## **References and Notes**

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## Time Differences in the Formation of Meteorites as Determined from the Ratio of Lead-207 to Lead-206

Abstract. Measurements of the lead isotopic composition and the uranium, thorium, and lead concentrations in meteorites were made in order to obtain more precise radiometric ages of these members of the solar system. The newly determined value of the lead isotopic composition of Canyon Diablo troilite is as follows:  $\frac{206Pb}{204Pb} = 9.307$ ,  $\frac{207Pb}{204Pb} = 10.294$ , and  $\frac{208Pb}{204Pb} = 10.294$ 29.476. The leads of Angra dos Reis, Sioux County, and Nuevo Laredo achondrites are very radiogenic, the <sup>206</sup>Pb/<sup>204</sup>Pb values are about 200, and the uraniumthorium-lead systems are nearly concordant. The ages of the meteorites as calculated from a single-stage <sup>207</sup>Pb/<sup>206</sup>Pb isochron based on the newly determined primordial lead value and the newly reported <sup>235</sup>U and <sup>238</sup>U decay constants, are  $4.528 \times 10^9$  years for Sioux County and Nuevo Laredo and  $4.555 \times 10^9$  years for Angra dos Reis. When calculated with the uranium decay constants used by Patterson, these ages are  $4.593 \times 10^9$  years and  $4.620 \times 10^9$  years, respectively, and are therefore 40 to  $70 \times 10^6$  years older than the  $4.55 \times 10^9$  years age Patterson reported. The age difference of  $27 \times 10^6$  years between Angra dos Reis and the other two meteorites is compatible with the difference between the initial 87 Sr/86 Sr ratio of Angra dos Reis and that of seven basaltic achondrites observed by Papanastassiou and Wasserburg. The time difference is also comparable to that determined by <sup>129</sup>I-<sup>129</sup>Xe chronology. The ages of ordinary chondrites (H5 and L6) range from 4.52 to  $4.57 \times 10^9$  years, and, here too, time differences in the formation of the parent bodies or later metamorphic events are indicated. Carbonaceous chondrites (C2 and C3) appear to contain younger lead components.

The absolute ages of meteorites have been determined by 207Pb/206Pb, 87Rb-<sup>87</sup>Sr, and <sup>40</sup>K-<sup>40</sup>Ar methods to be about  $4.5 \times 10^9$  years. Of these methods, the 207Pb/206Pb method has an advantage in that it requires only that the isotopic ratio be measured in order that the age of the meteorites be determined, on the assumption either that the initial Pb isotopic composition is known or that some meteorites are cogenetic.

Significant progress has recently been made by Wasserburg and his coworkers (1, 2) in the Rb-Sr chronology of meteorites. Their precise measurements of Sr isotopic ratios and superior mineral separation techniques have effectively confirmed the time difference in meteorite formation which was first found by Reynolds (3) from gas retention ages based on 129Xe derived from extinct <sup>129</sup>I. Wasserburg's group obtained many excellent meteorite ages by means of internal isochrons. However, the Rb/Sr ratios are relatively low in most stone meteorites,

especially achondrites; thus only a small change in Sr isotopic composition occurs during the lifetime of meteorites. This has sometimes made Rb-Sr chronology difficult. Furthermore, the decay constant of <sup>87</sup>Rb is largely dependent upon the accuracy with which the decay constant of U is known.

Recent precise redeterminations of the decay constants of <sup>238</sup>U and <sup>235</sup>U (4) and <sup>232</sup>Th (5) and of the abundance ratio of  $^{238}$ U to  $^{235}$ U (6) (the values are listed in footnote § of Table 2) and recent improved analytical techniques have encouraged us to carry out the radiometric study presented here of meteorites by the U-Th-Pb system. In the following discussion the reported ages, which were based on old U decay constants, are recalculated on the basis of the new decay constants, and the recalculated values are listed in parentheses for comparison.

Patterson (7) analyzed several meteorites for their Pb isotopic composition and obtained for the first time a pre-

cise age for the meteorites of  $4.55 \pm$  $0.07 \times 10^9$  years (4.49 × 10<sup>9</sup> years). Because Pb in Pacific sediment fits the Pb array in the meteorites, he concluded that the earth formed contemporaneously with meteorites  $4.55 \times$ 109 years ago. Similar data on meteorites obtained later by many scientists (8-10) fell for the most part on an isochron like that which Patterson reported. However, Hamaguchi et al. (11) and Bate et al. (12) analyzed U and Th concentrations in stony meteorites by neutron activation and reported that the U and Th contents are far lower than those estimated by Patterson to produce the observed Pb isotopic composition and Pb concentration. One might therefore suspect the <sup>207</sup>Pb/<sup>206</sup>Pb age of meteorites from these results (13). The <sup>207</sup>Pb/<sup>206</sup>Pb ages of the meteorites obtained in our study range from 4.56 to  $4.64 \times 10^9$ years (4.50 to  $4.57 \times 10^9$  years by newly reported U decay constants) which are 10 to  $90 \times 10^6$  years older than the age that Patterson reported. In this report, instead of discussing the mean age of the formation, we will discuss the time difference in the formation of meteorites which was obtained from the <sup>207</sup>Pb/<sup>206</sup>Pb ratio,

In the work reported here extensive precautions were taken to avoid possible contamination of samples before and during the analyses. Small (10 to  $\sim 50$  g) specimens of meteorites, except Allende (14), which were free of fusion crust, were washed for 1 minute in cold 2.5N HCl by means of an ultrasonic vibrator and were rinsed repeatedly with water prior to pulverization in a boron carbide mortar. The analytical procedures for silicates reported previously (15) consist of decomposition by HClO<sub>4</sub>-HF in a Teflon bomb and Pb separation by coprecipitation with  $Ba(NO_3)_2$  and anodic electrodeposition. Troilites were dissolved in a HCl-HNO<sub>3</sub> mixture in a Teflon bomb, and Pb was separated by an anion-exchange (Br- form resin) technique (16) and anodic electrodeposition; U and Th were separated by an anion-exchange ( $NO_3^-$  form resin) method (15). The Pb isotopic ratios were measured by the phosphate-silica gel method (17) in a mass spectrometer equipped with a digital voltmeter and an on-line computer. A correction for the fractionation of +0.1 percent per mass unit was applied to the raw ratios. The correction factor was obtained from replicate measurements on standard SRM-982 of the National



Fig. 1. The <sup>200</sup>Pb/<sup>201</sup>Pb versus <sup>207</sup>Pb/<sup>201</sup>Pb evolution diagram for meteorites;  $\bigcirc$ , troilite; +, chondrite;  $\blacktriangle$ , achondrite;  $\bigtriangleup$ 's are the slopes for the <sup>207</sup>Pb/<sup>200</sup>Pb isochron between the Canyon Diablo troilite and the individual meteorites. The nonradiogenic part is expanded in the insert. A line which intersects isochrons at the Canyon Diablo troilite illustrates a possible change in the initial Pb isotopic composition with increasing  $\mu$  from 4.56 to 4.55  $\times$  10° years before the present. The solid dot indicates Oversby's value (16) for Canyon Diablo troilite.

Bureau of Standards. The mass fractionation data obtained during the last 3 years ranged from -0.07 to -0.15percent for ionizing filaments operating at a temperature of  $1280^{\circ} \pm 30^{\circ}$ C. The variation may be due to small differences in the operating temperature, and in the quantity and quality of silica gel used. The Pb blanks were 3 to 5 ng per 1-g sample, and the U and Th blanks were 0.02 and 0.06 ng per 1-g sample, respectively.

The concentrations of U, Th, and Pb determined by isotope dilution in the study presented here are given in Table 1, along with previously determined values from neutron activation analysis (11-13, 18-21). The U concentrations in the chondrites range from 10 to 19 parts per  $10^9$  (ppb) and are about 10 percent lower than the previously reported values determined by activation analysis. Beardsley and Modoc are exceptions and exhibit values

20 percent higher than those previously reported. For achondrites, the U concentrations in Angra dos Reis and Nuevo Laredo agree with earlier results, whereas the concentration in Sioux County is 20 percent higher than earlier values (11, 13). Meteorites are demonstrably heterogeneous, and these small differences between the values obtained by isotope dilution and those obtained by neutron activation analysis probably are due to sample heterogeneity.

The Th concentrations obtained in our study are in good agreement with the activation analysis data of Bate *et al.* (12) and Morgan and Lovering (20, 21). The Th/U ratios of chondrites and achondrites range from 3.10 (Modoc) to 4.29 (Angra dos Reis), and average 3.7; these values are in good agreement with Murthy and Patterson's (8) ratio estimated from the Pb isotopic composition.

The Pb concentrations in carbonaceous chondrites are about 1.5 parts per  $10^6$  and are 3 to 8 times higher than the Pb concentrations in achondrites, whereas achondrites have 10 times higher U concentrations than carbonaceous chondrites. The Pb concentration in ordinary chondrites ranges from 75 to 250 ppb.

The Pb isotopic compositions of the meteorites are shown in Table 2, and a plot of the  ${}^{206}\text{Pb}/{}^{204}\text{Pb}$  ratio versus the  ${}^{207}\text{Pb}/{}^{204}\text{Pb}$  ratio is given in Fig. 1. The  ${}^{207}\text{Pb}/{}^{206}\text{Pb}$  age of meteorites of  $4.55 \pm 0.07 \times 10^9$  years (4.49 × 10<sup>9</sup> years) obtained by Patterson (7) is large-

Table 1. Uranium, thorium, and lead concentrations in meteorites (ppb); I.D., isotope dilution (this study); A.A., activation analysis.

Sample	Class	U		Th			Pb			Th/U	
		I.D.	A.A.	I.D.	A	A.	I.D.	A	.A.	I.D.	A.A.
				Troilite, iron	meteorites			arian dika sama na diliking dika dipiking di akang			dia in the state on the destination of the state of the
Canyon Diablo	Og	< 0.1	0.4 (13)	<0.1			6860	5100-7840	(13)		
Odessa	Og	1.0		<0.1			4215				
				<b>Car</b> bonaceous	chondrites						
Murray	C2	{ 10.7 { 10.9	15-28 (19) 11.3 (21)	39.3 39.8	45.4	(21)	1516 1500			3. <b>67</b> 3. <b>65</b>	4.0
Allende	C3	15.3	16.2 (35)	62.2			1097			4.07	
				Bronzite c.	hondrites						
Beardsley	H5	14.6	10-17 (13) 10.3 (21)	49.6	47.7 36.3	(12) (21)	259	170	(13)	3.40	4.8–2.8 3.5
Plainview*	H5	10.9	12.5 (19)	37.6		``·	195			3.45	
Richardton	<b>H</b> 5	9.97	$   \left\{ \begin{array}{ccc}     11.3 & (11) \\     10.7 & (21)   \end{array} \right. $	37.3	38.0 29.6	(12) (21)	74.7			3.75	3.36 2.8
				Hypersthene	chondrites						
Modoc	L6	19.0	(10.8 (11) 14 (13)	58.5	39.2	(12)	79.0	60	(13)	3.10	3.63 2.80
				Achon	drites						
Sioux County	Eucrite	78.0	63.0 (18)	302.4			193	100		3.87	
Nuevo Laredo	Eucrite	131.6 123.6	$126 (11) \\ 132, 139 (20)$	483.7 475.9	476 459, 492	(12) (20)	328 320	420 630	(13) (13)	3.67	3.78 3.5
Angra dos Reis	Angrite	202.8 210.7	<b>198 (2</b> 0)	869.6 901.0	967	(20)	541 550			4.29 4.28	4.88

\* This sample is a find; all other samples in this table are falls.

Table 2. Lead isotopic composition and  ${}^{367}\text{Pb}/{}^{269}\text{Pb}$  ages of meteorites. The analytical blank was corrected. The errors are one standard deviation (one  $\sigma$ ).

Sample	Class	<sup>208</sup> Pb/ <sup>204</sup> Pb	<sup>207</sup> Pb/ <sup>204</sup> Pb	<sup>208</sup> Pb/ <sup>204</sup> Pb	Blank* (%)	μŤ	( <sup>207</sup> Pb/ <sup>206</sup> Pb) <sub>R</sub>	Age§ $(\times 10^9 \text{ years})$
			Troilites, iro	n meteorites				
Canyon Diablo	Og	$9.307 \pm 0.003$	$10.294 \pm 0.003$	<b>29.476 ±</b> 0.009	<0.1			
Odessa	Og	9.995 ± .004	$10.691 \pm .005$	$30.087 \pm .016$	0.1			
			Carbonaceoi	is chondrites				
Murray	C2	$9.806 \pm .006$	$10.594 \pm .007$	$29.939 \pm .020$	0.5	0.32	0.6012	$4.511 \pm 0.042$
Allende	C3	$11.250 \pm .003$	$11.451 \pm .004$	$31.352 \pm .011$	0.5	0.51	0.5953	4.49 <b>6</b> ± .010
			Bronzite	chondrites				
Beardsley	H5	$12.193 \pm .033$	$12.106 \pm .033$	$31.973 \pm .086$	1.4	2.8	0.6271	$4.574 \pm .012$
Plainview	H5	$13.682 \pm .015$	$12.958 \pm .015$	$33.447 \pm .037$	0.9	2.9	0.6089	4.529 ± .010
Richardton	H5	$32.931 \pm .325$	$24.575 \pm .244$	52.017 ± .521	4.5	12.7	0.6044	$4.519 \pm .015$
			Hypersthen	e chondrite				
Modoc	L6	41.308 ± .411	$29.788 \pm .298$	<b>57.27</b> 3 ± .575	2.5	29.6	0.6092	4.530 ± .015
			Achor	ndrites				
Sioux County	Eucrite	$195.34 \pm 5.41$	$123.29 \pm 3.45$	$201.78 \pm 5.31$	1.8	180	0.6074	$4.526 \pm .010$
Nuevo Laredo	Eucrite	$222.38 \pm 1.07$	$140.05 \pm .69$	$233.64 \pm 1.06$	0.8	203	0.6089	$4.529 \pm .005$
Angra dos Reis	Angrite	$214.62 \pm 1.34$	$137.53 \pm .86$	$254.64 \pm 1.60$	1.4	19 <b>7</b>	0.6197	4.555 ± .005
* Corrected Ph blan	k divided b	w the total Ph in the	sample + Measur	ed 238II/204Pb (today's	) ratios	+ Padion	nic component	ofter substraction

\* Corrected Pb blank divided by the total Pb in the sample. † Measured  $^{238}U/^{204}Pb$  (today's) ratios. ‡ Radiogenic component after substraction of the primordial Pb.  $$\lambda of ^{238}U = 0.15525 \times 10^{-9} \text{ years}^{-1}, \lambda of ^{235}U = 0.98485 \times 10^{-9} \text{ years}^{-1}, \lambda of ^{232}Th = 0.049475 \times 10^{-9} \text{ year}^{-1}, ^{238}U/^{235}U = 137.88.$ Errors are the sum of each  $2\sigma$  of the mean in the  $^{297}Pb/^{290}Pb$  measurement between the individual sample and Canyon Diablo.

ly constrained by nonradiogenic Pb in the troilite phase of Canyon Diablo and by the very radiogenic Pb in the Nuevo Laredo achondrite. Many later investigations conducted on troilite (8, 16, 22, 23) have confirmed that the Pb in Canyon Diablo is the least radiogenic yet discovered. Because the troilite contains a substantial amount of Pb but not enough U and Th to measurably change the Pb isotopic composition within  $\sim 4.5 \times 10^9$  years, the Pb of Canyon Diablo troilite has been considered to be the "primordial" Pb existing when the parent bodies of meteorites and the earth accreted. All the primary ages of solar bodies obtained by the <sup>207</sup>Pb/<sup>206</sup>Pb method were calculated from this primordial Pb as the initial point. Oversby (16) measured the Pb isotopic composition of the primordial Pb by a double-spike technique which she firmly claimed had an accuracy of 0.1 percent in the <sup>206</sup>Pb/<sup>204</sup>Pb ratio measurement and 0.15 percent in the <sup>207</sup>Pb/<sup>204</sup>Pb ratio measurement. However, our measurements made with the phosphate-silica gel method revealed that Oversby's values for the primordial Pb isotopic composition, which have been widely accepted in Pb isotope studies, are in error far beyond the stated uncertainties. Her double-spike technique apparently introduced too large a mass-fractionation correction, which might be due to computation error or related to blank correction (24), so that the reported ratios of larger masses became smaller and were off the Pb array in the meteorites (Fig. 1). Interestingly, Ostic's values (23) for the Pb isotopic

composition in the troilite of the Toluca iron meteorite, which presumably were measured by a PbS method in a solid-source mass spectrometer, are in good agreement with the Canyon Diablo troilite data presented here. (Ostic, however, did not report details of the mass fractionation correction.)

The precision of our measurements for the isotopic composition of Canyon Diablo troilite (Table 2) is  $\pm 0.03$  percent ( $\sigma$ ) for <sup>206</sup>Pb/<sup>204</sup>Pb, <sup>207</sup>Pb/<sup>204</sup>Pb, and <sup>208</sup>Pb/<sup>204</sup>Pb ratios. Our measurements of Canyon Diablo troilite showed the presence of slightly more U than was found in the measurement blank; however, the U does not affect the Pb isotopic ratio beyond the instrumental measurement error stated above. The Pb blank, which influences primarily the accuracy of the isotopic ratio, was less than 0.05 percent of the total sample Pb. Our best values for the "primordial Pb isotopic composition" are as follows:  ${}^{206}Pb/{}^{204}Pb =$  $9.307 \pm 0.006$ ,  ${}^{207}\text{Pb}/{}^{204}\text{Pb} = 10.294 \pm$ 0.006, and  $208Pb/204Pb = 29.476 \pm$ 0.018.

The Pb isotopic composition of the Odessa troilite is more radiogenic than that of the Murray carbonaceous chondrite (Fig. 1), even though the U concentration is nearly as low as that for the Canyon Diablo troilite. We presume that the radiogenic Pb of the Odessa troilite could have been introduced by terrestrial contamination, as Oversby concluded (16).

The  ${}^{206}Pb/{}^{204}Pb$  ratio of Nuevo Laredo was previously reported to be about 50 to 60 (7, 25); however, we suspected, on the basis of a comparison of the U/Pb ratios (13, 26) and the abundance of volatile elements (27) in lunar rocks and achondrites, that Pb in the achondrite might be more radiogenic. The observed Pb isotopic composition for three achondrites (Table 2)-Nuevo Laredo (eucrite), Sioux County (eucrite), and Angra dos Reis (angrite)—is far more radiogenic than the values previously reported, and the 206Pb/204Pb ratio is about 200, which very much resembles that of lunar rocks. If we assume a single-stage Pb development from the troilite Pb of Canyon Diablo as primordial Pb, the <sup>207</sup>Pb/<sup>206</sup>Pb ages of Nuevo Laredo and Sioux County are 4.529 and 4.526  $\times$  $10^9$  years, respectively, using the new U decay constants. The age obtained for Nuevo Laredo is about  $0.05 \times 10^9$ years older than the value previously reported by Patterson or the recomputed values. However, it is close to the value of Silver and Duke (25), even though the radiogenic character is much different.

The <sup>207</sup>Pb/<sup>206</sup>Pb age of Angra dos Reis,  $4.555 \times 10^9$  years, is distinguishably greater than the ages of the other two achondrites reported above. These results are consistent with those of Papanastassiou and Wasserburg (1, 28) that the initial <sup>87</sup>Sr/<sup>86</sup>Sr ratio of Angra dos Reis, (87Sr/86Sr) ADOR, is distinctly smaller than that of seven basaltic achrondrites,  $({}^{87}Sr/{}^{86}Sr)_{\rm BABI}$ . On the assumption that Angra dos Reis formed from a chondritic-type reservoir, Papanastassiou (28) concluded that the achondrite formed  $14 \times 10^6$  years before the other basaltic achondrites, whereas the data presented here indicate that the time difference is about  $27 \times 10^6$  years. This age difference seems also to be in accord with the results obtained by Hohenberg (29) that Angra dos Reis contains a large <sup>244</sup>Pu-fission Xe component.

In the initial <sup>87</sup>Sr/<sup>86</sup>Sr method used by Papanastassiou in the interpretation of the age difference one of the requirements was the assumption of an elemental abundance ratio of Rb/Sr in the parent bodies of achondrites. In contrast, the principal requirement of the <sup>207</sup>Pb/<sup>206</sup>Pb method is that all meteorites must have contained the same primordial Pb, if the ages of the individual meteorites are compared; however, the ages of these achondrites are little affected by a small difference in the primordial Pb because Pb in achondrites is extremely radiogenic. For the age calculation, we assume a single-stage development in <sup>207</sup>Pb/<sup>206</sup>Pb ratios from the primordial Pb after formation (or metamorphic events) of the parent bodies of meteorites, namely, that the U-Th-Pb system was a closed system in these parent bodies. In the absolute sense, however, the U-Th-Pb system is probably not a closed system, and the U/Pb and Th/Pb ratios in the parent bodies may have changed during early metamorphic events. A later event, collision which broke up the parent bodies to meteorities, may little affect the 207Pb/206Pb ages if we consider the shorter cosmic-ray exposure ages of stone meteorites to be of the order of  $10^7$  years (30). Terrestrial contamination, if it occurred as the latest event, did not appreciably change the <sup>207</sup>Pb/<sup>206</sup>Pb ratio, even though it affected the radiogenic character of the <sup>206</sup>Pb/<sup>204</sup>Pb, <sup>207</sup>Pb/<sup>204</sup>Pb, and <sup>208</sup>Pb/ <sup>204</sup>Pb ratios, because common terrestrial Pb has a 207Pb/206Pb ratio similar to those in meteorites. The U-Th-Pb systematics of the achondrites studied in the present investigation are almost concordant within the error of blank correction, and the assumption of the closed system may be held for age calculation.

Class H5 bronzite chondrites show a significant variation in the Pb isotopic composition. Leads of Beardsley and Plainview are less radiogenic and that of Richardton is more radiogenic than common terrestrial Pb. The data also indicate different ages of formation or metamorphism. The U-Th-Pb system of Beardsley is almost concordant, and the  ${}^{207}\text{Pb}/{}^{206}\text{Pb}$  age is  $4.574 \times 10^9$  years, on the basis of the new U decay constants. The Beardsley

age is the highest obtained in the investigation. Plainview (which is a "find"the other meteorites used in this study are "falls") and Richardton have  $^{207}\mathrm{Pb}/^{206}\mathrm{Pb}$  ages of 4.529 and 4.519  $\times$ 109 years, respectively. However, we were not able to show that the U-Th-Pb systematics of the meteorites are concordant. The Pb concentrations of Plainview and Richardton are 20 and 40 percent, respectively, in excess of that produced from U and Th decay in the meteorites. On the other hand, our U value in these two meteorites is about 15 percent lower than that determined by activation analysis. Because the U, Th, and Pb concentrations are extremely low, experimental error might have introduced the discordancy and we discuss them with some reservation until additional experiments have been carried out. However, as noted above, the terrestrial contamination affects the <sup>207</sup>Pb/<sup>206</sup>Pb ages only slightly, and there appears to be an age difference between Beardsley and the two meteorites, Plainview and Richardton, if these had the same initial Pb isotopic composition. Alternatively, it could be interpreted that these bronzites are the same age, but that a later metamorphic event increased  $\mu$  (<sup>238</sup>U/<sup>204</sup>Pb; today's ratio = 12.7) in Richardton as compared to Beardsley (the observed  $\mu = 2.8$ ) to produce more radiogenic Pb, and so the age of Richardton referred to the initial Pb appears younger. Interestingly, Beardsley contains exceptionally high concentrations of Rb and K as compared to other chondrites in the H group (31), and Kaushal and Wetherill (32) observed a slightly apparent older age for Beardsley than for others in the group in their Rb-Sr chronological study, even though they considered that the apparent age difference was within the range of experimental error and therefore was not significant.

Lead of a hypersthene chondrite (L6), Modoc, is more radiogenic than that of the bronzite chondrites but less radiogenic than the achondrites. The age of the L6 chondrites is similar to that of Nuevo Laredo and Sioux County.

Some investigators (33) have suggested (but subsequently rejected the idea) that carbonaceous chondrites would be a close approximation of primordial matter, because of their high content of volatile elements, and that ordinary chondrites were derived from carbonaceous chondrites. Podosek and Lewis (34) concluded from

tremely high <sup>244</sup>Pu/<sup>238</sup>U ratio that the white inclusions in Allende formed relatively earlier than most meteorites; however, the <sup>207</sup>Pb/<sup>206</sup>Pb ages of C2 and C3 carbonaceous chondrites, Murray  $(4.511 \times 10^9 \text{ years})$  and Allende  $(4.496 \times 10^9 \text{ years})$ , appear to be younger than that of other meteorites studied in this report. The Pb isotopic composition of Murray obtained in this study is less radiogenic and the <sup>207</sup>Pb/<sup>206</sup>Pb age is younger than Marshall's values (10). The Pb in Murray is similar to that in Canyon Diablo, and the increments of 206Pb and 207Pb are small enough to introduce easily an error in the 207Pb/206Pb ratio. However, the 207Pb/206Pb age of Allende is also young, and we do not believe that the younger age for carbonaceous chondrites was introduced by our measurement error. It can be argued that the initial Pb for the carbonaceous chondrites might be slightly different from the primordial Pb which was assumed to be a common initial Pb for all of the meteorites-moon-earth system. If the accretion of volatile-rich carbonaceous chondrites caused the increase in the U/Pb ratio in the nebula and other meteorites formed later from the nebula with the increased U/Pb ratio, the initial leads, then, are different between carbonaceous chondrites and others. The primary <sup>207</sup>Pb/<sup>206</sup>Pb ages of carbonaceous chondrites appear to be younger than the true ages, if the initial Pb for carbonaceous chondrites is more primitive than the primordial Pb reported here. In Fig. 1, an example of the initial Pb change (in  $10 \times$ 10<sup>6</sup> years) is illustrated where the  $\mu$ (<sup>238</sup>U/<sup>204</sup>Pb, today's ratio) increased from 1 to 2 at  $4.56 \times 10^9$  years ago (namely, 2.7 times the increase in the U/Pb ratio). However, this argument appears to be untenable because the tie line of data points of Murray and Allende falls on Canyon Diablo and indicates a common primordial Pb. Allende has been thought to be the least terrestrially contaminated meteorite. The U-Th-Pb systematics indicate that this meteorite contains 6 percent more Pb than the concordant value. Even though we use the slightly higher U concentration, which is obtained by activation analysis (35), in our measurements there is still 5 percent more Pb than the concordant value. Because the C2 chondrite, Murray, also contains 3 percent more Pb than the concordant value, the most plausible interpretation is that the black matrix,

their I-Xe chronology and the ex-

which is enriched in volatile elements (27), accreted later than the other meteorites, even though the white inclusions in the carbonaceous chondrites are older.

We are presently investigating further the initial Pb problem by means of an internal isochron. However, the small initial Pb change discussed above hardly affected the ages of achondrites because these leads are very radiogenic and the time difference is significant.

In conclusion, we observed clear evidence for a time difference either in the formation of meteorites or in metamorphic events. Averaging the ages of the same type of meteorites or of a class of meteorites may not have significance beyond enabling us to obtain a mean age of formation. The older ages reported here,  $4.56 \times 10^9$ years for Angra dos Reis and  $4.57 \times$ 109 years for Beardsley, coincide well with the moon's age. The moon's model age, originally reported from the Apollo 11 study and further documented from subsequent missions as 4.63 to  $4.65 \times 10^9$  years (26), is recalculated to be 4.56 to  $4.58 \times 10^9$ years on the basis of new U decay constants. This mutual consistency in the age allows us to state the age of these members of the solar system as about  $4.57 \times 10^9$  years. In the present investigation emphasis has been placed on obtaining the precise age measurement, which is of great significance in resolving small time differences, as well as on obtaining the primordial Pb isotopic composition which is the most fundamentally important parameter for Pb isotope research.

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22 JUNE 1973

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Fossil Parasitic Copepods from a Lower Cretaceous Fish

Abstract. Well-preserved Lower Cretaceous fossil copepods related to the superfamily Dichelesthioidea have been collected from the gill chamber of the ichthyodectid fish Cladocyclus gardneri. The fossils provide conclusive evidence supporting recent theories that link caligid copepods, which are parasitic on fish, to the invertebrate-associated siphonostomes. This is the first discovery of fossil parasitic copepods, and they are by far the oldest copepods of any sort known.

Copepod crustaceans, both free-living and parasitic, are abundant in marine and freshwater communities today. In spite of this ubiquity, and a taxonomic rank that implies an origin in Paleozoic times, copepods are extremely rare as fossils. A few fossils of free-living forms are recorded, the earliest from the middle or upper Miocene (10 to 20 million years ago) (1). This is a report of the first discovery of fossil parasitic copepods, dating back approximately 100 million years.

The specimens reported here were found by one of us (C.P.) in the gill chambers of two skulls of the teleost fish Cladocyclus gardneri Agassiz (family Ichthyodectidae) during an acetic acid preparation of the fish by the transfer technique (2). These fish are from the Santana Formation, Serra do Araripe, Ceara, Brazil, which is well known for abundant, uncrushed fish preserved in limestone nodules. The Santana Formation is of Lower Cretaceous age, probably Aptian (3, 4). One skull yielded parts of three copepods, one of them almost complete; the other skull yielded fragments of two or three individuals. The specimens were coated with platinum and examined with a Cambridge Stereoscan microscope.

The fossil copepods are preserved in the round and are solid objects, not exoskeletons. They are composed of a substance identical in appearance to