

Scintillator Distribution in High-Speed Autoradiography

Durie and Salmon (1) describe the technique of impregnation of photographic emulsion with liquid scintillator which allows rapid autoradiographic labeling. In attempting to use this method, we found that freezing the impregnated slides without special conditions led to the formation of large ice crystals and irregularities in the frozen scintillator. They appeared even when the dioxane did not contain either 2,5-diphenyloxazole or 1,4-bis-2-(4-methyl-5-phenyloxazolyl)-benzene. It could be seen clearly with the ultraviolet lamp (UVSL-58, Product Inc., San Gabriel, California).

Crystalline pictures of the scintillator luminescence show that molecules of the fluorescent substance are distributed nonhomogeneously over the biological sample. Hence, distorted distribution of silver grains in developed emulsion layers seems to be inevitable. A crystalline structure can also be seen under light microscopy. In order to circumvent these difficulties, we attempted to obtain uniform coating of the slides with the scintillator by quick deep freezing. The slides were dipped in horizontal position into the precooled isopentane so that the surface of the slide containing the biological sample and the impregnated emulsion remained free. Isopentane was cooled by liquid nitrogen. Rapid deep freezing of the scintillator can be obtained also by spraying the substance on the previously frozen slides, as above.

Examination with an ultraviolet lamp and light microscope of the slices frozen by described techniques shows that crystals are not formed and fluorescent molecules are distributed in a homogeneous way.

We think that the suggested system of rapid freezing can contribute to more correct readings of autoradiograms prepared by the high-speed scintillation autoradiography technique.

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References and Notes

1. B. G. M. Durie and S. E. Salmon, *Science* **190**, 1093 (1975).
2. We thank Dr. Y. Meckler for his comment on using the ultraviolet light.

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The suggestion to circumvent the problem of crystal formation is good and should be helpful for workers who require cooling to low temperatures to

maximally enhance grain formation. In the original development of the high-speed scintillation autoradiographic method, moderately rapid freezing (in a deep freezer) to -85°C was used. In our laboratory minimal crystal formation resulted from this cooling procedure. However, cooling to much lower temperatures resulted in serious crystal formation and a cracking of the photographic emulsion. Increasing the concentration of 2,5-diphenyloxazole (PPO) in the scin-

tillator clearly accentuated crystal formation. An unfortunate error in the original report on scintillation autoradiography listed the concentration of PPO as 35 g, which is a very high concentration and does cause significant crystal formation. With the correct concentration of 5 g of PPO we did not find crystal formation to be a significant problem.

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Strange Xenon, Extinct Superheavy Elements, and the Solar Neutrino Puzzle

Anders and co-workers (1, 2) suggest in situ decay of a volatile superheavy element 115 (or 114, 113) to explain the enrichment of heavy Xe isotopes in mineral fractions of the Allende meteorite. We will show (i) that the scenario described by Anders *et al.* (2) cannot account for their experimental results, (ii) that these new data provide additional evidence in support of our earlier suggestion (3) that the isotopically anomalous Xe in Allende and other carbonaceous chondrites was produced prior to incorporation into meteorite minerals, (iii) that the elemental and isotopic ratios of noble gases in these meteorite minerals contain an important record of the nuclear processes that produced our chemical elements, and (iv) that this record of element synthesis offers a possible explanation for the solar neutrino puzzle (4).

In 1957 Burbidge *et al.* (5) presented a detailed theory of the synthesis of elements in stars. They showed that fusion and other nuclear reactions that are expected at different stages of stellar evolution could have produced the present

elemental and isotopic abundance patterns of all the chemical elements from the lightest element, hydrogen. According to their theory, the heavy and light isotopes of Xe were synthesized in supernova envelopes by rapid neutron capture and proton capture or (γ , n) reactions (r - and p -processes), respectively. Before reaching the supernova stage, nuclear evolution would have consumed light elements from central regions of the star, but the envelopes would remain relatively rich in hydrogen and other light elements. These light elements participate in the rapid nuclear reactions which synthesize the light and heavy Xe isotopes at the time of the supernova, H is consumed by the p -process, and neutrons for the r -process are generated by reactions of ^4He on ^{13}C and ^{21}Ne (5). Recently there have been several more detailed discussions of yields of the r - and p -processes for all stable Xe isotopes (6).

In 1972 we noted (3) that carbonaceous chondrites contained an isotopically distinct component of Xe which was enriched in the light and heavy iso-

Table 1. Elemental and isotopic ratios of noble gases in acid-treated mineral fractions of the Allende meteorite (1).

Sample	Elemental ratios					Isotopic ratios			
	^4He	^{22}Ne	^{36}Ar	^{82}Kr	^{132}Xe	$^{124}\text{Xe}/^{132}\text{Xe}$	$^{136}\text{Xe}/^{132}\text{Xe}$	$^{86}\text{Kr}/^{82}\text{Kr}$	$^{80}\text{Kr}/^{82}\text{Kr}$
3C1	6,230	3.79	122	0.254	≈ 1.00	0.00471	0.338	1.54	0.197
3CS1	7,010	4.08	122	0.241	≈ 1.00	0.00474	0.338	1.54	0.198
3CS3	11,200	6.91	106	0.225	≈ 1.00	0.00492	0.354	1.56	0.197
3C3	13,900	8.42	112	0.238	≈ 1.00	0.00491	0.354	1.56	0.196
3C2	40,100	26.4	92.6	0.176	≈ 1.00	0.00586	0.461	1.68	0.197
3CS2	47,400*	29.1*	92.2	0.169	≈ 1.00	0.00616	0.477	1.70	0.197†
3CS4	83,300	48.0	93.4	0.131	≈ 1.00	0.00727	0.583	1.91	0.190†

*Gases were extracted from 3CS2 by stepwise heating; the He and Ne released at 700° and 1600°C were excluded from the summation for reasons given by Lewis *et al.* (1). †Experimental uncertainties are ± 9 percent in these ratios because of corrections for hydrocarbon background at mass 80.

topes. We concluded that the anomalous Xe could not be produced by nuclear or fractionation processes within the meteorites and suggested that the Xe component might represent material from a nearby supernova. Lewis *et al.* (1) suggest that we were too hasty in concluding that excess light and heavy Xe isotopes were mixed before trapping in meteorite minerals, and Anders *et al.* (2) suggest that an "isotopic fractionation by an unknown mechanism took place during trapping that enriched the light isotopes of Xe and the heavy isotopes of the other four gases."

The elemental and isotopic ratios of noble gases in mineral fractions of the Allende meteorite are presented in Table 1 so that the reader can judge the merits of alternate interpretations in explaining the experimental observations. We suggest that mass fractionation cannot account for the enrichment of light Xe isotopes and heavy Kr isotopes because (i) the data show an excellent correlation between excess light Xe isotopes and excess heavy Kr isotopes, which is contrary to the definition of mass fractionation; (ii) the $^{80}\text{Kr}/^{82}\text{Kr}$ ratios do not vary inversely with the $^{86}\text{Kr}/^{82}\text{Kr}$ ratios as expected from fractionation; and (iii) the $^4\text{He}/^{22}\text{Ne}$ ratios are essentially constant in all Allende mineral fractions, in sharp contrast to the large changes expected in the $^4\text{He}/^{22}\text{Ne}$ ratios if the minerals had sampled a single, well-homogenized mixture of noble gases and fractionation had acted to alter either the $^{124}\text{Xe}/^{132}\text{Xe}$ ratios by ≈ 50 percent or the $^{86}\text{Kr}/^{82}\text{Kr}$ ratios by ≈ 25 percent.

We regard the data in Table 1 as additional evidence that the anomalous Xe component was produced in supernova envelopes prior to incorporation into meteorite minerals because (i) the *r*-process is expected to produce excess heavy Xe isotopes and excess heavy Kr isotopes, but not ^{80}Kr or ^{82}Kr (because of shielding by ^{80}Se and ^{82}Se) in supernova envelopes (5); (ii) the *p*-process is expected to produce excess light Xe isotopes and excess ^{78}Kr [hydrocarbon contamination at mass 78 prohibits any conclusions about the latter from the analyses by Lewis *et al.* (1)] in supernova envelopes (5); (iii) relatively high abundances of He and Ne are expected in minerals which trapped noble gases from supernova envelopes; (iv) the experimental results in Table 1 show a linear correlation between excess heavy Xe isotopes, excess light Xe isotopes, excess heavy Kr isotopes, and He and Ne, as expected if some Allende minerals sampled noble gases from the supernova en-

velopes; and (v) the anticorrelation between the abundances of the two lightest noble gases, He and Ne, and the three heavy noble gases, Ar, Kr, and Xe, is expected if other Allende minerals sampled noble gases from a region depleted in He and Ne (and products of the *r*- and *p*-processes)—that is, from central regions of the supernova.

The elemental and isotopic ratios of noble gases shown in Table 1 provide a very important clue to the location of the supernova. A recent study (7) showed that Xe implanted in lunar soils from the solar wind contains the anomalous Xe component from supernova envelopes and concluded that the supernova must have exploded in the immediate vicinity of the solar system. A plot of $^4\text{He}/^{132}\text{Xe}$ against $^{22}\text{Ne}/^{132}\text{Xe}$ from Table 1 yields a straight line, which passes through the point (0,0) in the manner expected if Allende minerals sampled two very different reservoirs of noble gases, one being rich in He and Ne ($^4\text{He}/^{22}\text{Ne} \approx 1650$) and the other containing Ar, Kr, and Xe but no He or Ne. It should be stressed that fractionation cannot account for the paucity of He and Ne in the latter since there is no evidence for fractionation of the He/Ne ratios or for a selective depletion of Ar or Kr from Xe in these minerals. We regard the occurrence of minerals containing Ar, Kr, and Xe, without He, Ne, or any clear indication of fractionation, as evidence that these minerals sampled noble gases from the central regions of the supernova and conclude that the early solar system contained materials with isotopic and elemental inhomogeneities expected in a supernova. These observations suggest that the entire solar system may have condensed primarily from a single, local supernova.

According to Burbidge *et al.* (5) the iron-group metals are produced in the supernova core by the equilibrium (*e*) process. We regard the iron cores of the inner planets, the iron meteorites, and the core of the sun as likely condensation products from the supernova core. Hoyle (8) has suggested that initially low He concentrations and high concentrations of the iron-group metals might explain the low flux of solar neutrinos (4), and Clayton *et al.* (9) have suggested a central black hole inside the sun to explain the solar neutrino puzzle. Either of these suggestions could be understood as remanence of the local supernova core.

More quantitative details of the noble gas record of element synthesis and its implications for the isotopic anomalies of other elements may be found in (7, 10,

11). These were summarized in our presentation at the 1976 annual meeting of the American Geophysical Union, but our work on this subject was apparently overlooked in a subsequent Research News article (12).

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Manuel and Sabu (1) have misunderstood our noble gas data (2) upon which they build their hypothesis. Our samples are mixtures of two distinct phases, chromite/carbon and Q [presumably an Fe,Ni,Cr-sulfide, insoluble in HCl-HF, but soluble in HNO₃ (3, 4)]. Each phase has its own characteristic noble gas component. Phase Q contains small (but not zero) amounts of He and Ne and large amounts of isotopically normal (essentially solar) Ar, Kr, and Xe, whereas chromite/carbon contains more He and Ne but less Ar, Kr, and Xe, all of anomalous isotopic composition. The correlations stressed by Manuel and Sabu are a trivial consequence of the fact that all our samples are mixtures of these two phases.

A crucial question is whether the above minerals and their gas components are local products from the solar nebula or exotic imports from a supernova. Manuel and Sabu have repeatedly (1, 5) stated their preference for a supernova, without giving heed to the evidence against such an origin (2, 3), which has been further strengthened by recent work (6–8). The latest form of their hypothesis, which derives the bulk of the sun and planets from a supernova remnant, is too extreme to merit discussion.

Instead, we shall recapitulate the arguments against a less extreme form of this hypothesis, which derives only the gas-bearing minerals from a supernova. There are three kinds of evidence against such a supernova origin: isotopic, chemical, and astrophysical.

Isotopic evidence. If the noble gases, with isotopic anomalies of up to 90 percent, came from a supernova, then their host minerals should be similarly anomalous. They are not. The lead in Q is isotopically identical to primordial solar-system lead to within 2 percent (9); $^{190}\text{Os}/^{184}\text{Os}$ in chromite is identical to that in terrestrial Os to within 0.7 percent (7); and carbon has a $^{12}\text{C}/^{13}\text{C}$ ratio within 1 percent of the mean terrestrial value (6, 10).

Chemical evidence. Thermodynamic and kinetic considerations show (3, 6) that chromite and Q must have formed at low temperatures ($\sim 500^\circ\text{K}$) and high pressures (on the order of 10^{-5} atm), the very opposite of conditions prevailing in a supernova envelope.

Astrophysical evidence. Because of their higher Coulomb barriers, heavier elements require higher temperatures for thermonuclear reactions, and are consequently synthesized at greater depths in an exploding supernova [see for example, figure 14 of (11)]. Thus the main constituents of the host minerals (^{12}C , ^{24}Cr , ^{56}Fe , and ^{58}Ni) originate at greatly different depths, as do the five noble gases (^2He to ^{54}Xe) and the proton and neutron capture processes that are to change their isotopic compositions. Yet chromite and carbon, whose constituents form farthest apart in this picture, contain noble gases of identical isotopic composition, identical elemental composition, and identical concentration (6), whereas chromite and Q, which are made of the same elements Fe and Cr, contain very different noble-gas components (2, 6, 12).

All these difficulties, as well as the law of parsimony, point to the solar nebula as a more plausible locale. The lack of isotopic anomalies in the host minerals is no longer a fault but a virtue, and so are the low temperatures and high pressures implied by the mineralogy.

As for the *elemental* ratios of noble gases cited by Manuel and Sabu, there is no need to invoke nuclear processes because chemical processes will do. When noble gases are trapped in a growing crystal, their proportions vary with the trapping temperature as well as the nature of the crystal. It has been shown experimentally (13) that the three heavy gases have much larger negative heats of solution than do the two light gases, and so ratios such as $^4\text{He}/^{132}\text{Xe}$ or $^{22}\text{Ne}/^{132}\text{Xe}$ rise by some 3 orders of magnitude between 400° and 700°K [see figure 9 of (2) and figure 5 of (13)]. Manuel and Sabu ignore this evidence and assume that the trapping process is completely non-selective, so that each gas component requires a separate "reservoir" of the same composition. The experimental facts clearly contradict their assumption.

Whether trapping of gases in solids can also account for *isotopic* trends, such as the enrichment in light Xe isotopes, remains an open question, as we have freely admitted (2, 3, 6, 7, 12). Cosmochemistry has lived with this problem for 15 years, and most workers, including Manuel himself (14), have assumed without proof that some sort of mass fractionation process was responsible for the observed trends. Qualitatively, our new data are no different from the previous ones, and since virtually no experimental or theoretical work has been done on this subject, the categorical statement that "mass fractionation cannot account for the enrichment of light Xe isotopes" (1) is somewhat premature. Indeed, the two brief studies available, of Xe in UO_2 (15) and BaSO_4 (16), do in fact show such enrichments. Nor is it necessary that all five gases show fractionations of the same sign. As early as 1961, Lewis (15) pointed out that trapping might involve separate stages of adsorption and surface migration to traps. Because the first step would enrich the heavy isotopes and the second step the light isotopes, the net effect would depend on the relative activation energies, and could therefore change sign somewhere between He and Xe.

Lastly, the extinct superheavy element. Our 1975 articles presented further

evidence for the hypothesis (17) that the excess heavy isotopes of Xe and Kr (incidentally, Manuel and Sabu misrepresent our views on the latter) most likely were produced by decay of an extinct superheavy element. One year and four experimental papers later (4, 6, 7, 12), we still regard this hypothesis as the best available, although we shall continue to refrain from any discovery claims until the remaining uncertainties (2, 6) have been resolved to our satisfaction. A detailed discussion of the current state of the problem, in the light of our latest results, is given in (6, 12) and will not be repeated here. We believe that further progress in this area will come only from new experiments.

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