Iodine-129 in Terrestrial Ores

Abstract. *Xenon extracted from natural iodyrite (silver iodide) from Broken Hill, New South Wales, Australia, contains excess xenon-129 from the in situ decay of naturally occurring iodine-129 and excess xenon-128 from neutron capture on iodine-127. On the basis of the amount of radiogenic xenon-129, it is estimated that, prior to the nuclear age, terrestrial iodine contained an equilibrium ratio of iodine-129 to iodine-127 of between 3.3×10^{-15} and 2.2×10^{-15}.*

The existence of ^{129}I in the early history of the solar system has been established from measurements of the decay product, ^{129}Xe, in meteorites. Since ^{129}I has a half-life of 17×10^6 years, a negligible fraction ($\approx 10^{-80}$) of the original supply of this radioactive isotope would have survived over the 4.5×10^9 year history of the earth and the meteorites.

Radiogenic ^{129}Xe was discovered in meteorites by Reynolds (*1*) in 1960 and the I-Xe dating method has since been used to date the formation times of different meteorites (*2*), the formation times of different parts of a single meteorite (*3*), and the cooling rates of meteorites (*4*).

Although the primordial supply of ^{129}I should now be extinct, this isotope has been continually produced on earth by spontaneous fission (*5*), by spallation reactions on xenon in the upper atmosphere (*6*), and by (n,γ) and (n,2n) reactions on ^{128}Te and ^{130}Te (*7*), respectively. Since the advent of the nuclear age concentrated quantities of ^{129}I have also been produced on the earth's surface as a fission product of nuclear reactors and weapons.

Purkayastha and Martin (*5*) have set an upper limit for the ratio of ^{129}I to ^{127}I in natural iodine of $\gtrsim 10^{-8}$. In 1962 Edwards (*6*) pointed out the potential utility of ^{129}I for atmospheric or hydrologic tracer studies. He estimated that natural reactions would result in a steady-state equilibrium ratio of $^{129}\mathrm{I}/^{127}\mathrm{I}$ of $\gtrsim 10^{-14}$ prior to the nuclear age. More recently, Kohman and Edwards (*8*) estimated that natural nuclear reactions would produce a present-day marine equilibrium value of $^{129}\mathrm{I}/^{127}\mathrm{I} \approx 2.2 \times 10^{-12}$. They also noted that fallout from weapons testing in the atmosphere mixed with the natural iodine content of rainwater and aerosols to produce an $^{129}\mathrm{I}/^{127}\mathrm{I}$ ratio of $\approx 10^{-5}$ to 10^{-4}.

In order to use the present distribution of ^{129}I to follow the cycle of radioactive iodine introduced by reactors and nuclear weapons, it is essential to know the abundance of ^{129}I prior to the nuclear age. Since some terrestrial ores are highly enriched in iodine but contain only trace quantities of xenon, the amount of radiogenic ^{129}Xe present in old iodine-rich ores can be used to estimate the amount of ^{129}I that was present when the ore formed.

In this investigation the abundance and isotopic composition of xenon were measured in gases released by melting samples of iodyrite (AgI) under vacuum. The samples were from the Broken Hill district of New South Wales, Australia. This region shows evidence of two periods of metamorphism (*9*), one about 500×10^6 years ago and the other about 1700×10^6 years ago, but the occurrence of iodyrite is restricted to the oxidized zone of the ore body. Although the geologic age of Broken Hill iodyrite is not known conclusively, Lawrence (*10*) suggested that the iodyrite was formed in the Tertiary period during an extended period of peneplanation. This would have exposed the ore body to the atmosphere and to downward moving groundwater, which would take up halogen ions. The halogen compounds were later concentrated under severe arid conditions which have prevailed in this region since late Tertiary time. According to this view, the upper and lower age limits of Broken Hill iodyrite would be within the Tertiary period.

Two iodyrite samples, one weighing 1.133 g (sample 1) and the other weighing 11.240 g (sample 2), were used for our analysis. Both samples were from the same ore. Sample 1 was melted in a previously outgassed molybdenum crucible which was in a vacuum system attached directly to the mass spectrometer. The abundance and isotopic composition of the evolved xenon were measured by mass spectrometry by comparison with calibrated air spikes. The detailed procedures for gas cleanup, analysis, and data reduction are given elsewhere (*3, 7*). The low amount of the gas observed in sample 1 made it necessary to repeat the experiment with a larger sample. Sample 2 was melted under vacuum in a separate vacuum line, and the gases released were collected on a charcoal finger cooled by liquid nitrogen. The charcoal finger was sealed off, attached by means of a break seal to the vacuum system of the mass spectrometer, and the adsorbed gases were released by breaking the seal and heating the charcoal to $\approx 200°$C. The total xenon content of sample 2 was about 30 times that of sample 1. The isotopic composition of xenon from the two samples is shown in Table 1 together with the isotopic composition of xenon in the gas of a deep well (*11*) and in the atmosphere (*12*). Except at masses 128 and 129, the isotopic composition of xenon in sample 1 is identical within the limits of statistical error (± 1 standard deviation) to that of the xenon observed in deep gas wells (*11*). However, the xenon in sample 2 is identical to atmospheric xenon except at masses 128 and 129. We interpret these observations to indicate that both iodyrite samples contain excess xenon at masses 128 and 129, that sample 1 and probably sample 2

Table 1. Xenon in iodyrite and in the earth. STP, standard temperature and pressure.

Mass	Iodyrite sample 1 (1.133 g)	Deep well gas (*11*)	Iodyrite sample 2 (11.240 g)	Atmosphere (*12*)
		Isotopic ratios		
124			0.024 ± 0.003	0.024 ± 0.0002
126			0.025 ± 0.004	0.022 ± 0.0002
128	2.23 ± 0.01	0.49 ± 0.01	0.725 ± 0.003	0.470 ± 0.001
129	7.77 ± 0.08	6.67 ± 0.11	6.81 ± 0.02	6.48 ± 0.02
130	≡1.00	≡1.00	≡1.00	≡1.00
131	5.41 ± 0.04	5.43 ± 0.05	5.22 ± 0.05	5.19 ± 0.02
132	6.89 ± 0.07	6.91 ± 0.09	6.60 ± 0.05	6.59 ± 0.02
134	2.76 ± 0.03	2.75 ± 0.02	2.54 ± 0.02	2.56 ± 0.01
136	2.42 ± 0.03	2.38 ± 0.02	2.16 ± 0.01	2.17 ± 0.02
		Gas concentrations (cm^3/g, STP)		
^{130}Xe	$2.3 \pm 0.2 \times 10^{-13}$		$7.0 \pm 0.6 \times 10^{-13}$	
^{129r}Xe	$2.1 \pm 0.2 \times 10^{-13}$		$2.3 \pm 0.2 \times 10^{-12}$	
^{128r}Xe	$4.0 \pm 0.4 \times 10^{-13}$		$1.8 \pm 0.2 \times 10^{-13}$	

contain a trapped component of xenon which is from deep inside the earth, and that the higher xenon content of sample 2 is due to a component of atmospheric xenon in this sample. It seems likely that there was a larger degree of atmospheric contamination in sample 2 owing to the use of the auxiliary vacuum system for melting the sample and collecting the gases on charcoal.

The excess amounts of xenon at masses 128 and 129 are indicated as ^{128r}Xe and ^{129r}Xe, respectively, at the bottom of Table 1. The amounts of ^{128r}Xe and ^{129r}Xe were computed relative to the xenon from deep well gas for sample 1 and relative to the xenon in the atmosphere for sample 2. The concentration of ^{129r}Xe is the same in both samples in spite of the large variation in total xenon content. This ^{129r}Xe is attributed to the in situ decay of ^{129}I that was present with the more abundant ^{127}I isotope when the iodyrite formed. The concentration of ^{128r}Xe varies by about a factor of 2 in these two samples. This ^{128r}Xe is probably due to neutron capture on ^{127}I over the lifetime of the iodyrite.

A minimum value of the ^{129}I/^{127}I ratio that was incorporated into the ore can be obtained by assuming that all of the ^{129}I has since decayed to ^{129}Xe. This yields an ^{129}I/^{127}I ratio of $\geqq 2.2 \times 10^{-15}$ for sample 1 and of $\geqq 2.4 \times 10^{-15}$ for sample 2.

To estimate an upper limit for the ^{129}I/^{127}I ratio that was incorporated into the iodyrite, it is necessary to estimate a minimum age for the iodyrite. An approximate value for the minimum age of the iodyrite can be obtained from the amount of ^{128r}Xe in the ore if the neutron flux rate can be estimated during the lifetime of the ore. If the iodyrite was exposed to the typical neutron flux reported for granitic rocks, ≈ 10 neutron cm^{-2} yr^{-1} (*13*), then the excess ^{128r}Xe and the thermal neutron cross section of ^{127}I (≈ 6.2 barns) can be used to estimate the age of the iodyrite samples. This calculation yields an age of 67.7×10^6 years for sample 1 and an age of 30.5×10^6 years for sample 2, in good agreement with the age limits estimated for iodyrite (*10*). These ages yield maximum values of the ^{129}I/^{127}I ratio incorporated in the ore: $^{129}\text{I}/^{127}\text{I} \leqq 2.3 \times 10^{-15}$ for sample 1 and $^{129}\text{I}/^{127}\text{I} \leqq 3.3 \times 10^{-15}$ for sample 2.

Thus it is estimated that the iodyrite samples formed from iodine with an isotopic composition of $3.3 \times 10^{-15} \geqq {}^{129}\text{I}/^{127}\text{I} \geqq 2.2 \times 10^{-15}$. Since the value calculated for the maximum ^{129}I/^{127}I ratio depends critically on the neutron flux assumed, it should be noted that a higher neutron flux would increase our estimate of the maximum ^{129}I/^{127}I ratio. However, gas loss over geologic time would reduce the content of both ^{129r}Xe and ^{128r}Xe, and this process would cause minimum errors in the limit calculated for the maximum ^{129}I/^{127}I ratio.

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Microearthquakes in the Ahuachapan Geothermal Field, El Salvador, Central America

Abstract. *Microearthquakes occur on a steeply dipping plane interpreted here as the fault that allows hot water to circulate to the surface in the geothermal region. These small earthquakes are common in many geothermal areas and may occur because of the physical or chemical effects of fluids and fluid pressure.*

The study of microearthquakes provides a way of mapping active faults in regions with few large earthquakes (*1*, *2*). The microearthquakes discussed here, with magnitudes as small as 0 (*3*, *4*), can best be recorded by ultrasensitive, high-frequency seismographs operated at hypocentral distances of less than a few tens of kilometers. More than 10,000 microearthquakes may occur in a region where only one earthquake has been located during the same period by the U.S. Coast and Geodetic Survey (now the National Ocean Survey) with teleseismic data. Thus, microearthquake data sufficient to outline faults can often be collected in a few days, weeks, or months.

Microearthquakes are observed in many major geothermal areas studied in Iceland, California, and Japan (*3*–*6*). The epicenters typically occur within the region of geothermal alteration of the surface rocks and the foci are often between 1 and 10 km deep. Earthquakes usually occur on faults, and, in many geothermal areas, faults provide the main channel along which hot water reaches either the surface or a highly permeable horizon near the surface (*7*).

Recent data suggest that fluids present along faults play a significant role in determining where, when, and in what time sequence tectonic stress is relieved as seismic energy (*4*, *8*). Two effects of fluids may be important: pore pressure (*9*) and stress corrosion (*10*). High pore pressure reduces the frictional resistance to fracture. High stresses at crack tips increase the chemical reaction of the rock with the pore fluid and thus enhance fracturing. These effects may also lead to an increase in microearthquake activity when geothermal brines are reinjected for disposal purposes.

Three vertical-component seismometers were operated 2 to 3 km apart in the Ahuachapan geothermal area in El Salvador from 7 December 1969 to 10 August 1970 (Fig. 1). Two horizontal-component seismometers were also placed at site A. The system response had maximum gain of about 500,000 at 17 cycle/sec. All seismic signals and a time signal were connected by cable to a magnetic tape recorder situated at point A. About 500 earthquakes were recorded and analyzed during the total of 6 months that the equipment at site