Correlation between Fission Tracks and Fission Type Xenon in Meteoritic Whitlockite[1, 2]

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Whitlockite from the St. Severin chondrite, previously shown to contain excess fission tracks, is here shown to have a large concentration of excess neutron-rich xenon isotopes. The concentration of excess heavy Xe in the whitlockite is about twenty-five times that calculated from the track density. An isotopic spectrum is deduced that is identical to the spectrum calculated previously for excess heavy xenon in the Pasamonte achondrite. These results uniquely associate this xenon spectrum with in situ fission in meteorites. Chemical arguments support the correlation of this with Pu^{244}. Identification of the fissioning nucleus as Pu^{244} gives $Pu^{244}/U^{238} \approx 1/30$. Neither 'sudden' nor 'uniform' nucleosynthetic models give consistent solutions for Pu^{244}/U^{238} and U^{235}/U^{238}.

INTRODUCTION

In this paper we report a measurement of fission type xenon in whitlockite from the St. Severin amphoterite. This is correlated with fission track data and discussed in the context of the Pu^{244} hypothesis.

There are two independent lines of evidence that suggest the presence of fission products in meteorites:

1. Several meteorites contain distinct enrichments in the isotopic abundance of the neutron-rich Xe isotopes suggestive of fission [*Rowe and Kuroda,* 1965; *Kuroda et al.,* 1966; *Munk,* 1967].
2. Studies on minerals from several other meteorites have revealed high densities of charged particle tracks with properties characteristic of fission tracks rather than tracks of heavy cosmic-ray ions [*Fleischer et al.,* 1968; *Schirck et al.,* 1969; *Cantelaube et al.,* 1967].

In all cases studied, the concentrations of U appear to be inadequate to account for either the observed track densities or the amounts and isotopic composition of the excess Xe in terms of U^{238} spontaneous fission. Induced fission does not appear to be an adequate explanation because:

1. There is no independent evidence for the required particle fluxes.
2. The isotopic spectra do not match presently known fission yield curves.
3. Fission track densities in coexisting minerals are not strictly proportional to the U contents [*Fleischer et al.,* 1968].

The excess fission tracks and excess neutron-rich Xe are usually attributed to the spontaneous fission of Pu^{244} (82-m.y. half-life) during the early history of the solar system. Strong evidence already exists for the presence of I^{129} (17-m.y. half-life) at the time of formation of many meteorites [*Reynolds,* 1967]. Thus, it is reasonable that Pu^{244}, with a half-life five times greater than I^{129}, should also be present. However, one should not necessarily equate the nucleosynthetic processes for Pu^{244} with those of I^{129}. Pu^{244} can only be formed in galactic *r*-process nucleosynthesis [*Seeger et al.,* 1965], whereas I^{129} could be formed in local particle irradiations within the solar system following its isolation from the galaxy as a whole [*Fowler et al.,* 1962]. The estimated abundance of Pu^{244} relative to U^{238} is relatively high ($Pu^{244}/U^{238} \sim 0.012$–$0.1$) at the end of nucleosynthesis [*Fowler,* 1962].

Efforts have been made to correlate the excess Xe^{136} with Xe^{129} from I^{129} decay. Some auth-

[1] This paper is to be included in a collection of reprints in memory of the late Craig M. Merrihue.
[2] Contribution 1630 of the California Institute of Technology.

ors [*Kuroda et al.*, 1966] have stated that the excess Xe^{136}-U data for many meteorites define a formation interval of 3×10^9 years (assuming the Pu^{244} hypothesis), whereas the I^{127}-Xe^{129} data scatter more or less randomly. Other workers [*Kuroda*, 1967; *Sabu and Kuroda*, 1967] attempt to show a rather precise correlation between Xe^{136}/U and the time interval calculated from the Xe^{129}/I^{127} data on the same meteorites. This attempt has been seriously criticized by *Reynolds* [1968] in a detailed analysis.

The known chemical properties of Pu suggest that it would be associated with U, Th, and/or the rare earths. Some correlation between the amount of excess Xe^{136} and the U content has been found in Ca-rich achondrites [*Kuroda et al.*, 1966; *Munk*, 1967]. Studies of excess fission tracks show them to be most dense in the phases that have the highest U content and to be essentially absent in phases with negligible U [*Fleischer et al.*, 1968; *Price et al.*, 1968]. Since the observed results cannot be explained by U fission, these correlations give some support to the Pu^{244} hypothesis. However, even in the absence of any Pu-U chemical fractionation, this argument is weakened by several factors that tend to destroy a perfect Pu-U correlation:

1. Several *different*, so-called fission Xe components have been claimed to be present in meteorites.
2. Some of the fission Xe might not be the result of in situ decay.
3. Differences in the formation times of meteorites comparable to the Pu^{244} half-life would cause Pu^{244}/U^{238} variations due to Pu^{244} decay that would not correlate with U concentration.
4. Cooling rates that were not fast when compared with the lifetime of Pu^{244} would lead to time differences in the retention of daughter products (tracks or Xe) in different minerals, so that the inferred Pu/U ratios in various phases of a single meteorite may not be the same.

Fleischer et al. [1968] have attempted to relate the fission tracks to Pu^{244} by comparing cooling rates deduced from track densities in different minerals with cooling rates from metallurgical data. Their conclusions are critically dependent on the conditions of track retentivity and the assumed metallurgical cooling rate.

Although the Pu^{244}/U^{238} ratios inferred from both observations have been roughly compatible, neither the track data alone nor the excess heavy Xe is sufficient to demonstrate the existence of Pu^{244} at the time of formation of the solar system. Moreover, in the absence of an experimentally established correlation between the excess tracks and the excess Xe there is no direct proof that the same phenomena are being investigated by the two methods. In addition to this basic qualitative question, it is of great importance to establish whether or not the Pu^{244}/U^{238} values obtained by the two methods can be quantitatively compared because of problems related to track annealing, inherited fission Xe, etc. Further, for phases with a high concentration of fission tracks, the expected high enrichment of the fission component would allow a precise determination of the fission Xe spectrum. Comparison of a precise meteoritic spectrum with the laboratory Pu^{244} Xe spectrum, when this becomes available, will provide an unequivocal test of the Pu^{244} identification.

Cantelaube et al. [1967] have made detailed track measurements on the minerals from the St. Severin chondrite. The observed track densities in pyroxene and plagioclase are comparable. Pyroxene track densities show a marked dependence on distance from the preterrestrial surface of the meteorite and are anisotropic. They are identified as heavy ion tracks. However, the observed track densities in the trace mineral whitlockite, $\beta Ca_3(PO_4)_2$, are consistently higher ($7.4 \pm 1.5 \times 10^6$ cm^{-2}) and show no depth distribution. The whitlockite was found to be comparatively rich in U (0.4 ppm), but U^{238} spontaneous fission could account for only 1/7 of the tracks. Consequently, the whitlockite tracks were attributed by these workers to be Pu^{244} fission tracks.

Experimental Procedure and Data Analysis

Through the generous cooperation of P. Pellas, F. Kraut, and J. Orcel of the Museum National d'Histoire Naturelle, Paris, France, a sample of St. Severin was provided to us for study (H2402b). A 100-gram piece of St. Severin was crushed and sized into a 105- to 25-μ fraction. Forty milligrams of whitlockite were separated by both heavy liquid and magnetic separations. Microchemical tests and electron microprobe

analyses showed this separate to contain approximately 90% whitlockite by grain count, the principal contaminant being feldspar.

The total meteorite samples were loaded in the sample arm as single pieces, whereas the whitlockite and feldspar separates were wrapped in Ni foil containers. A description of the rare gas extraction system has been given previously [*Bogard et al.*, 1969*a*]. The extraction bottle, manifold, and sample arm were kept at 65°–80°C throughout the extractions.

The samples were heated by induction in a W crucible with an Al_2O_3 insert for 35 min at 1800°C, in the presence of two Ti getters at 850°C and activated charcoal cooled to −196°C. The gases adsorbed on the charcoal were released for cleanup during the latter stages of the extraction. Finally, the Ti getters were cooled consecutively to 80°C. The Xe and part of the Kr (60%) were adsorbed on charcoal at −78°C, and the sample tube was removed from the system. The remaining Kr and Ar were adsorbed onto charcoal at −196°C and removed.

The sample tubes were then sealed onto the spectrometer manifold. The Xe fraction was further cleaned by hot Ti and hot Cu-CuO-Pd getters immediately before analysis. The Kr and Xe were analyzed separately by adsorbing the Xe and part of the Kr (15% of that present in the manifold) on activated charcoal at −46°C and first admitting the remaining Kr for analysis and then the Xe. The sample manifold was maintained at 70°C throughout, and the charcoal from each sample tube was removed from the system subsequent to the sample analysis.

The gases were analyzed in an all-metal, 15-cm-radius, Nier-type mass spectrometer with an electron multiplier (gain about 3×10^5) at 2.5-kv accelerating voltage and 1-ma emission at 60 volts with a source magnet. The mass spectrometer sensitivity and mass discrimination were obtained by analyzing standard pipettes of 10^{-9} cc STP of atmospheric Xe. The sensitivity, as monitored throughout the series of sample measurements, was 6.4×10^{-15} cc STP Xe^{132}/mv with a total spread of 3% on the 10^9-Ω resistor. The mass discrimination was (0.41 ± 0.08)% per mass unit favoring the lighter masses. The analysis of a smaller quantity ($\approx 10^{-10}$ cc STP) of Xe (comparable to the amounts found in the samples) gave a similar value for mass discrimination but a 20% higher value for the sensitivity. This was not taken into account in the calculation of errors.

Isotopic ratios were obtained by extrapolation to zero time to correct for memory effects. The errors quoted for the tabulated Xe isotopic ratios reflect primarily the estimated maximum uncertainties in these zero time extrapolations. The Xe^{132} blanks listed in Table 1 are estimated from an analysis of Ni foil (6.6×10^{-11} cc STP/g foil) and the re-extraction of the empty crucible ($0.75–1.8 \times 10^{-12}$ cc STP) after the initial extractions of each sample. The amounts of non-atmospheric Xe found in the re-extractions were negligible in all cases. Hydrocarbon corrections, estimated primarily from the signal at mass 127, were negligible. The largest correction was 3% for the less abundant isotopes (124, 126, 128), and the correction was less than 0.2% for all others.

Analytical Results and Discussion

The analytical results are presented in Table 1. For comparison we have included the earlier results of *Funkhouser et al.* [1967] and *Marti et al.* [1969]. The samples investigated were mineral separates of whitlockite and feldspar and samples of whole meteorite fragments composed mostly of light and dark structure. These are visually distinctive and were not previously recognized. The Rb/Sr in the light phase is twice that in the dark.

Inspection of the results for all samples (particularly the whitlockite) shows that they have a xenon composition that, relative to either atmospheric or average carbonaceous chondrite Xe (AVCC), has (1) clear excesses of Xe^{124} and Xe^{126} attributable to spallation, (2) Xe^{129} attributable to I^{129}, and (3) excess Xe^{134} and Xe^{136}.

The total sample results of *Marti et al.* [1969] appear to be intermediate between our light and dark samples. The less precise data of *Funkhouser et al.* [1967] show a much larger excess of Xe^{124} and Xe^{126}, which we presume is due to a difference in shielding or chemical composition. The observed ratios for all St. Severin samples plot on a Xe^{124}/Xe^{130} versus Xe^{129}/Xe^{130} diagram within the linear array for many other meteorites.

Figure 1 shows a Xe^{134}/Xe^{132} versus Xe^{136}/Xe^{132} correlation plot [*Hohenberg et al.*, 1967]

TABLE 1. Xe Analysis of St. Severin

	Sample Weight, mg	Xe^{132},* 10^{-12} cc STP/g	Xe^{132} Blank,† %	Isotopic Analysis									Xe_f^{136}, 10^{-12} cc STP/g
				Xe^{124}	Xe^{126}	Xe^{128}	Xe^{129}	Xe^{130}	Xe^{131}	Xe^{132}	Xe^{134}	Xe^{136}	
Mineral separates				1.33	1.79	6.37	65.4	9.87	63.1	100	63.4	63.2	180
Whitlockite	31	390	35	±0.07	±0.05	±0.11	±1.2	±0.14	±0.5		±0.6	±0.6	±30
Feldspar	124	54	88	0.75	1.11	7.92	121.2	15.5	79.4	100	39.7	34.2	1.1
				±0.07	±0.10	±0.20	±1	±0.2	±0.5		±0.3	±0.2	±0.8
Total samples													
Light	432	19	9	0.65	0.77	7.90	253.	14.9	78.2	100	42.9	38.2	1.5
				±0.05	±0.05	±0.15	±5.	±0.2	±0.6		±0.4	±0.3	±0.4
Dark	435	35	5	0.58	0.64	8.05	156.	15.9	81.2	100	39.6	34.1	1.0
				±0.03	±0.05	±0.14	±1	±0.1	±0.4		±0.2	±0.3	±0.5
Marti et al. [1968]		21		0.64	0.72	8.17	173.5	15.80	80.8	100	39.7	34.0	
				±0.02	±0.02	±0.10	±1.5	±0.12	±0.5		±0.3	±0.3	
		27		0.615	0.67	8.18	169.0	15.86	81.1	100	39.5	33.7	
				±0.015	±0.015	±0.15	±1.2	±0.10	±0.5		±0.3	±0.3	
Funkhouser et al. [1967]‡		41		1.8	2.5	8.3	147±6	17	80	100	40	34	
Atmosphere				0.357	0.335	7.14	98.3	15.17	78.8	100	38.8	33.0	
[*Nier*, 1950]				±0.004	±0.004	±0.02	±0.4	±0.05	±0.2		±0.1	±0.1	
Average carbonaceous chondrite (AVCC)				0.459	0.410	8.20		16.08	81.7	100	38.2	32.1	
[*Eugster et al.*, 1967]				±0.010	±0.004	±0.04		±0.11	±0.3		±0.2	±0.2	

* No blank corrections applied; concentration uncertain to ±15% for our analyses.
† Calculated from re-extractions and on Ni foil extractions on a per gram basis for each sample.
‡ Average of five analyses.

for the present samples along with data on other meteorites. These results define a precise linear array, which indicates that the Xe^{134} and Xe^{136} in these samples is almost certainly a mixture of two components. All St. Severin samples lie on this line. The whitlockite is seen to be greatly enriched in Xe^{134} and Xe^{136}. An excess is also evident in the light fraction of the total meteorite, whereas the dark fraction and the feldspar show only minor enrichments.

For a mixture of two components (α and β) the slope s of the mixing line is

$$s = \frac{(Xe^{134}/Xe^{132})\alpha - (Xe^{134}/Xe^{132})\beta}{(Xe^{136}/Xe^{132})\alpha - (Xe^{136}/Xe^{132})\beta}$$

For a given mixture m the ratio

$$(Xe^{134}/Xe^{132})_m/(Xe^{136}/Xe^{132})_m = (Xe^{134}/Xe^{136})_m$$

provides limits on the (Xe^{134}/Xe^{136}) of the end members. For the present case (Figure 1), the mixing line has a positive slope and a positive intercept on the (134/132) axis. The $(Xe^{134}/Xe^{136})_{max}$ value for the mixture most enriched in component α (ADR on Figure 1) is equal to the slope of the line drawn from the origin through this point. The point for the α end member must lie farther out on the mixing line. Thus, the slope of the line drawn from the origin to the α end member point (equal to $(134/136)_\alpha$) will be less than $(134/136)_{max}$. Similarly, the positive intercept of the correlation line implies that s is less than $(134/136)_\alpha$, as can be seen by inspection of Figure 1. Hence,

$$s < (Xe^{134}/Xe^{136})_\alpha < (Xe^{134}/Xe^{136})_{max}$$

From this relation we derive the strict limits $0.82 \leqq (Xe^{134}/Xe^{136})_\alpha \leqq 0.929 \pm 0.007$, where the upper limit is set by the 1300°C point from Angra dos Reis [*Hohenberg*, 1968]. The calculated value of $(Xe^{134}/Xe^{136})_F$ from the whitlockite is 0.93 ± 0.01 (see below). Because the extension of the line almost passes through the origin, any effects of variations in the yield of Xe^{132} from fission (and also spallation) would be small.

In addition to the precise linear array defined by different meteorites and air Xe (Figure 1), the beautifully detailed temperature study by *Hohenberg* [1968] on a single meteorite (Angra dos Reis) shows an identical line. We conclude

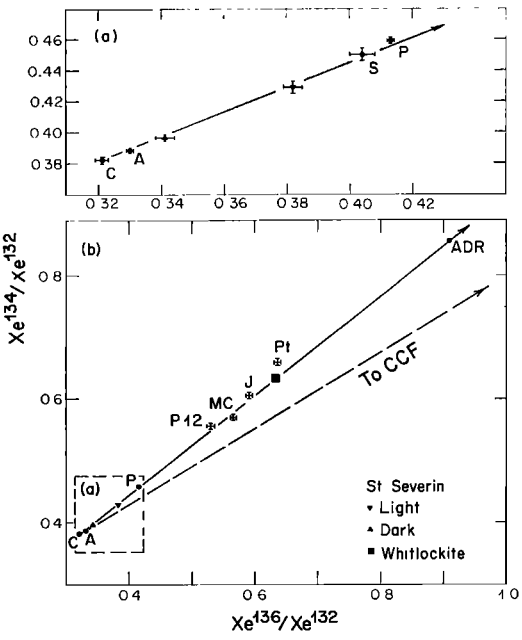

Fig. 1. Correlation diagram of Xe^{134}/Xe^{132} versus Xe^{136}/Xe^{132}. Figure 1*a* is an enlargement of the region indicated in 1*b*. The full line C to ADR passes through almost all the data shown to within experimental error and is evidence for a two-component mixture, the end point being meteoritic fission xenon. The feldspar and data of *Marti et al.* [1969] plot within the St. Severin dark point. The mixing line (dashed) of C and CCF is distinct. Literature data points are C, average carbonaceous chondrites [*Eugster et al.*, 1967]; A, atmosphere [*Nier*, 1950]; S, Stannern [*Marti et al.*, 1966] P and P12, Pasamonte total and 1200° fraction [*Hohenberg et al.*, 1967]; J, Juvinas, MC, Moore County, PT, Petersburg [*Kuroda et al.*, 1966]; ADR, Angra dos Reis [*Hohenberg*, 1968].

that a two-component model is adequately justified for Xe^{134} and Xe^{136}.

Decomposition of the spectrum. To calculate the amounts and spectrum of the whitlockite fission Xe, we assume that the following components are present: (1) Xe of atmospheric composition, (2) spallation Xe produced from cosmic-ray reactions with Ba and the rare earths, (3) radiogenic Xe^{129} from the decay of I^{129}, and (4) fission type Xe (i.e., excess heavy xenon isotopes assuming that Xe^{130} from fission is zero).

The proportions of these components for each isotope are shown in Figure 2, which schematically summarizes the results of the following

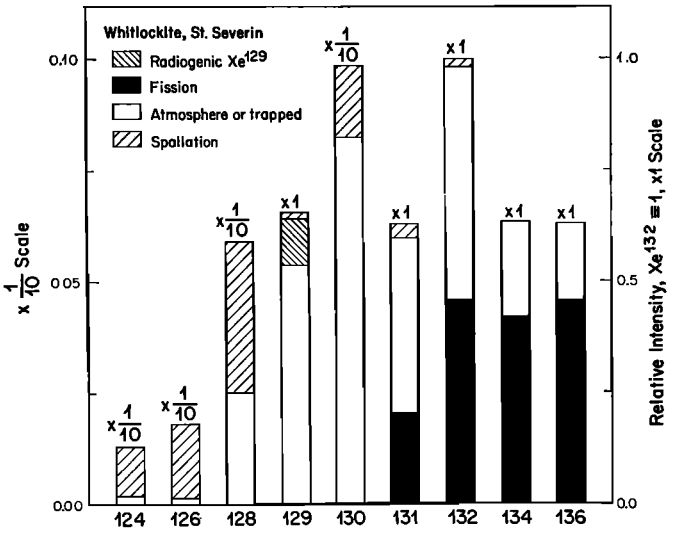

Fig. 2. Histogram of the isotopic ratios for xenon extracted from whitlockite separated from the St. Severin amphoterite. Note that the scales on the left and right are different. The fraction of each isotope due to the four components is shown. The shape of the fission spectrum is given by the dark columns at mass 131 through 136.

calculations based essentially on the procedure of *Marti et al.* [1966]. The choice of atmospheric Xe for the trapped component is justified by the fact that 54% of the observed Xe^{130} can be accounted for by our estimated blank and that only a negligible amount could be due to fission. It is quite possible, within the variability of our blanks, that the whitlockite itself contains no extraterrestrial trapped Xe. In any case, the calculated amounts and spectrum of fission type Xe would only be slightly affected if AVCC were chosen as the primordial component, and this effect is allowed for in the estimated errors. The calculation of the fission type spectrum requires the estimation of the relative spallation yields at masses 126, 130, 131, and 132. These three ratios are generally not well known, nor are they constants because of variations in the relative abundances of target elements and in the cosmic-ray energy spectrum from one sample to the next. Table 2 summarizes the relative Xe spallation yields obtained from the meteorite analyses that yield a well-defined spallation spectrum, and it shows our adopted values for these quantities. We assume that the spallation yields of Xe^{134} and Xe^{136} are negligible. Although the measured $(130/126)_{sp}$ ratios for the meteorites in Table 2 agree within the quoted errors, *Hohenberg* [1968] presents evidence for values as low as 0.55 from stepwise heating experiments on Angra dos Reis. Because of this and because of the relatively small number of data available, our error limits on $(130/126)_{sp}$ have been set to allow deviations of $\pm 50\%$. With these adopted spallation isotopic ratios, the decomposition of the spectrum was made as follows:

1. The amount of atmospheric Xe^{130} was calculated by assuming that the measured Xe^{130}/Xe^{126} ratio represented a mixture of spallation plus atmosphere.
2. The amounts of atmospheric gas at each mass number were calculated from the amount of atmospheric Xe^{130}.
3. The remaining gas at masses 124 and 128 was assigned to the spallation component, although it is possible that some of the excess Xe^{128} reflects neutron capture on I^{127}. The excess Xe^{129} represents the combination of spallation Xe^{129} and radiogenic Xe^{129} from the decay of I^{129}.
4. After correcting for spallation at masses 131 and 132, the remaining excesses at masses 131, 132, 134, and 136 constitute the fission type Xe spectrum.

The calculated Xe^{128} spallation yield for St. Severin whitlockite (Table 2) agrees within experimental error with previous measurements.

TABLE 2. Meteoritic Xe Spallation Spectra

Values in parentheses are assumed.

	Xe^{124}/Xe^{126}	Xe^{128}/Xe^{126}	Xe^{130}/Xe^{126}	Xe^{131}/Xe^{126}	Xe^{132}/Xe^{126}
Stannern*	0.590 ± 0.015	1.45 + 0.25 − 0.12	0.97 + 0.50 − 0.25	3.9 + 2.5 − 1.1	0.9 + 2.3 − 0.9
Lafayette†	0.58 ± 0.02	1.45 ± 0.2	0.88 ± 0.10	2.6 ± 0.3	0.3 ± 0.3
Weekeroo station silicate‡	0.526 ±0.013		1.12 ±0.04	3.25 ±0.16	0.90 ±0.17
Pasamonte (method A)§	0.59 ± 0.08 0.51 ± 0.08		1.01 ± 0.13 1.16 ± 0.13	4.5 ± 0.7 4.8 ± 0.7	(0) (1.0)
St. Severin whitlockite	0.71 ±0.05	1.54 ±0.13	(1.0 ± 0.5)	(4 ± 2)	(1 ±1)

* *Marti et al.* [1966].
† *Rowe et al.* [1966].
‡ *Bogard et al.* [1969*b*].
§ *Hohenberg et al.* [1967].

However, the Xe^{124} yield is slightly higher. If this difference is due to a somewhat harder flux spectrum for St. Severin, our adopted $(130/126)_p$ value may be somewhat high.

From the decomposition of the spectrum we obtain the results shown in Figure 2. The isotopic composition of the fission type xenon spectrum and the yield of Xe^{136} are given in Table 3 and shown in Figure 3.

Figure 3*a* compares the fission type xenon from the whitlockite with the values of the fission type xenon from Pasamonte. These values are the same to well within all errors.

An alternative fission type xenon has been obtained by *Eugster et al.* [1967], following a suggestion of *Krummenacher, Merrihue, Pepin, and Reynolds* [1962] that the differences between atmospheric Xe and AVCC were a result of fractionation plus the addition of a fission component. Several stepwise heating experiments on carbonaceous chondrites have also yielded fractions enriched in the heavy xenon isotopes [*Reynolds and Turner*, 1964; *Pepin*, 1968; *Rowe*, 1968]. These yield a fission type spectrum (extrapolated to $Xe^{130} = 0$) that is the same as obtained by *Eugster et al.* [1967] (Figure 3*b*). This type of 'carbonaceous chondrite type fission xenon' (CCF) is shown in Figures 3*b* and 1 and is compared with our results and the achondrite data. These are obviously different, and hence both spectra are not compatible with the products of a single fissionable nucleus.

Kr results. The total measured amounts and

TABLE 3. Calculated Fission Spectrum

	Xe^{131}/Xe^{136}	Xe^{132}/Xe^{136}	Xe^{134}/Xe^{136}	Xe_f^{136} atoms/g
St. Severin whitlockite	0.31 ± 0.08	0.97 ± 0.08	0.93 ± 0.01	$(5.3 \pm 0.8) \times 10^{9}$*

U = 1.0×10^{15} atoms/g [*Cantelaube et al.*, 1967]

Total track density 7.4×10^{6} tracks/cm² [*Cantelaube et al.*, 1967]

Measured $(Pu^{244}/U^{238}) = 0.035$*†

* Purity of whitlockite separate ~90%.
† For Pu^{244}: $\tau_{1/2}$ = 82 m.y., $\lambda_f/\lambda_{total} = 1.25 \times 10^{-3}$ [*Fields et al.*, 1966]. Assumed fission yield of Xe^{136} = 6%.

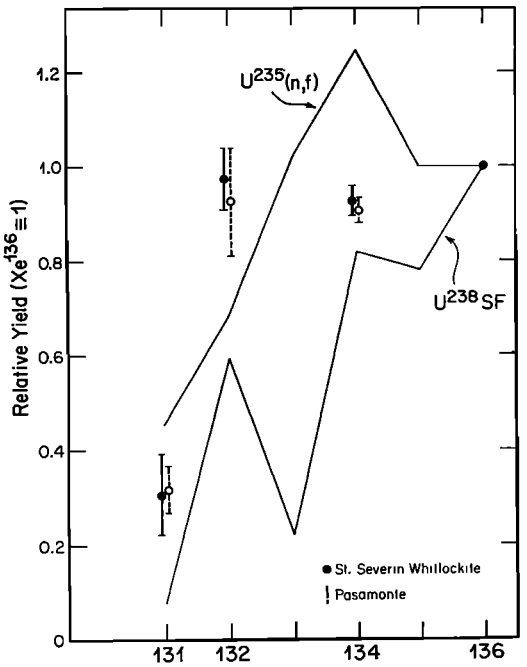

Fig. 3*a*. Neutron-rich excess xenon spectrum normalized to $Xe^{136} = 1$ for the St. Severin whitlockite. This spectrum is attributed to fission because of the correlation with fission tracks. The Pasamonte fission spectrum of *Eberhardt and Geiss* [1966] is shown for comparison. These spectra are identical to within experimental error. Note how these differ from the two uranium fission yield spectra [*Wetherill*, 1953; *Wanless and Thode*, 1955].

isotopic composition of Kr in the whitlockite were $Kr^{84} = (4.9 \pm 1) \times 10^{-11}$ cc STP, $Kr^{86}/Kr^{84} = 0.306 \pm 0.002$, $Kr^{83}/Kr^{84} = 0.211 \pm 0.002$, $Kr^{82}/Kr^{84} = 0.207 \pm 0.002$, and $Kr^{80}/Kr^{84} = 0.0447 \pm 0.0015$. Measurement of Kr^{78} was precluded by a large background contamination at this mass. The hydrocarbon correction applied was largest (1.1%) for Kr^{80} and significantly less for all others.

There is no large excess of Kr^{86}. A small amount of spallation Kr is mixed with Kr having an isotopic composition intermediate between air and AVCC. Within the uncertainties in the composition of the trapped component and the measured ratios, there may be no excess. Strong limits may be set for the $(Kr^{86}/Xe^{136})_{excess}$ ratio of

$$0 \leq (Kr^{86}/Xe^{136})_{excess} \leq 0.12$$

The upper limit is 30% lower than previously set by *Hohenberg et al.* [1967] and while it is only slightly less than the value of 0.13 for spontaneous fission of U^{238} [*Wetherill*, 1953], it is much lower than the value of 0.35 for thermal fission of U^{235} [*Arrol et al.*, 1949; *Wanless and Thode*, 1955]. This is in accord with the evidence given by the isotopic spectrum of the excess heavy Xe (see Figure 3*a*) and strengthens the conclusion that the excess Xe is not the result of U fission.

Track-xenon-U comparison. Track density measurements on seven whitlockite grains from the sample analyzed for Xe gave densities of 6–10 $\times$ 10^6 tracks/cm^2 for individual grains based on 50 to 700 tracks per grain. This is in agreement with the value of $7.4 \pm 1.4 \times 10^6$ reported by *Cantelaube et al.* [1967]. If a mean track length of 6 μ [*Cantelaube et al.*, 1967], a Xe^{136} fission yield of 6%, and 90% pure whitlockite are assumed, the expected amount of Xe^{136} would be lower by a factor of about 25

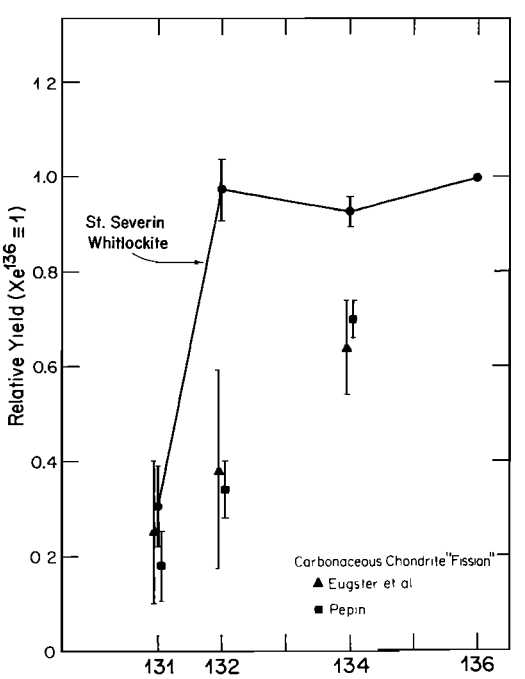

Fig. 3*b*. Comparison of the fission spectrum for St. Severin whitlockite (line with error bars) and the 'fission type' spectrum obtained by other workers on carbonaceous chondrites (CCF). These two spectra are distinct. There is no evidence that the CCF spectrum is related to fission.

from what is observed (Table 3). Thus, the correlation between fission tracks and fission Xe is qualitative but not quantitative. This difference can be explained by a later retention time for tracks than for fission Xe. The annealing curves given by *Cantelaube et al.* [1967] indicate that fission tracks would be annealed at low temperature, whereas it is plausible to expect the xenon to be retained. The difference between Xe and track retention might result from relatively slow initial cooling or by a reheating event at a time before the complete decay of Pu^{244}. The possibility that the present low density is due to slow annealing at low temperature over 4.6×10^9 years is less plausible.

It is possible to calculate a 'track Xe' interval which could be used to define a thermal history for specific models, if it is assumed that temperatures could be assigned to track retention and Xe retention. For St. Severin this time interval corresponds to about 380 m.y. The study of initial $(Sr^{87}/Sr^{86})_I$ in this meteorite on the same whitlockite gives a time of 'metamorphism' relative to the initial $(Sr^{87}/Sr^{86})_{BABI}$ of basaltic achondrites of $\Delta T_{BAGB} \cong 320 \pm 160$ m.y. (see *Papanastassiou and Wasserburg* [1969] for notation). This time of Sr re-equilibration may be the same event as reflected in the 'track Xe' interval.

The measured fission Xe and U in the whitlockite and our estimated whitlockite abundance (4×10^{-4} g/g) can account for only about 10% of the fission Xe and 2% of the U observed in the total meteorite samples, if a total meteorite U content of 10 ppb is assumed. This assumption is in part justified by the He^4 content which we have measured to be 1.5×10^{-5} cc STP/g in both the dark and the light phases ($He^4/He^3 = 71$) as well as the results by *Marti et al.* [1969] (1.9×10^{-5} cc STP/g). These results are typical for a chondrite that has not suffered serious diffusion loss. Our data (Table 1) show that the feldspar has not concentrated fission Xe and probably not U. Thus, the mineral phases in which the bulk of these elements occur are unknown. The phosphorus contributed by the whitlockite is only 7% of the total P in the meteorite [*Müller*, 1968] and is similar to the proportion of fission Xe^{136} in the total meteorite attributable to whitlockite. Although other phosphorus-bearing minerals may be present, it is possible that the whitlockite recovery is grossly incomplete.

Summary of Results

We now focus on the important results from this work. The conclusions stated here are insensitive to the assumptions made in the data analysis. The critical observations are as follows:

1. In addition to both a high density of excess fission tracks and a high U concentration, the whitlockite has a high concentration of excess neutron-rich xenon isotopes (see Figure 2). The number of excess Xe^{136} atoms is about twenty-five times greater than expected from the fission track density.

2. The Xe^{136} excess observed in other minerals with a low track density and in the total meteorite is a factor of ~200 less in concentration than in the whitlockite.

3. A significant excess of Xe^{129} is observed in the whitlockite (see Figure 2) as well as in the other samples.

4. The calculated isotopic spectrum of the neutron-rich xenon isotopes (Xe^{131} to Xe^{136}) is defined with high precision (see Figure 3*a*) and is identical with the previous best estimates of the so-called fission xenon component of the Pasamonte achondrite [*Rowe and Kuroda*, 1965; *Rowe and Bogard*, 1966; *Eberhardt and Geiss*, 1966; *Hohenberg et al.*, 1967]. The fission Xe^{134}/Xe^{136} ratio is compatible with a type of excess heavy Xe found in other achondritic meteorites. This Xe^{134}/Xe^{136} composition appears to be ubiquitous and uniquely defined. This is the first clearly established occurrence of this type of Xe in chondrites.

Conclusions

The results show that there is a strong qualitative correlation between the excess heavy Xe isotopes and excess fission tracks in meteorites. The presence of excess heavy Xe in the same grains as excess tracks establishes that these effects arise from the *same* process and that this process is nuclear fission.

From the apparent uniqueness of the isotopic spectrum we conclude that the fission products (Xe and tracks) are not a mixture of several components. Despite the lack of a quantitative correlation between the tracks and excess Xe, the high concentrations of both fission Xe and fission tracks in the whitlockite as compared

with that in the other minerals (e.g., feldspar) strongly argue that the Xe excess is most reasonably understood as the result of in situ decay of a single fissionable isotope and not as an inherited or trapped fission component.

Neither the tracks nor the Xe excess can be accounted for by induced fission. Furthermore, the concentration of the fission products in a phase rich in U and, presumably, the rare earths is to be expected from the chemical properties of Pu. These provide the most direct arguments that we are observing the effects of the spontaneous fission of Pu^{244} contained in these meteorites at the time of their formation. If so, then the fission spectrum as reported here is the one to be tentatively associated with Pu^{244}. Other examples of fission type xenon [*Munk*, 1967; *Reynolds and Turner*, 1964; *Pepin*, 1968; *Funk et al.*, 1967; *Eugster et al.*, 1967; *Rowe*, 1968], such as shown in Figure 3*b* for carbonaceous chondrites, cannot, from our considerations, be attributed to Pu^{244} spontaneous fission and must be due to some other process. This was previously speculated by *Pepin* [1968] and *Rowe* [1968] although on a much more tentative basis than is possible in the present work. Whether the CCF Xe spectrum is, in fact, due to fission remains to be resolved. A suggestion by *Munk* [1967] of another fission type spectrum in Angra dos Reis is subject to considerable doubt for reasons given by *Hohenberg* [1968].

The amount of excess Xe^{129} is much too large to be accounted for by spallation only, and some must result from the decay of I^{129}. The presence of radiogenic Xe^{129} is consistent with the identification of the excess neutron-rich Xe as a product of Pu^{244} fission. The possibility that some of the I^{129} was formed as a fission product cannot be excluded in the present case. If a $(129/126)_{sp}$ ratio of 0.9 ± 0.5 [*Rowe*, 1967] is assumed, a concentration of excess Xe^{129} of $41 \pm 22 \times 10^{-12}$ cc STP/g is obtained for the whitlockite.

Assuming that the U concentration of *Cantelaube et al.* [1967] (Table 3) is representative of our samples, we calculate a Pu^{244}/U^{238} ratio of 1/30 at the time when the whitlockite first began to retain fission Xe. If Pu was not enriched with respect to U in the formation of the whitlockite, this value is a lower limit to the Pu^{244}/U^{238} ratio at the time of formation of the solar system.

Previous reports of high Pu^{244}/U^{238} ratios were based on measurements of excess heavy Xe isotopes having the 'carbonaceous chondrite fission' spectrum [*Pepin*, 1968; *Funk et al.*, 1967]; however, this spectrum should not be associated with Pu^{244}. A report of a high Pu/U ratio based on track measurements [*Price et al.*, 1968] has since been retracted [*Schirk et al.*, 1969]. In general, Pu^{244}/U^{238} ratios calculated from Pasamonte-type fission Xe measurements [*Hohenberg et al.*, 1967; *Sabu and Kuroda*, 1967] and other track studies [*Fleischer et al.*, 1968; *Schirk et al.*, 1969] are much lower than values obtained in this work.

If the separation time of the solar system from the interstellar medium is assumed to range from 0 to 1.3×10^8 years, the range in Pu^{244}/U^{238} is 1/30 to 1/10 at the termination of nucleosynthesis. This range of values can also be obtained from the calculations of *Fowler* [1962] if continuous uniform nucleosynthesis is assumed for durations ranging from 3.3 to about 1×10^9 years. However, these duration times are incompatible with the values (greater than 5.5×10^9 years) necessary to account for the solar system U^{235}/U^{238} ratio on the same model.

An upper limit of 360 m.y. for the separation time can be set by assuming all the Pu^{244} was synthesized in a sudden event. This upper limit is insensitive to errors in the relative production rates. However, a sudden synthesis model [*Fowler*, 1962], even allowing for a wide range in relative production rates, fails to give consistent solutions for Pu^{244}/U^{238} and U^{235}/U^{238}. Unless the fissioning nucleus is not Pu^{244} or unless there is considerable chemical fractionation, it appears that the theoretical relative production rates are considerably off or that a more complex model for galactic nucleosynthesis is necessary.

Note added in proof. We have recently measured the He^4 content of the whitlockite to be 8×10^{-4} cc STP/g whitlockite with $He^4/He^3 = 2.6 \times 10^3$. Assuming no He loss, these data are consistent with the U concentrations measured by *Cantelaube et al.* [1967] and a Th/U ratio of ~4.

As only ⅓ of the He is produced by Th decay, this is not very sensitive to the Th/U ratio.

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