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University of California, San Diego
La Jolla, California 92037

Title: "The Distribution and Origin of the Elements
and Their Isotopes in Nature"

Period of Contract: August 15, 1962 to August 15, 1970

Principal Investigator: Dr. Harold C. Urey [REDACTED]

Co-Investigator: Dr. Kurt Marti [REDACTED]

**There is no objection from the patent
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1. Introduction

The research on this contract is a continuation of work on the general subject of the origin of the solar system and the planets which was started some twenty years ago with the publication of a book, The Planets, Their Origin and Development, by H. C. Urey in March, 1952. In this book, many aspects of the problem were touched upon--the atmospheres of the planets, the differentiation of the planets, the origin of the meteorites, the abundances of the elements, the character of the moon, etc. At that time almost no one was paying any attention to this subject with the exception of a colleague, Harrison Brown. Since then, many people have entered the field--a couple of astronomers, G. P. Kuiper and Zdenek Kopal, many people in the field of the composition of the meteorites, some geophysicists, people interested in the atmospheres of the planets, and others. The work done under this contract is only a small fraction of the total work being done in this country and abroad. The National Aeronautics and Space Administration has entered the field and with it a large number of young people investigating the moon, the planets, atmospheres, particles in space, etc. However, the interests of the principal investigator have remained rather broad in spite of the fact that it is physically impossible to keep up with all aspects of the problem. It is also true that many conflicting ideas in regard to the many questions have been advanced.

The principal investigator has been discussing a theory for the origin of the solar system which assumes that the solar nebula broke up into gaseous spheres. Unlike a considerable number of other investigators in the field, he still believes that this is a good idea. Work in recent years has been continued in the direction of understanding this theory. The most favored origin for the moon appears to be that it was captured by the earth. It has

a particularly low density, similar to that that might be expected for solar non-volatile material. It seems improbable that one such object should have been formed in the solar system and captured by the earth. It seems far more likely that many such objects existed at one time. Due to a very considerable number of lectures being given in this country and abroad, I think this idea has been brought more effectively to the attention of students than has been true previously.

2. Facilities and Instruments

All of the work was performed in the Department of Chemistry, UCSD. The equipment used in our laboratories for these studies include:

- (a) A six inch, single focussing noble gas spectrometer with electron bombardment source, electromagnetic source and deflection magnets and with two detection systems. The instrument is operated at ultrahigh vacuum ($\sim 10^{10}$ torr) and achieves a detection limit of 3×10^{-15} cc STP per isotope.
- (b) A 12 inch solid source spectrometer, thermal ionization, single focussing with both straight and multiplier detection systems. The latter is being used in either the integrating or the pulse-counting modes.
- (c) Two single focussing ratio machines with dual detector systems which can be used for dynamic runs of gaseous samples.
- (d) A Techtron Atomic Absorption Instrument with analog output and hollow cathode lamps of a large number of chemical elements. A precision of 2% was achieved on this instrument.

These instruments are complete systems with integrated electronic and vacuum supplies.

3. Personnel

Research:

H. C. Urey, Professor of Chemistry, Principal Investigator	
V. Rama Murthy, Co-Investigator	1962 - 1965
Kurt Marti, Assistant Professor of Chemistry Co-Investigator	1965 - 1970
Ron Ostic, Postgraduate Research Chemist	1963 - 1965
Charles E. Rees, Postgraduate Research Chemist	1965 - 1967
W. Brian Clarke, Postgraduate Research Chemist	1963 - 1965
Günter W. Lugmair, Postgraduate Research Chemist	1968 - 1970
H. Roy Krouse, Visiting Professor	1967 - 1968
John S. Lewis, Graduate Student	1965 - 1968
Janet Bainbridge, Research Associate	1960 - 1962 1967 - 1969

Technical:

Don Sullivan, Principal Electronics Technician	1962 - 1970
Jack Hollon, Senior Laboratory Mechanic	1962 - 1970
Mrs. Paula Sandoval	1962 - 1969
Mrs. Arlene Schimmel	1969 - 1970
Miss Mary McCue	1962 - 1969
Mrs. Mary Irene Lord	1969 - 1970
Mr. Keith Goldman	1962 - 1970

4. Publications

- Marti, K. "Mass-Spectrometric Detection of Cosmic-Ray Produced Kr⁸¹ in Meteorites and the Possibility of Kr-Kr Dating"; Phys. Rev. Letters 18, 264-266 (1967).
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- H. C. Urey, "Dalton's Influence on Chemistry," John Dalton and the progress of science, Ed. by D.S.L. Cardwell, Manchester Univ. Press (1968), speech given at Manchester, September, 1966.
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- H. C. Urey, "Origin and Distribution of the Elements" "The Problem of Elemental Abundances," Paris Conf. Vol. 30, pp. 247-253, Ed. by L. H. Ahrens.

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- H. C. Urey, "The Moon and Its Nature," Chapter 1, US/USSR Joint Publication Editorial Board, NASA, Headquarters, Washington, D. C.

5. Research Topics - Results and Interpretation

5.1 Diamonds in Meteorites:

A very considerable amount of study on the question of diamonds in meteorites has occurred. If the diamonds have been produced by steady pressure, then large objects must have been formed in the solar system at some time. By large objects, we mean the moon and its cosmic quota of gases. It is not certain that the diamonds in meteorites are produced by steady pressure, but strong proponents of this idea have been recruited during recent years--Professor Clifford Frondel at Harvard and Professor George Kennedy at UCLA, for example. It is interesting that the Novo Urei meteorite contains about 1% of diamonds and little or no evidence for shock effects. To many, this problem is a closed one, but to me (HCU), it is not. However, it is not possible to pursue this subject at the present time except under very unpleasant conditions.

5.2 Dr. Ostic continued the calculations of Bainbridge on gas spheres, assuming imperfect gases, and found that under these conditions, pressures and temperatures appropriate for the formation of diamonds could be secured. He also investigated the isotopic composition of lead in the Tuluca iron meteorite.

Dr. Rama Murthy investigated some nuclear abundances of rare earth elements which led to negative results, and only recently have we secured sensitivities of our instruments such that effects of this kind could be detected, and positive results have been secured on gadolinium in meteoritic and lunar samples. Dr. Murthy is a professor at the University of Minnesota.

We had difficulties in this laboratory with variations in isotope abundances as measured by mass spectrometers. This lead Murthy and

Umemoto to false conclusions in regard to variations in isotopic abundances. The problem proved to be very complex. Dr. Rees succeeded in unraveling the difficulties in a paper published in 1969 in the Journal of Mass Spectroscopy and Ion Physics 3, 71-80.

Dr. Rees and Dr. Brian Clarke are at McMaster University, Hamilton, Ontario.

Dr. Makoto Shima was with us in La Jolla and did very nice work on the abundances of lithium, beryllium and boron isotopes on the earth and in meteorites. His wife, Dr. Masako Shima, investigated the abundances of isotopes in germanium and tin, and concluded that there was no effect. In fact, this work corrected some erroneous results on other elements that deceived us for some time. The Shimas have returned to Japan where they are in the University in Tokyo.

The work of this laboratory during the mid sixties is rather old, at the present time, and has been superseded by an enormous amount of work in the last few years.

5.3 Stable Isotope Variations in Meteorites (C^{12}/C^{13}):

Professor H. Roy Krouse of the University of Alberta has been with us for one year, and has been investigating the variation in the isotopic ratios of carbon in meteorites and terrestrial samples. Variations in these isotopes were observed by Clayton in Chicago some years ago, and Clayton's results have been confirmed. The Orgueil meteorite contains two varieties of carbon. The carbonate carbon is remarkably high in C^{13} , higher than corresponding samples on the earth by approximately 6%, while the carbonaceous material in this meteorite is lower than the carbonate carbon of the earth by approximately 25%. The difference between these two amounts is between 8 and 9%, and this is larger than corresponding differences on the earth, 2% or 3% being typical for this difference. Krouse has extended this to other meteorites of

this class and finds similar differences in other carbonaceous chondrites. This probably means that a mixture of material from two different sources makes up these carbonaceous chondrites. The Orgueil meteorite has minerals in it which are precisely those expected for the primitive ocean of the earth or any other planetary body before erosion and sorting of materials began. The Orgueil meteorite must have been subjected to liquid water at some time, and it looks like the sediment at the bottom of some primordial lake. The time that it has been in space, some 2 or 3 m.y., is consistent with the time expected for an object dislodged from the moon and arriving on the earth. Similar things can be said for other carbonaceous chondrites, though they are best interpreted as mixtures of the Orgueil meteorite with other types of meteoritic material, which of course might well have occurred on the surface of an object such as the moon.

5.4 Cosmogenic Rare Gases and the Cavities in Iron Meteorites:

Mr. John Lewis has written a dissertation covering two aspects of these planetary problems. In the first place, there are large holes in iron meteorites. Some people have argued that these were present in the objects before they entered the atmosphere, and this argues against the iron objects being fragments of a large iron body, since it is not possible to break iron objects in such a way that highly re-entrant holes would be produced. The principal investigator has argued that this indicates that the iron meteorites come from moderate-size objects embedded in silicate masses. Mr. Lewis was investigating the concentration of cosmic-ray-produced isotopes at various positions in an iron meteorite to determine whether these holes could possibly have been produced by melting effects as the meteorite came through the atmosphere. He investigated the abundance of the cosmogenic Ne^{21} , Ar^{38} and He^3 as a function of positions in the meteorite with this objective in view.

It turned out that all the investigated samples were located in the interior of large preatmospheric bodies and were almost completely shielded from cosmic-rays. No gradients in the spallation products were found.

Lewis also has been investigating the conditions that exist in the atmosphere of Venus using chemical calculations to account for the composition of the atmosphere in high altitudes. He found that the existence of certain compounds, HCl and HF, in the high atmosphere of Venus is consistent with equilibrium of these compounds with the rocks of Venus at a temperature of approximately 500°K. This calculation of John Lewis compares favorably with any other evidence we have in regard to the surface temperature on Venus. It, of course, cannot be regarded as exact for one does not know whether the atmosphere is exactly adiabatic or not.

5.5 Mass-Spectrometric Detection of Cosmic-Ray Produced Kr^{81} in Meteorites.

The abundances of some isotopes of the heavy rare gases in meteorites are of the order of 10^{-13} cc STP/g. The detection limit of our gas spectrometer has therefore been lowered to about 3×10^{-15} cc STP per isotope (for 2mA emission current). Efforts have been made to reduce the hydrocarbon background and memory effects.

A search for cosmic-ray-produced radioactive Kr^{81} has been carried out, because calculations indicated that the 2.1×10^5 y Kr^{81} isotope should be present with abundances of about 3×10^{-14} cc STP/g in chondrites and about 2×10^{-13} cc STP/g in Ca-rich achondrites. Because of its very small activity, this nucleus cannot be detected with counters. The Kr peaks could be resolved from the remaining small isobaric hydrocarbon background in the Kr mass region while a possible interference by the bromine isotope Br^{81} could be monitored using mass 79. The extraction blanks were 0.6×10^{-12} cc STP of

atmospheric Kr. A calcium-rich achondrite was used in this first investigation, because the elements Sr, Y and Zr are enriched by about a factor of 7 in this meteorite class. Kr^{81} was found to be present in two analyses of 0.5- and 2-g samples. The Kr^{81} peak varied with the sample size, but the small isobaric background peak remained constant. A Kr^{81} concentration of $(0.20 \pm 0.03) \times 10^{-12}$ cc STP/g was calculated. The results are given in Table 1. Kr^{81} is the first cosmic-ray-produced radioactive nucleus heavier than the Fe-Ni group found so far. It is produced by spallation reactions mainly from Sr, Y and Zr.

5.6 The Kr^{81} -Kr Dating Method

The discovery of Kr^{81} in meteorites suggested the possibility of obtaining cosmic-ray exposure ages from measured isotope ratios of Kr^{81} and the spallation yield of a stable Kr isotope. This method is applicable to any material containing the target elements Sr, Y and Zr in about stone-meteoritic abundances and with detectable stable spallation Kr, e.g. the lunar samples.

Assuming constant production rates P_M for the Kr isotopes, the concentration of Kr^{81} as a function of time is given by

$$\text{Kr}^{81}(t) = (P_{81}/\lambda_{81}) (1 - e^{-\lambda_{81}t}),$$

and the spallation yield of a stable isotope, e.g., Kr^{83} , by $(\text{Kr}^{83})_{\text{spall}}(t) = P_{83}t$. Therefore, at the time of fall, we expect a ratio

$$\frac{(\text{Kr}^{83})_{\text{spall}}(T_r)}{\text{Kr}^{81}} = \frac{P_{83}}{P_{81}} \lambda_{81} \frac{T_r}{1 - \exp(-\lambda_{81}T_r)}$$

and obtain the radiation age

$$F(T_r) \equiv \frac{T_r}{1 - \exp(-\lambda_{81}T_r)} = \frac{(\text{Kr}^{83})_{\text{spall}}}{\text{Kr}^{81}} \frac{P_{81}}{P_{83}} \frac{1}{\lambda_{81}} .$$

Table 1. Isotopic composition and concentrations of Kr and Ar in the Macibini meteorite.

Isotope	Concentration	Isotope	Concentration
Kr ⁸⁶	$(23 \pm 3) \times 10^{-12}$ cc STP/g	Ar ³⁶	$(8.0 \pm 0.5) \times 10^{-18}$ cc STP/g
Kr ⁷⁸	0.388 \pm 0.005	Ar ³⁶	1.00
Kr ⁸⁰	1.288 \pm 0.013	Ar ³⁸	1.12 \pm 0.02
Kr ⁸¹	0.0087 \pm 0.0002	Ar ⁴⁰	172 \pm 8
Kr ⁸²	2.31 \pm 0.02	(Ar ³⁸) _{spall}	1.05 \pm 0.02
Kr ⁸³	2.85 \pm 0.02		
Kr ⁸⁴	4.32 \pm 0.03		
Kr ⁸⁶	1.00		

For $T_r \gg 1/\lambda_{81}$, $F(T_r) \approx T_r$. For Macibini, we found $(\text{Kr}^{83})_{\text{spall}}/\text{Kr}^{81} = 258 \pm 10$. Using $P_{81}/P_{83} = 0.61 \pm 0.03$ and $\lambda_{81} = 3.3 \times 10^{-6} \text{ yr}^{-1}$,⁶ the radiation age is $T_r = 48 \pm 4$ Myr. The advantages of Kr-Kr dating of stone and stony-iron meteorites are similar to those of Kr^{41} - Kr^{40} dating of iron meteorites developed by Voshage and Hintenberger(9): (1) The age determination is based on the isotopic composition of one element. (2) The relative Kr spallation yields are expected to depend only slightly on the energy spectrum. In favorable cases the relative production cross sections can be determined. (3) Diffusion losses are negligible.

5.7 Trapped Xenon and Krypton and the Classification of Chondrites:

The Novo Urei meteorite has the highest concentrations of Xe and Kr found so far in a meteorite. The measured ratio $^{40}\text{Ar}/^{36}\text{Ar} = 0.35$ is a new experimental upper limit for primordial Ar, but ^{40}Ar is not corrected for the radiogenic component. Since the sample was crushed shortly before it was stored in the vacuum, adsorbed atmospheric ^{40}Ar is probably small.

The ureilites are classified among the achondrites, but Mason(13) states that although the composition of Novo Urei resembles that of an olivine-hypersthene chondrite (deficient in Ni, S, Fe, Na and Al), their closest relatives are possibly the carbonaceous chondrites with respect to carbon content. The rare gas evidence gives strong support to this view.

The abundance patterns of ^{36}Ar , ^{84}Kr and ^{132}Xe are similar for all the investigated meteorites, except for Barratta which will be discussed later. Compared with cosmic abundances (10) the ratio $^{84}\text{Kr}/^{132}\text{Xe}$ is fractionated by a factor of about 20, and the ratio $^{36}\text{Ar}/^{132}\text{Xe}$ by about 1000. Obviously, all these meteorites reveal different concentrations of the same strongly fractionated rare gas component. Signer and Suess (11) proposed the term "planetary component" in their two-component model, because the fractionation of the gases is somewhat similar in the terrestrial atmosphere, whereas Anders (12) used the term "fractionated" to avoid any implications as to the origin.

The isotopic composition of Xe_{trapped} and Kr_{trapped} is almost the same in all these meteorites. The $^{36}\text{Ar}/^{38}\text{Ar}$ ratios in meteorites are identical to the values for the terrestrial atmosphere and to that found for the unfractionated "solar" component in gas-rich meteorites. A strong elemental fractionation of "primordial" gases without isotopic alteration is inconsistent with fractionation in gravitational fields or by diffusion losses. It is best explained by selective trapping of gases from a common reservoir, probably by adsorption at low temperature, as previously suggested by Zähringer (7) and Anders (12). A classification according to the $^{132}\text{Xe}_{\text{trapped}}$ ($^{132}\text{Xe}_T$) concentration agrees perfectly with the petrological one of Van Schmus and Wood (1). Based on the grid of the latter, we can set limits for the $^{132}\text{Xe}_T$ concentrations (in $10^{-10} \text{ cm}^3 \text{ STP/g}$) of the different types:

- Type 6: $^{132}\text{Xe}_T < 1.4$;
- Type 5: $1.4 \leq ^{132}\text{Xe}_T < 3.5$;
- Type 4: $3.5 \leq ^{132}\text{Xe}_T < 10$;
- Type 3: $10 \leq ^{132}\text{Xe}_T < 40$.

The isotopic compositions of Kr and Xe in some carbonaceous and disequilibrium chondrites are reported. The following isotopic abundances of Kr and Xe in the trapped primordial heavy rare gas component are obtained:

$$^{78}\text{Kr}: ^{80}\text{Kr}: ^{82}\text{Kr}: ^{83}\text{Kr}: ^{84}\text{Kr}: ^{86}\text{Kr} = 0.01920: 0.1263: 0.6490: \\ 0.6507: 3.230: 1.000$$

and

$$^{124}\text{Xe}: ^{126}\text{Xe}: ^{128}\text{Xe}: ^{130}\text{Xe}: ^{131}\text{Xe}: ^{132}\text{Xe}: ^{134}\text{Xe}: ^{136}\text{Xe} = 0.00452: \\ 0.00406: 0.0809: 0.1613: 0.815: 1.000: 0.381: 0.320.$$

5.8 Solar-Type Xenon: A New Isotopic Composition of Xenon in the Pesyanoe Meteorite

Since the first report of a new isotopic composition of xenon in the Pesyanoe enstatite achondrite (2), additional data have been obtained (Table 2). There can be little doubt that the new isotopic composition of xenon in the Pesyanoe meteorite is due to the presence of a solar-type gas component. A report at the present time is desirable for the following reasons. (1) Solar wind gases, including probably Xe, are present in samples from the lunar surface. It will be of great interest to compare the solar wind composition on the lunar surface with solar-type gases in the Pesyanoe meteorite. (2) The fact that the neutron-rich isotopes of xenon have lower relative abundances in the Pesyanoe meteorite than in either chondritic meteorites or atmospheric xenon is important for future work on fission-type xenon.

The discrepancies of the Xe^{134} and Xe^{136} points indicate that xenon in the Pesyanoe meteorite presents a new isotopic composition. While excesses in the heavy Xe isotopes due to fission xenon have recently been found in several meteorites. Pesyanoe is the first one with a clear-cut deficiency. The 1000° fraction has even lower relative abundances. The data from a stepwise heating of the Fayetteville meteorite, another gas-rich meteorite (8), also show a temperature fraction (800°C) with deficiencies of Xe^{134} and Xe^{136} . The importance of this fact has not been realized. Closer inspection of the Pesyanoe data reveals a perfect agreement of the two sample totals, but there are differences well outside the limits of error among the three temperature fractions. The δ -values for the isotopes Xe^{132} , Xe^{134} , and Xe^{136} form nearly linear arrays of different slopes. Such a variation in the Xe released at different temperatures (variation in δ_{136}^{132} up to 7 percent) can be understood only for a system containing more than one component. In a mixture of two or

Table 2. Isotopic composition and contents of xenon ($\times 10^{-12} \text{ cm}^3$ (At standard temperature and pressure) in two samples of the Pesyanoe meteorite. The isotopic abundances are normal. $\text{Xe}^{132} = 100$ percent.

Xenon	Xe^{132}	Isotopic composition (percent)								
		Xe^{124}	Xe^{126}	Xe^{128}	Xe^{129}	Xe^{130}	Xe^{131}	Xe^{132}	Xe^{134}	Xe^{136}
Sample I	96 \pm 12	0.488 \pm 0.015	0.504 \pm 0.012	8.28 \pm 0.10	103.3 \pm 0.8	16.40 \pm 0.11	82.0 \pm 0.4	100	37.3 \pm 0.3	30.7 \pm 0
Sample II										
700° fraction	10.7	0.44 \pm 0.02	0.45 \pm 0.03	7.80 \pm 0.15	101.6 \pm 1.0	15.9 \pm 0.2	79.4 \pm 0.5	100	38.4 \pm 0.4	31.9 \pm 0
1000° fraction	24.1	0.49 \pm 0.02	0.53 \pm 0.02	8.30 \pm 0.15	104.1 \pm 0.9	16.5 \pm 0.2	81.7 \pm 0.5	100	36.5 \pm 0.3	29.5 \pm 0
1600° fraction	29.2	0.52 \pm 0.02	0.55 \pm 0.02	8.14 \pm 0.12	103.7 \pm 0.8	16.3 \pm 0.2	82.4 \pm 0.6	100	37.7 \pm 0.4	31.0 \pm 0
Total	64.0 \pm 7.7	0.495 \pm 0.022	0.526 \pm 0.022	8.14 \pm 0.14	103.5 \pm 0.9	16.3 \pm 0.2	81.6 \pm 0.5	100	37.4 \pm 0.35	30.6 \pm 0
Trapped Xe (3)		0.452	0.406	8.09	(102.4)	16.13	81.5	100	38.1	32.0
Atmospheric Xe (4)		0.357	0.335	7.14	98.3	15.17	78.8	100	38.8	33.0

more components, one component must have isotopic abundances of Xe^{134} and Xe^{136} equal to or lower than those of the 1000° fraction, and must have the trapped chondritic composition for all other isotopes. This is a new Xe component which we call "solar-type" for the following reasons. The Pesyanoe pattern is typical for gas-rich meteorites and is different from either the atmospheric or the trapped chondritic composition. Signer and Suess (3) termed this component "solar," because of a close resemblance to the solar or cosmic abundance pattern. Ratios of Kr^{84}/Xe^{132} greater than 4 are found in only a few gas-rich meteorites, and, therefore, are diagnostic for the presence of solar-type gases. Therefore, some of the Xe and a sizable fraction of the Kr must belong to this component. The Xe^{136}/Xe^{132} ratio of the 1000° fraction is used here as the best available choice for solar-type xenon. It is interesting, however, to speculate that the δ -values of solar-type Xe^{134} and Xe^{136} could be slightly more negative. This would be an indication that terrestrial atmospheric and solar-type xenon may be related to each other by a strong mass fractionating process (4). Trapped chondritic xenon, on the other hand, may possibly be related to solar-type xenon by the superposition on the latter of fission-xenon components with the required relative mass yields.

5.9 Gd Isotopic Analyses

To improve the sensitivity of the solid source mass spectrometer a new ion-source with combined thermal and electron-bombardment ionization has been built. Because of the large capture cross-section, Gd is a good indicator for thermal neutrons. The isotopic composition of gadolinium has been investigated in the meteorites, Barratta, Orgueil and Pueblito de Allende, and was found to be the same within 0.2% as the terrestrial

composition. The precision recently was further improved to below 0.1% in order to apply the technique to a study of neutrons in lunar material.

5.10

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