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EXTINCT RADIOACTIVITIES: TRAPPED RESIDUALS OF
PRESOLAR GRAINS

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ABSTRACT

The xenon anomalies trapped in meteorites and the Moon may have first been trapped in circumstellar grains formed in or outside of postexplosive stars. In that case, the initial solar nebula need not have contained most of their radioactive progenitors, and this would necessitate major revision of the history of solar system formation.

Subject headings: meteorites, meteors — nucleosynthesis — solar system

I. INTRODUCTION

In this paper I propose a new interpretation of the extinct radioactivities, which at present consist of ^{129}I , ^{244}Pu , and perhaps a superheavy nucleus. The decays of these nuclei have resulted in documented anomalies within the isotopic composition of the trapped xenon daughters, where their presence is noticeable due to the relative scarcity of average xenon in solid objects. It is the conventional view that these now-extinct radioactive nuclei existed in the gas of the primeval solar system, and that a portion of them did not decay until after the solidification of the meteorites. On this picture one gains information concerning the absolute and relative solidification times of the meteorites after the isolation of the solar nebula; specifically, nucleosynthesis added to the solar gas until about 10^8 years before meteorite solidification and, furthermore, the various meteorites solidified at the end of that period within a relatively short 14-million-year interval (Podosek 1970). I will argue that both conclusions may be incorrect due to a faulty premise in this conventional point of view.

I propose instead that the extinct radioactivities were trapped and decayed within interstellar grains formed in part in the circumstellar envelopes of exploding stars. If these grains can trap the xenon atoms resulting from the radioactive decays, there will be a reservoir of *evidence of extinct radioactivities* of abnormally high concentration in interstellar space. These grains concentrate in regions of star formation and could have been mixed rather uniformly through the early solar nebula. On this picture the radioactive atoms did not themselves exist in the solar nebula, and their concentrations reveal meteorite chronology not in the way that is commonly supposed, but rather in a different way that I will suggest. For purposes of this paper, I will assume that all extinct radioactivities are of this type except for whatever portion of ^{244}Pu that can be shown to have actually existed in the solar system, although a mixture of the conventional picture and the one I propose is also possible. I will first dis-

cuss key points of interpretation of each anomalous xenon component.

II. IODINE-129

Jeffery and Reynolds (1961) performed stepwise release of anomalous $^{129}\text{Xe}^*$ and showed that it is correlated with the ^{128}Xe created by neutron irradiation of ^{127}I within the meteoritic samples. This demonstration, showing that the $^{129}\text{Xe}^*$ is linearly associated with iodine in the meteorites, refuted the suggestion of Eberhardt and Geiss (1960) that the decay was not *in situ* but was instead accreted ^{129}Xe -rich gas from a xenon-rich atmosphere. This argument has since been taken as proof that ^{129}I existed in the solar system and that the $^{129}\text{Xe}^*$ anomaly measures the relative solidification times of the meteorites. This last conclusion has not been totally justified, because the stepwise-release experiments are consistent with ^{129}I decay to ^{129}Xe in interstellar grains long before the solar system existed. On the picture I suggest, the almost uniform correlation exists because the grain population from which the meteorites assembled contained an almost uniform admixture of grains that had been formed prior to the solar system.

Let r_i designate the fraction of interstellar iodide-bearing grains that were formed within the matter being ejected from the events of r -process nucleosynthesis before ^{129}I ($\tau_{1/2} = 17 \times 10^6$ yr) has decayed. In these grains one may later find $^{129}\text{Xe}^*/^{127}\text{I}$ abundance ratios approaching unity (the approximate production ratio of $^{129}\text{I}/^{127}\text{I}$ in the r -process), although that ratio is diminished by partial decay, by escape of ^{129}Xe , and by any dilution of ejected iodine with interstellar iodine before grains have formed. As a guess, I have found it useful to imagine $(^{129}\text{Xe}^*/^{127}\text{I})_G \sim 0.1$ in these grains. On the other hand, the fraction $1 - r_i$ of interstellar iodide-bearing grains is characterized by much less $^{129}\text{Xe}^*$; tentatively I assume there is none, which is strictly true only if interstellar iodine atoms are pure ^{127}I .

Now let the function $f_i(x, t)$ represent the fraction

of iodide-bearing grains in the solar system that were residuals of the interstellar grains (as opposed to those assembled within the solar nebula from solar gas):

$$f_A(\mathbf{x}, t) = \frac{N_A(\text{presolar grains})}{N_A(\text{presolar grains}) + N_A'(\text{solar grains})} \quad (1)$$

It is a function of position and time, and is schematically illustrated in Figure 1 for both a refractory component (e.g., silicates) and a volatile (e.g., iodine). The value of f_A is initially near unity because (by definition) all of the grains were initially presolar. As the nebula begins to make its own grains, f_A falls to some lower value. Since a large fraction of refractory elements (aluminum, silicon, etc.) are bound up in grains, f_A never falls to a very low value for those species. The nebular gas just does not contain a sufficient preponderance of these species to eventually dominate the presolar grains. For a volatile (iodine), on the other hand, such a small fraction was trapped in interstellar grains that the solar grains N_A' eventually dominate. Figure 1 also indicates the epoch δ in this process during which the meteorites formed.

It will be clear from the foregoing that the average concentration of *anomalous* $^{129}\text{Xe}^*$ in the solar material is

$$\left\langle \frac{^{129}\text{Xe}^*}{^{127}\text{I}} \right\rangle_{\odot} = \left(\frac{^{129}\text{Xe}^*}{^{127}\text{I}} \right)_G \langle f_I(\mathbf{x}, t) r_I \rangle \approx 10^{-1} \cdot 10^{-2} \cdot 10^{-1} = 10^{-4}, \quad (2)$$

where I have chosen suggestive values of these quantities as indicated. The average $\langle f_I \rangle$ is over the position

and time interval for meteorite formation, and I have guessed it at 10^{-2} . I have also supposed for illustration that one-tenth of the interstellar iodine-bearing grains were formed in the expansion of the nucleosynthesis events; viz., $r_I \approx 0.1$. This product is proposed to result in the observed average concentration $\langle ^{129}\text{Xe}^*/^{127}\text{I} \rangle \approx 1.1 \times 10^{-4}$ (Reynolds 1967; Podosek 1970).

This picture fits the facts with wholly new chronological implications. The mean time of meteorite formation is not given by this picture relative to cessation of nucleosynthesis. Nor is the time interval for meteorite solidification, which is estimated at about 14 million years on the basis of variation of a factor 2 in observed $^{129}\text{Xe}^*/^{127}\text{I}$ ratios in individual meteorites (Podosek 1970), related in this picture to the ^{129}I half-life. The factor 2 results instead by variations of that amount in $f_I(\mathbf{x}, t)$ for different meteorites forming at different times during the transition interval, as indicated in Figure 1. The spread in meteorite ages is approximately equal to the half-life for the decline of $f_I(t): 1/\tau_{1/2} \approx (f \ln 2)^{-1} df/dt$. This change in interpretation may be essential, because some models (e.g., Cameron 1973) suggest that the meteorites must physically form in a much shorter period than 10^7 yr.

Having presented this picture, I should comment on the relevance of some of the outstanding puzzles surrounding ^{129}Xe . That all classes of meteorites seem to contain the same ratio $^{129}\text{Xe}^*/^{127}\text{I}$ requires on my picture that the assembling of meteorites from small grains has not substantially fractionated $^{129}\text{Xe}^*$ from iodine. That it is not expected to do so is confirmed by the dramatic discovery of Hohenberg and Reynolds

FIG. 1.—That fraction f_A of grains bearing species A in the solar nebula that had prior existence in the interstellar medium. The figure is schematic. It suggests that this presolar fraction remains higher for refractory minerals than for volatiles. The time of the solar Hayashi contraction relative to the epoch of meteorite formation is not given.

(1969) that preheating the enstatite chondrite Abee to 1200° C *before* neutron irradiation removed 90 percent of the xenon but did not destroy the correlation line. This remarkable property nicely fits the picture of homogeneous grain admixture, but makes it harder to justify the traditional interpretation as differences in cooling time. More experiments of this type on both natural samples and artificially prepared iodine-bearing minerals are called for.

Lewis and Anders (1975) have advanced as evidence for decay within the meteorites themselves the fact that a magnetic fraction from Orgueil shows a smaller release of $^{129}\text{Xe}^*$ near 1000° C which is isochronous with the main peak at 1200° C. The presolar-grain picture seems to therefore require that the presolar admixture of the 1000° C mineral be the same as the presolar admixture of the 1200° C mineral. Although this is a special requirement, it does not seem an extreme or unlikely one.

Unequilibrated chondrites such as Manych have a special ^{129}Xe anomaly that is not isochronous with the ordinary chondrites (Podosek and Hohenberg 1970). Manych seems an assembly of chondrules containing varying ratios for $^{129}\text{Xe}^*/^{127}\text{I}$, all of which are smaller than in the equilibrated chondrites. On the present picture this suggests that Manych was assembled from constituents that formed at less common times and/or places where $f_{\text{I}}(\mathbf{x}, t)$ was smaller than for the common chondrites; i.e., the presolar grains were a smaller and less homogeneous admixture of the total grain population. If the sense of Figure 1 is correct, these facts imply either later formation or formation at a different place. In the unequilibrated chondrite Chainpur the chondrules have less $^{129}\text{Xe}^*/^{127}\text{I}$ than has the matrix, suggesting that the matrix formed when and/or where f_{I} was greater than for the chondrules, and the two were then gently mixed. This situation is less awkward than the conventional interpretation that the chondrules formed 7.4 million years *after* the matrix.

III. PLUTONIUM-244

Since the measurement of the xenon yields in ^{244}Pu fission (Alexander *et al.* 1971) it has been certain that this fission anomaly exists in meteorites. The usual interpretation is that the ^{244}Pu decayed after the meteorites formed, but I suggest here that (most of) the ^{244}Pu may instead have fissioned in presolar grains. Let r_{U} equal the fraction of interstellar uranium-bearing grains that have formed outside the events of nucleosynthesis before ^{244}Pu has decayed. Because these elements reside in the refractory silicates, $r_{\text{U}} > r_{\text{I}}$, and I take $r_{\text{U}} \approx 1$. Because a higher fraction of the refractory elements are in presolar grains, f_{A} will be larger for them than for iodine; I guess $\langle f_{\text{U}} \rangle > 0.1$ as in Figure 1. Then

$$\left\langle \frac{^{244}\text{Pu Xe}}{^{238}\text{U}} \right\rangle = \left(\frac{^{244}\text{Pu Xe}}{^{238}\text{U}} \right)_{\text{g}} \langle f_{\text{U}}(\mathbf{x}, t) \rangle r_{\text{U}}. \quad (3)$$

The abundance ratios are usually discussed in terms of the amount of ^{244}Pu necessary to produce the ob-

served concentration of fission xenon. The abundance in the interstellar grain fraction approaches $^{244}\text{Pu}/^{238}\text{U} \approx 1$ for grains forming before Pu decay, but it is probably effectively reduced to 0.1 by xenon loss and dilution of the ejected gas. The observed concentrations in meteorites suggest instead $^{244}\text{Pu}/^{238}\text{U} \approx 2 \times 10^{-2}$ in most achondrites (Reynolds 1967) but ranging up to 9×10^{-2} in the carbonaceous chondrite Allende (Podosek and Lewis 1972). These high concentrations are easily achieved with $r_{\text{U}} \leq 1$ and $\langle f_{\text{U}} \rangle \geq 0.1$.

Since the presolar-grain picture works well, it seems worthwhile to comment on features suggested by it or clarified by it. The most important feature is that ^{244}Pu fission tracks are also found in the meteorites, so unless the presolar grains were large enough to record and retain these tracks, one must conclude that ^{244}Pu was physically present in the meteorites (Fleischer, Price, and Walker 1968). The puzzle here is that the excess fission xenon is about 25 times more abundant in the St. Severin whitlockite than would be expected from the density of fission tracks (Wasserburg, Huneke, and Burnett 1969), and the same discrepancy exists for the white inclusions within the Allende chondrite (Shirck 1974), where annealing data show that the paucity of fission tracks is not due to low track retentivity. If the fission xenon, but not the tracks, is mostly retained from presolar grains, the discrepancy is explained. In that case it is the tracks and not the fission xenon that provide the measure of continuous nucleosynthesis within the Galaxy.

Hoffman *et al.* (1971) detected terrestrial ^{244}Pu , although no more than a kilogram is expected in the entire Earth. Instead of assuming that this ^{244}Pu existed initially in the Earth, however, it may make more sense to assume that it was deposited more recently by accretion of interstellar grains as the solar system has made several hundred orbits of the Galactic center. It must often have passed through dustier environs than today, and grains formed within the past 10^9 years will be surprisingly rich in Pu, which can then be metamorphosed by crustal evolution into terrestrial minerals. In the same way, the "parentless" fission xenon found in lunar breccia (Reynolds *et al.* 1974; Behrmann, Drozd, and Hohenberg 1973) can be explained in terms of lunar accretion over the old age of the lunar surface. If $f_{\text{U}} \approx 0.1$, the interstellar grains contain 10 times or more fission xenon per ^{238}U than is found in average meteoritic material. When accreted as surficial contaminations of lunar breccia, they would reveal far too much xenon in terms of the uranium concentration, as observed. This point of view, though promising, requires detailed experiments and investigations to assess. Fission track density corresponding to $(\text{Pu}/\text{U}) \approx 0.1$ at 4.6×10^9 yr ago have been found in a whitlockite crystal in lunar breccia 14321 (Hutcheon and Price 1972), which also contains an igneous clast having a Rb-Sr age of 3.95×10^9 years. At least three possibilities suggest themselves: (1) those ^{244}Pu concentrations did exist initially and were left from nucleosynthesis, (2) the Moon accreted the large (500 by 300μ) whitlockite crystal, or (3) the early Moon accreted dust from fresh

nucleosynthesis that was set into a whitlockite crystal by an ancient violent lunar event. Each of these three possibilities is consistent, moreover, with the idea that the parentless xenon was accreted.

This entire discussion highlights one of the predictions of this model—the existence of grains in the solar system having a different age than the solar system. Rubidium-strontium or U isotope ages could be different from those expected. The U and Sr isotopic shifts could be especially dramatic. Grains accreted by the Moon are probably younger but could be older than the Moon, whereas presolar grains in the meteorites are necessarily older than the meteorites. The St. Severin whitlockite (Croizat 1974) has the highest $^{232}\text{Th}/^{235}\text{U}$ ratio ever measured, for example; and this is taken as proof of severe Th/U chemical fractionation; however, one would like to see the U isotope ratio measured for evidence of presolar grains. Although smaller $^{235}\text{U}/^{238}\text{U}$ ratios are expected in individual presolar grains, an ensemble of presolar grains might have the same average isotope ratio as that found in the solar system. This would come about if the presolar grains have the same age distribution as the uranium nuclei.

IV. COMPONENT X

Manuel, Hennecke, and Sabu (1972) have given an intriguing characterization of this anomalous xenon component, which is characterized by a ^{136}Xe -rich fission-like yield in the heaviest four isotopes and a correlated overabundance in the two lightest isotopes. Anders and Heymann (1969) argued for spontaneous fission from an extinct superheavy nucleus that could live long enough ($\tau > 10^7$ yr) to be incorporated into the meteorites at the level 6×10^{-4} (^{238}U). Nuclear systematics make it doubtful that such long-lived superheavies exist, however (Howard and Nix 1974). The presolar grain picture makes a fission interpretation more plausible, because, in terms of it, the transuranic or superheavy nucleus need live only long enough to be incorporated into grains outside the synthesizing object. This could happen in years, for example, rather than in millions of years.

The enrichment of the lightest isotopes, ^{124}Xe and ^{126}Xe , is not accounted for by fission, however, so perhaps that concept is wrong. These two nuclei exist abundantly in the ejecta from the sites of p -process nucleosynthesis, which is responsible for them (Burbidge *et al.* 1957). They would thus seem to be xenon gas accreted by grains forming in the ejecta of those objects, although perhaps the isotope ratio was modified also by fractionation. If those two isotopes are accreted, it seems natural to regard the remainder of X as also having been accreted during grain formation in circumstellar envelopes. The situation then becomes complicated by the s -process, which synthesizes the intermediate isotopes of xenon. The explosive ejecta should be deficient in the s -process isotopes relative to air. Subtraction of the s -process contributions to the xenon isotopes (Seeger, Fowler, and Clayton 1965) does produce xenon richer at $A = 124, 126, 134$, and 136 as required for X, but it also

produces xenon richer at $A = 131$ and 132 (all relative to $A = 130$), so it does not seem that this kind of renormalization is substantially involved. It may also happen that the r -process events producing X could have a deficiency of the short-time r -process relative to the long-time r -process (Seeger *et al.* 1965). A more fascinating possibility lies in the chain reaction of neutron-induced fission that will occur to some extent in the expanding transbismuth ejecta of a supernova (either in free atoms or in forming grains). Manuel *et al.* (1972) argued that the heavy components of X look like neutron-induced fission, either on ^{235}U or on the many extinct heavy radioactivities present at the time of ejection. Grains forming specifically in regions rich in p -process and in transbismuth ejecta may thus trap component X, in which case it is a very peculiar component indeed. The point is that grain formation during ejection is perhaps the key to understanding, in which case it fits the picture I propose even if an extinct superheavy is not involved.

V. ANOMALOUS OXYGEN-16

Clayton, Grossman, and Mayeda (1973) have detected a component of relatively pure ^{16}O in the carbonaceous chondrites—even higher in concentrated small inclusions. They must surely be correct in supposing that this oxygen resides primarily in refractory silicates formed outside an object ejecting ^{16}O . The importance of this discovery for the picture I have presented here is that it demonstrates that of order 1 percent (at least) of refractory solar-system grains had prior existence. That fact removes my own suppositions from the realm of arbitrary postulate. As I indicated in Figure 1, I expect $\langle f_0 \rangle \approx 0.1$ for the presolar fraction of refractory grains during the epoch of meteorite formation. It is not reasonable that all interstellar grains be ^{16}O -rich grains, however, so I would estimate $r_0 \approx 0.1$ for the interstellar fraction of ^{16}O -rich grains. This results in

$$\left\langle \frac{^{16}\text{O}^*}{^{16}\text{O}} \right\rangle_{\odot} = \langle f_0 \rangle r_0 \approx 10^{-2}, \quad (4)$$

as observed.

VI. DISCUSSION

I have presented a new framework for interpreting the extinct radioactivities. It is premature to say whether it or the picture we have traditionally assumed is the correct one. The many experiments and correlations that suggest themselves will decide. The outcome is of first rank in importance, since the correct picture sets the constraints for the formation of the solar system.

The idea of forming grains in supernovae need not be as bizarre as it at first sounds. Salpeter (1974) has argued for the existence of a cold ($T \leq 100$ K) and dense ($n \approx 10^4 \text{ cm}^{-3}$) gas in a shell surrounding supernovae where grains may form. Nor can one lightly dismiss the possibility of grain formation within the denser and deeper supernova envelope when the adiabatic expansion has cooled it to the vicinity of

10^3 K, because expansion of the hot and dense interior is a very efficient cooler that converts the thermal content to bulk cold streaming matter. The ^{16}O anomaly proves that it is possible to form grains before extensive dilution with interstellar matter.

The conventional interpretation of the extinct radioactivities contains an important philosophical point, the "proof" of continuous nucleosynthesis and the measure of its rate. Unless the picture I am giving can be disproven, we can with certainty use only natural ^{244}Pu on the Earth and fission tracks in minerals demonstrably formed in the solar system for this proof.

Many facets of the role of dust in cosmogony have been discussed in a wide-ranging paper by Cameron (1973), who apparently came close to developing the ideas of this paper. I am grateful to him for pointing out this paper to me. I appreciate also helpful conversations with E. Anders. This paper was twice presented verbally: (1) in 1974, August, at the NATO Advanced Study Institute on the Origin and Abundances of the Chemical Elements in Cambridge, England, and (2) at the 1975 Anaheim meeting of the American Physical Society (Clayton 1975). The research was supported in part by NSF and in part by NASA.

REFERENCES

- Alexander, E. C., Lewis, R. S., Reynolds, J. H., and Michel, M. C. 1971, *Science*, **172**, 837.
 Anders, E., and Heymann, D. 1969, *Science*, **164**, 821.
 Behrmann, C. J., Drozd, R. J., and Hohenberg, C. M. 1973, *Earth Planetary Sci. Letters*, **17**, 446.
 Burbidge, E. M., Burbidge, G. R., Fowler, W. A., and Hoyle, F. 1957, *Rev. Mod. Phys.*, **29**, 547.
 Cameron, A. G. W. 1973, Center for Astrophysics Preprint No. 21, to appear in *Proceedings of the Symposium on the Dusty Universe*.
 Clayton, D. D. 1975, *Bull. A.P.S.*, Ser. 2, **20**, 79.
 Clayton, R. N., Grossman, L., and Mayeda, T. K. 1973, *Science*, **182**, 485.
 Crozaz, G. 1974, *Earth Planetary Sci. Letters*, **23**, 164.
 Eberhardt, P., and Geiss, J. 1960, *Zst. Naturforschung*, **15a**, 547.
 Fleischer, R. L., Price, P. B., and Walker, R. M. 1968, *Geochim. Cosmochim. Acta*, **32**, 21.
 Hoffman, D. C., Lawrence, F. O., Mewherter, J. L., and Rourke, F. M. 1971, *Nature*, **234**, 132.
 Hohenberg, C., and Reynolds, J. H. 1969, *J. Geophys. Res.*, **74**, 6679.
 Howard, W. M., and Nix, J. 1974, *Nature*, **247**, 17.
 Hutcheon, I. D., and Price, P. B. 1972, *Science*, **176**, 909.
 Jeffery, P. M., and Reynolds, J. H. 1961, *J. Geophys. Res.*, **66**, 3582.
 Lewis, R. S., and Anders, E. 1975, *Proc. Nat. Acad. Sci.*, in press.
 Manuel, O. K., Hennecke, E. W., and Sabu, D. D. 1972, *Nature*, **240**, 99.
 Podosek, F. A. 1970, *Geochim. Cosmochim. Acta*, **34**, 341.
 Podosek, F. A., and Hohenberg, C. M. 1970, *Earth Planetary Sci. Letters*, **8**, 443.
 Podosek, F. A., and Lewis, R. S. 1972, *Earth Planetary Sci. Letters*, **15**, 101.
 Reynolds, J. H. 1967, *Ann. Rev. Nucl. Sci.*, **17**, 253.
 Reynolds, J. H., Alexander, E. C., Jr., Davis, P. K., and Srinivasan, B. 1974, *Geochim. Acta.*, **38**, 401.
 Salpeter, E. E. 1974, *Rev. Mod. Phys.*, **46**, 433.
 Seeger, P. A., Fowler, W. A., and Clayton, D. D. 1965, *Ap. J. Suppl.*, **11**, 121.
 Shirck, J. 1974, *Earth Planetary Sci. Letters*, **23**, 308.
 Wasserburg, G. J., Huneke, J. C., and Burnett, D. S. 1969, *Phys. Rev. Letters*, **22**, 1198.

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