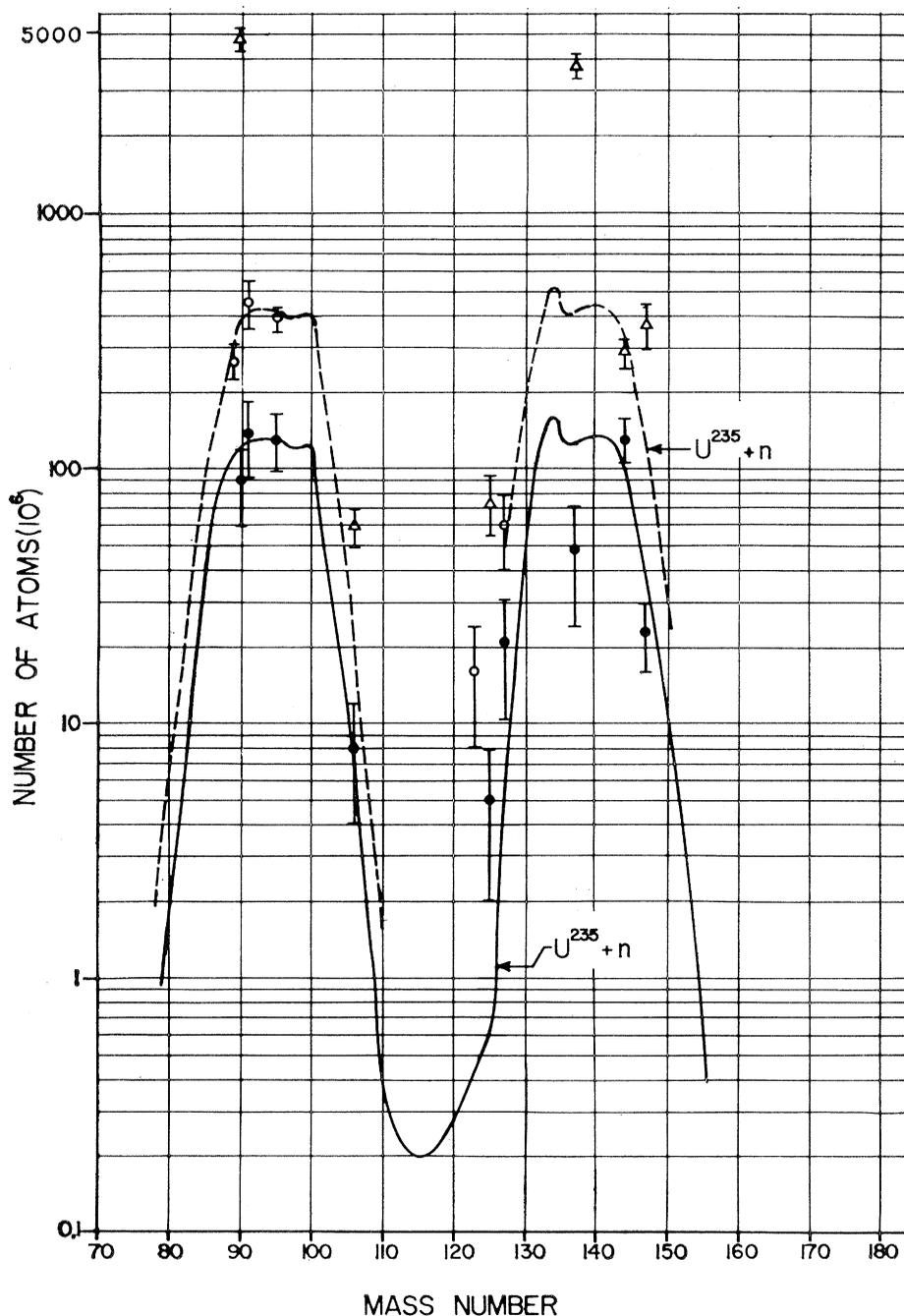


Fig. 1. Abundance patterns of the fission products in 20 single particles (solid curve) and in the bulk of rain water concentrate (dashed curve) collected in Osaka, Japan, on 20 May 1965. Verticals with filled circles, values for the combined sample of 20 single particles; verticals with open circles, values for the bulk of rain water concentrate (short-lived nuclides); verticals with open triangles, values for the bulk of rain water concentrate (nuclides with half-lives longer than about 0.8 year).

above was performed almost 300 days after the fallout particles were collected, and the radioactivities were many orders of magnitude less than the levels shortly after the nuclear explosion. Nevertheless, the experimental results presented in this report demonstrate the usefulness of this type of experiments. It is clear from these experiments that one single particle, instead of 20 particles, can easily be used to study the fission mass-yield pattern if the analysis is done shortly after the nuclear explosion.

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References and Notes

1. T. Mamuro, K. Yoshikawa, N. Maki, *Health Phys.* **11**, 199 (1965); T. Mamuro, K. Yoshikawa, T. Matsunami, A. Fujita, *Annual Report of the Radiation Center of Osaka Pre-*

- lecture, Osaka, Japan*, vol. 5, p. 16 (1964); T. Mamuro, K. Yoshikawa, T. Matsunami, A. Fujita, T. Azuma, *Nature* **197**, 964 (1963); T. Mamuro, A. Fujita, T. Matsunami, K. Yoshikawa, T. Azuma, *ibid.* **194**, 643 (1962); T. Mamuro, A. Fujita, T. Matsunami, K. Yoshikawa, *ibid.* **196**, 529 (1962); P. K. Kuroda, Y. Miyake, J. Nemoto, *Science* **150**, 1289 (1965).
2. M. N. Rao and C. J. Shahani, *J. Inorg. Nucl. Chem.* **27**, 2679 (1965).
 3. P. K. Kuroda, *Argonne Nat. Lab. Rept. ANL-5920* (Oct. 1958), p. 1.
 4. D. C. Hoffman, F. O. Lawrence, W. R. Daniels, *Los Alamos Sci. Lab. Rept. LA-1721*, No. 491 (1964).
 5. H. B. Evans, *Procedure No. 5, The Radiochemistry of Cesium* (NAS-NRC Nuclear Science Series), p. 44.
 6. C. D. Coryell and N. Sugarman, Eds., *Radiochemical Studies: The Fission Products* (McGraw-Hill, New York, 1951).
 7. H. L. Smith and D. C. Hoffman, *J. Inorg. Nucl. Chem.* **3**, 243 (1956).
 8. P. K. Kuroda, B. D. Palmer, M. Attrep, J. N. Beck, R. Ganapathy, D. D. Sabu, M. N. Rao, *Science* **147**, 1284 (1965).
 9. Supported by the U.S. Atomic Energy Commission, contract No. AT-(40-1)-2529. One of us (K.Y.) was on leave from the Radiation Research Center of the Osaka Prefecture, Osaka, Japan, during the period September 1965 through August 1966, and is grateful to Dr. Tetsuo Mamuro and his group.

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Doppler Interpretation of Quasar Red Shifts

Abstract. *The hypothesis that the quasistellar sources (quasars) are local objects moving with velocities close to the speed of light is examined. Provided there is no observational cutoff on apparent bolometric magnitude for the quasars, the transverse Doppler effect leads to the expectation of fewer blue shifts than red shifts for an isotropic distribution of velocities. Such a distribution also yields a function $N(z)$, the number of objects with red shift less than z which is not inconsistent with the present data. On the basis of two extreme assumptions concerning the origin of such rapidly moving sources, we computed curves of red shift plotted against magnitude. In particular, the curve obtained on the assumption that the quasars originated from an explosion in or nearby our own galaxy is in as good agreement with the observations as the curve of cosmological red shift plotted against magnitude.*

Arp (1) has suggested that the quasars are not located at cosmological distances, as previously argued (2), but rather that they appear to be associated with a class of peculiar galaxies falling within a range of distances of 10 to 100 megaparsecs from our own galaxy. If further investigation supports this interpretation, the large red shifts of these objects cannot be cosmological in origin. In that event, there remain two explanations. Either the red shifts are due to large gravitational fields in the emitting regions of the quasars, or they are Doppler shifts caused by large velocities of the objects.

If the shifts are gravitational, the quasars must be extremely dense for their sizes. Taking one second of arc as an upper limit to the angular diameter of the quasars, we find that their radii would range from 30 to 300 parsecs if we place them at the distances

suggested by Arp. For the observed shifts ($z \equiv \Delta\lambda/\lambda \approx 1$) to be gravitational, we must require

$$\frac{GM}{Rc^2} \approx \frac{\rho R^2 G}{c^2} \approx 1,$$

where R is the radius, M the mass, ρ the mean density, G the gravitational constant, and c the velocity of light. This gives a lower limit to the mean density of a quasar, $\rho > 10^{-10}$ g/cm³. The globular clusters, with which we are more familiar, have sizes of the same order of magnitude, but mean densities ranging from 10^{-18} to 10^{-20} g/cm³.

Arp's suggestion that the red shifts might be a consequence of high collapse velocities of the emitting material towards the centers of the quasars implies densities of the same order of magnitude, since the requirement that escape (and hence "collapse") veloc-

ities be close to c is the same as $GM/Rc^2 \approx 1$. We do not suggest that the gravitational hypothesis be discarded on these grounds, but rather wish to point out some logical consequences of the second, the Doppler shift, hypothesis, which is usually abandoned too easily, as judged from reading the literature.

The Doppler shift theories may be divided roughly into two classes. One can assume that the quasars are shot out with relativistic velocities from many explosion centers more or less isotropically distributed with respect to our own galaxy, or, following Terrell (3), one can postulate that the quasars have been ejected from the center of our own galaxy in perhaps just one catastrophic explosion. Burbidge and Hoyle (4) have proposed a modification of this second scheme, which would place the explosion outside our galaxy but sufficiently close so that quasars initially approaching us at the time of explosion would have long since passed us by. The discovery by Koehler (5) of absorption in atomic hydrogen by a hydrogen cloud associated with Virgo cluster and in the line of sight to quasar 3C273 casts some doubt that this object, at least, could have come from an explosion in or near our own galaxy. The motivation for postulating such a local explosion has been the lack of any observed blue shifts. One of the consequences of the Doppler shift hypothesis, however, is that even if the explosion responsible for accelerating the quasars did not occur in our own galaxy, their velocities must be so relativistic that one does not expect to see as many blue shifts as red shifts.

The theory is quite elementary. Suppose a source is moving with dimensionless velocity $\beta = v/c$ (velocity speed of light) in a direction characterized by an angle θ between its velocity vector and the line of sight from the source to the observer, so that $\theta < \pi/2$ corresponds to motion towards the observer. The Doppler shift is given by the well-known formula from special relativity,

$$1 + z = (1 - \beta \cos\theta) (1 - \beta^2)^{-\frac{1}{2}} \quad (1)$$

This formula displays the so-called "transverse" Doppler effect, which is a consequence of the relativistic time dilation. Even for a source with no component of velocity toward or away from the observer, there is a red shift. For any value of β , there is a critical