Age and isotopic relationships among the angrites Lewis Cliff 86010 and Angra dos Reis*

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Abstract—Results of a wide-ranging isotopic investigation of the unique Antarctican angrite LEW-86010 (LEW) are presented, together with a reassessment of the type angrite Angra dos Reis (ADOR). The principal objectives of this study are to obtain precise radiometric ages, initial Sr isotopic compositions, and to search for the erstwhile presence of the short-lived nuclei ^{146}Sm and ^{26}Al via their daughter products. The isotopic compositions of Sm, U, Ca, and Ti were also measured. This allows a detailed appraisal to be made of the relations between, and the geneology of, these two angrites.

LEW proves to be severely contaminated with modern terrestrial Pb, which is shown to result from terrestrial weathering. Nevertheless, concordant Pb-Pb model ages of pyroxene separates were obtained (2σ): 4.55784 ± 52 Ga for LEW and 4.55780 ± 42 Ga for ADOR. Uranium isotopic compositions are normal within error. The inferred initial Pb isotopic composition is within error of primordial Pb, as defined by Cañon Diablo troilite. Because of the extreme U/Pb ratios of both angrites, this places an upper limit of ~ 2 Ma on the time between volatile element-loss of the angrite parent body or its precursor planetesimals and the final crystallization of the angrites as differentiates. Initial $^{87}Sr/^{86}Sr$ ratios were found to be indistinguishable in LEW, ADOR, and the cumulate eucrite Moore County. The derived initial $^{87}Sr/^{86}Sr$ for all three meteorites is 0.698970 ± 15 (2σ). A ^{147}Sm-^{143}Nd isochron for LEW was obtained, yielding an age of 4.553 ± 34 Ga with an initial $^{143}Nd/^{144}Nd$ of 0.506682 ± 49 (2σ). ^{146}Sm-^{142}Nd isotope systematics were also measured. Anomalies on ^{142}Nd arising from extinct ^{146}Sm were clearly resolved, resulting in an initial $^{146}Sm/^{144}Sm$ of 0.0071 ± 17 with $\epsilon_{Nd}^{142} = -2.57 \pm 0.62$ at the time of isotopic closure. Both of these Sm-Nd methods imply derivation of LEW from a reservoir with chondritic Sm/Nd ratio; they are also quite consistent with the previously reported systematics for ADOR. Overall, the age and isotopic similarities between LEW and ADOR are striking; it suggests almost simultaneous production on the same asteroid, even though recent experimental studies imply that the two are not comagmatic.

Calcium and titanium do not exhibit enrichments in the *n*-rich isotopes, in contrast to CAIs, making it unlikely that the angrite parent body is inherently rich in early condensates from the solar nebula. No evidence was found for live ^{26}Al in LEW. When combined with the Pb-Pb age and initial $^{87}Sr/^{86}Sr$ data, this firmly excludes ^{26}Al being an important heat source in the early solar system; heat supplied for, for example, (1) the differentiation of the angrite and eucrite parent bodies, and (2) metamorphism of the chondrite parent bodies must come from some other source. The debate over live vs. extinct ^{26}Al in the earliest solar system remains unresolved by this new data from LEW, however.

Both the angrite and eucrite parent bodies have extremely low Rb/Sr and high U/Pb ratios compared to the solar nebula. Therefore, the common initial $^{87}Sr/^{86}Sr$ of 0.698970 can be firmly associated with the U-Pb angrite age above, and the formation of both bodies. This enables an absolute chronology of the early solar system to be established. Based upon $^{87}Sr/^{86}Sr$ differences, the oldest CAIs are a minimum of 11 ± 4 Ma older than the angrites, i.e., 4.569 ± 5 Ga. The age and isotopic constraints are discussed with respect to current collapse, condensation, and accretion timescales calculated for the solar nebula.

INTRODUCTION

THE ACHONDRITE LEW-86010 is unique but has sufficient mineralogical affinities to the achondrite Angra dos Reis to be tentatively classified as an angrite by MASON (1987). This has solicited considerable interest, not least for the reason that only three angrites are known—Angra dos Reis (1.5 kg) which fell in Brazil in 1869, and two recovered from the ice at Lewis Cliff in Antarctica. These are considerably smaller than Angra dos Reis (ADOR), being 6.9 g for LEW-86010 (hereafter LEW) and 0.6 g for LEW-87051. Although mineral compositions are unique to the angrites, the proportions of phases present differ considerably between them. ADOR is

composed primarily of fassaitic pyroxene, while the two Lewis Cliff specimens contain fassaite with subordinate amounts of plagioclase and olivine (e.g., MASON, 1987; PRINZ et al., 1988).

Aside from its compositional uniqueness, Angra dos Reis attracted particular attention originally because of its primitive initial $^{87}Sr/^{86}Sr$ isotopic composition (WASSERBURG et al., 1977). As reported by these authors, $^{87}Sr/^{86}Sr$ falls markedly below the mean basaltic achondrite value (BABI) as determined by PAPANASTASSIOU and WASSERBURG (1969), and is only slightly elevated from the lowest initial Sr ratio found in Allende CV3 calcium-aluminium-rich inclusions (CAIs) by GRAY et al. (1973). This result suggests that ADOR is extremely old. Moreover, it potentially limits the time period between the first condensation of solids within the solar

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* Dedicated to A. O. Nier on the occasion of his eightieth birthday.

nebula, as recorded in CAIs, and the differentiation of planetary-sized objects. Such an interval would include the processes of solar nebula condensation, accretion of the angrite parent body, its differentiation, and the isotopic closure of ADOR as it crystallized. For a solar Rb/Sr ratio this timescale would be of the order of just a few Ma (WASSERBURG et al., 1977).

The great antiquity of ADOR ($\sim$4.55 Ga) has been demonstrated more directly by radiometric dating using the Sm-Nd (LUGMAIR and MARTI, 1977; JACOBSEN and WASSERBURG, 1984) and Pb-Pb methods (TATSUMOTO et al., 1973; WASSERBURG et al., 1977; CHEN and WASSERBURG, 1981). In addition, there is firm evidence for the presence of now-extinct radionuclides at the time of crystallization—heavy xenon isotopes produced by ^{244}Pu fission (LUGMAIR and MARTI, 1977; WASSERBURG et al., 1977) and ^{146}Sm (LUGMAIR and MARTI, 1977; JACOBSEN and WASSERBURG, 1984).

We present here an extensive set of isotopic results for LEW-86010 and a reassessment, with new measurements of Angra dos Reis. Our studies, part of a larger consortium study of LEW-86010 led by M. Prinz, are aimed primarily at providing constraints on the crystallization age of LEW and determining what, if any, affinity it has with ADOR. However, this also allows some assessment to be made of the nature of the precursor materials involved and upon the timescale of solar nebula condensation. A variety of isotopic results is presented:

1) Precise initial ^{87}Sr/^{86}Sr ratios for LEW and ADOR have been measured and compared with the cumulate eucrite Moore County.

2) Both Pb and Nd isotopic ages have been obtained on LEW together with a new Pb model age for ADOR.

3) Internal Al-Mg isotope systematics have been examined to assess whether "live" ^{26}Al was present in the angrite parent body at the time of crystallization.

4) Calcium and titanium isotope compositions were measured to test the hypothesis of PRINZ et al. (1988) that the angrites originate from melting of highly refractory CAI-type material.

5) Neutron capture effects in ^{149}Sm/^{150}Sm due to cosmic ray exposure have been measured in LEW. These and those found in ADOR are discussed in the context of noble gas-derived cosmic-ray exposure ages.

Petrological and geochemical studies of LEW have been reported previously (e.g., MASON, 1987; PRINZ et al., 1988; GOODRICH, 1988; DELANEY and SUTTON, 1988; CROZAZ and MCKAY, 1990; MITTLEFEHLDT and LINDSTROM, 1990) as were noble gas data (EUGSTER et al., 1991; HOHENBERG et al., 1991) and oxygen isotope results (CLAYTON and MAYEDA, 1989). Preliminary data from our study were reported by LUGMAIR et al. (1989) and by LUGMAIR and GALER (1989).

EXPERIMENTAL PROCEDURES

Sample Processing

A piece of LEW-86010 weighing 0.98 g was received from the Meteorite Working Group (Johnson Space Center) for the consortium study. Fractions of ADOR were available from a previous study (LUGMAIR and MARTI, 1977). All sample preparation was done under clean room conditions. Exterior fusion crust was removed from LEW with the aid of a diamond drill. A chunk of 150 mg was then removed and sent to C. Hohenburg for noble gas analysis (cf. HOHENBERG et al., 1991). The remainder was then gently crushed in a boron carbide mortar and sieved for maximum yield in the 75–125 μm-size fraction. A representative "whole-rock" split (LEW-WR) was then taken for isotope work from this size fraction; 60 mg was sent to H. Palme for neutron activation analysis along with a 35 mg split of the finer fraction 45–75 μm.

Mineral separates were prepared from the 75–125 μm fraction using a clean Franz Isodynamic magnetic separator followed by hand picking. This resulted in pure (>99%) separates of plagioclase (LEW-Plag), olivine with kirschsteinite exsolution lamellae (LEW-Ol), and titanian fassaite pyroxene (LEW-Px). Because the clinopyroxene is chemically zoned (CROZAZ and MCKAY, 1990), small separates of interior, light, pinkish pyroxene (LEW-PxLt) and exterior, darker brown pyroxene (LEW-PxDk) were also produced for analysis; they represent very small fractions of a much larger pool of pyroxene. All trace element and isotopic analysis other than those for the Rb-Sr system were obtained on the same dissolutions of these mineral separates and whole rock. Small ($\sim$5 mg) splits of the mineral separates were sent to R. N. Clayton for oxygen isotope analysis (cf. CLAYTON and MAYEDA, 1989). Although MASON (1987) noted trace merrillite in the thin section of LEW, none was found in the nonmagnetic fraction processed. A $\sim$20 mg separate of unsieved, relatively coarse ADOR fassaite (ADOR-Px) was also prepared. This was used to obtain Pb isotopic data on ADOR for intercomparison with LEW.

Rb-Sr analyses for determining the initial ^{87}Sr/^{86}Sr were performed first using much smaller quantities of LEW and ADOR. Three small chips of ADOR ($\sim$3 mg) devoid of obvious phases other than fassaite were dissolved, together with a small separate of plagioclase from LEW; a solution of Moore County plagioclase was already available from a previous study by CARLSON and LUGMAIR (1988).

Washing, Dissolution, and Splitting

All the mineral separates of LEW and ADOR were gently washed in cold 2 N HCl (Pb blank $\sim$ 0.1 pg·mL^{-1}) after weighing and prior to dissolution. However, LEW-WR was analysed without washing as this would have removed any trace merrillite present. Mineral separates destined for Pb isotope analysis were ultrasonically agitated in 1.5 mL HCl with a trace of doubly distilled MeOH for 5 min in a closed Teflon beaker. The liquid was then allowed to wet fully the inside surfaces of the beaker and cap to catch any splashes from the ultrasonication, before pipetting the supernatant into a clean beaker. As will be discussed later, this was done either once (LEW-Plag), five times (LEW-Ol, LEW-PxLt, LEW-PxDk) or ten times (LEW-Px, ADOR-Px). The sample was then washed in a similar way, once only, with H$_2$O followed by decanting and rinsing with methanol. All washings of a particular sample were pooled into a single beaker for analysis.

Dissolution was then effected in the same beaker used for washing by the addition of HF/HNO$_3$ and application of gentle heat. The sample was eventually brought into total solution in HCl for aliquoting. In general, the parent solution was aliquoted into three parts: $\sim$5% spiked for Rb, Sr, Sm and Nd; $\sim$5% spiked for Pb; $\sim$90% for Pb, Nd, Sm, (Sr) isotopes. This scheme allowed quantitative isotope dilution determinations of Pb, Rb, Sr, Sm, and Nd without introducing unnecessary Pb blank from the other spike solutions. Neodymium, samarium (and in some cases strontium) isotopic compositions were measured on the 90% split after processing through the lead column. This fraction was also spiked for U after the Pb column, and therefore the determined U abundances are strictly lower limits—this was done to avoid unnecessary Pb blank from the ^{236}U spike. However, spiking with ^{236}U also allowed verification of the U isotopic composition. Lead abundances were determined from the second smaller 5% split using a high purity ^{206}Pb spike with ^{208}Pb/^{206}Pb = 0.000141 ± 2 (NBS-991). After separation of Pb from this split the remaining solution was, in some cases, used for determining Mg, Ca, and Ti isotopic compositions.

Pb Isotopes

Since this is one of the first contributions in which Pb data are reported from this laboratory, we will describe our techniques in some detail.

Chemistry

The chemical separation procedure for Pb closely followed that described by GALER (1986) which, in turn, was adapted from techniques developed in Paris (MANHÈS et al., 1984; GÖPEL et al., 1985). After aliquoting in HCl and spiking the 5% split with ^{206}Pb tracer, samples were dried down and converted to Br^--form. The columns used were fabricated of shrink-fit Teflon fitted with a polypropylene frit, containing a 50 μL anion exchange resin bed (AG-1, $\times 8$ in NO_3^--form) and a ~ 1 mL reservoir. Separation was achieved by using HBr-HNO_3 mixtures, which is more efficient than using HBr alone followed by stripping of the column with dilute HNO_3. Relevant distribution coefficients for Pb, competing elements, and the remaining elements of interest can be found in KORKISCH and HAZAN (1965), and STRELOW and TOERIEN (1966); those for HBr-HNO_3 mixed media are presented in STRELOW (1978), STRELOW and VAN DER WALT (1981), and VAN DER WALT et al. (1982). The elution behaviour of Pb under these conditions is, in general, bracketed by Zn on the one hand, and by Cd and Bi on the other. As discussed in GALER (1986), optimal retention of Pb while major elements and Zn are eluted occurs with "Solution-A" (0.2 N HBr–0.5 N HNO_3). Lead can then be preferentially taken off the column, leaving other elements such as cadmium and bismuth behind, using "Solution-B" (0.03 N HBr–0.5 N HNO_3). None of the other elements of interest in this study have a significant distribution coefficient using either of these two solutions. All samples were centrifuged in Solution-A just prior to loading onto the column. Any precipitate which might have been present was later recombined with the major element effluent from the column. It is emphasized that, although care was taken to ensure maximal column yields in each case, this may slightly affect the U concentrations, as the ^{236}U tracer was added after passage through the Pb column.

Procedural blanks

All reagents were distilled either once or twice in a quartz sub-boiling still except for H_2O and HF. HF was distilled once in a Teflon two-bottle still, and H_2O was taken directly from a commercial ion exchange purification system. The common Pb content of the reagents in $pg \cdot mL^{-1}$ was ~ 0.05 for water and ~ 0.08–0.5 for the mineral acids (1 pg Pb $\approx 4 \times 10^8$ atoms ^{204}Pb). For safety's sake, all the mineral separates and LEW-WR were passed twice through the Pb column to remove possible trace impurities from the first pass; the leaches of the mineral separates were only passed through once, however. Experience has shown that this extra pass is unnecessary for routine silicate or carbonate samples. The procedural Pb blank for the column separation (one pass) is ~ 1.2 pg.

Blanks were run alongside samples at every stage. This included all HCl washing steps—the sample and blank were agitated side-by-side, and approximately the same amount pipetted out in each case. The amount of reagent used in dissolution, and all heating and drying steps were applied to both equally. Both sample and blank solutions were aliquoted in the same proportions and spiked consecutively. Each batch of Pb column chemistry entailed processing of the 90% and 5% splits of the sample and its corresponding blanks, respectively.

Mass spectrometry

After separation, the Pb was loaded onto a flat zone-refined Re filament along with silica gel and phosphoric acid using leached lime glass pipettes (loading blank: ~ 0.2–0.3 pg Pb). Considerable care was taken to reproduce the heating conditions during loading as well as the quantity of reagent used between the samples and the Pb standards. In addition, the total amount of Pb loaded for isotope composition runs in each instance (~ 1–7 ng) was kept comparable to the 5 ng Pb loaded as standards. In some cases this meant that only

very small splits of the Pb extracted were loaded (e.g., LEW-WR and the leachates). Data were obtained by peak-switching on the La Jolla Micromass 54E single-collector machine, in all cases using the Faraday cup. Hydrocarbons were monitored during filament warmup by spectrum scans. In every case except sample ADOR-Px, which will be discussed later, the mass-204 hydrocarbon was determined to be ~ 100 ppm or less of the $^{204}Pb^+$ peak at the start of data collection. Data in this study were obtained at temperatures between 1120 and 1160°C, and with beam currents of 0.3–5 $\times 10^{-12}$ A $^{208}Pb^+$ depending on the sample size. The NBS-981 standards measured concurrently yielded a fractionation factor of 0.120 $\pm$ 0.027% AMU^{-1} (2σ external of 15 runs with the same batch of silica gel; Table 1d) for all ratios relative to accepted values (TODT et al., 1984). Ionization efficiencies were ~ 3–5% for standards of this size.

Treatment of errors

A proper treatment of errors during Pb isotopic data reduction is extremely complicated. The reason for this is that unlike other radiogenic isotope systems, the raw data is usually highly correlated between the ratios; this is especially true for radiogenic samples. These covariances have a marked effect on the propagation of errors. The errors on the fractionation and blank corrected ratios may be increased or decreased as a result. In addition to the isotope ratios themselves, this must also be taken into account for the final Pb concentration, μ (i.e., the $^{238}U/^{204}Pb$ ratio), and calculated Pb-Pb and U-Pb model ages, for example.

The propagation of uncertainties can be done using approximate analytical formulae or by Monte Carlo simulation. The method used, however, was adapted from the generalized numerical algorithm discussed by RODDICK (1987). This makes use of finite difference approximated partial derivatives in a truncated Taylor series propagation of the variances and covariances of measured quantities into those of the parameters desired. This allows the error propagation to be applied to all parameters and their covariations at the same time, and through each step involved. The resulting uncertainties, of course, account only for the formal uncertainties associated with the measurements. Accepted values for decay and other constants were used after STEIGER and JÄGER (1977)—$\lambda_{238} = 0.155125$ Ga^{-1}, $\lambda_{235} = 0.98485$ Ga^{-1}, $\lambda_{232} = 0.049475$ Ga^{-1}, and 137.88 for the $^{238}U/^{235}U$ ratio today. Primordial Pb isotopic compositions follow from TATSUMOTO et al. (1973) and GÖPEL et al. (1985).

LEAD ISOTOPIC RESULTS

Lead isotopic data were obtained on unwashed LEW-WR and washed mineral separates of LEW-Plag, LEW-Ol, LEW-Px, and small samples of interior (LEW-PxLt) and exterior (LEW-PxDk) pyroxene. New data were also measured for ADOR pyroxene (ADOR-Px) to facilitate intercomparison between the derived ages. Results for the pooled HCl leachates and residues are given in Table 1a–d. This is divided as follows: Table 1a contains Pb and U abundances, and Pb isotopic compositions of LEW-WR and in the mineral separates after washing; Table 1b contains Pb-Pb and U-Pb model ages on the samples reported in Table 1a; Table 1c contains total Pb procedural blanks (in picograms) measured during this study; lastly, Table 1d reports Pb abundances and isotopic compositions of the leaches. Data for the standard NBS-981 Pb and the isotopic compositions used for blank correction are also reported in Table 1d.

Terrestrial Contamination

The washing of the LEW mineral separates resulted in the removal of minor Fe staining present on some of the grains and was accompanied by the evolution of H_2S. The only

Table 1a. U–Pb isotopic results

Sample	Weight (mg)	^{206}Pb (nmol·g^{-1})	Pb (ppm)	U (ppm)		^{206}Pb/^{204}Pb	^{207}Pb/^{204}Pb	^{208}Pb/^{204}Pb	^{204}Pb/^{206}Pb	^{207}Pb/^{206}Pb	^{208}Pb/^{206}Pb
LEW-WR	56.30	12.60 (6)	10.300 (51)	0.1083 (3)	m†	18.891 (1)	15.969 (1)	38.670 (4)	0.052934 (3)	0.84539 (1)	2.04698 (12)
					f	18.937 (10)	16.026 (13)	38.856 (42)	0.052807 (29)	0.84640 (23)	2.0519 (11)
LEW-Plag	52.09	0.0806 (12)	0.05186 (77)	0.01214 (4)	m	37.006 (31)	27.197 (22)	49.848 (45)	0.027023 (22)	0.73493 (13)	1.34707 (55)
					f	37.095 (37)	27.295 (31)	50.087 (70)	0.026958 (26)	0.73581 (24)	1.3503 (9)
					fb	37.167 (61)	27.341 (44)	50.131 (77)	0.026906 (44)	0.73562 (27)	1.3489 (13)
LEW-Ol	44.04	0.1636 (15)	0.1291 (12)	0.006686 (10)	m	20.067 (9)	16.672 (9)	38.725 (24)	0.049834 (22)	0.83079 (18)	1.92964 (54)
					f	20.115 (14)	16.732 (16)	38.910 (48)	0.049714 (35)	0.83179 (29)	1.9343 (12)
LEW-PxLt	4.17	0.226 (18)	0.128 (10)	0.06150 (7)	m	96.75 (1.52)	63.28 (95)	104.87 (1.63)	0.01045 (18)	0.65401 (48)	1.0851 (14)
					f	96.98 (1.52)	63.51 (95)	105.37 (1.64)	0.01043 (18)	0.65480 (51)	1.0877 (15)
					fb	100.6 (3.0)	65.7 (1.8)	108.5 (2.8)	0.01006 (31)	0.6533 (11)	1.0792 (58)
LEW-PxDk	9.03	0.4315 (74)	0.2317 (40)	0.1020 (2)	m	285.1 (5.4)	181.0 (3.4)	273.5 (5.2)	0.003520 (60)	0.63494 (8)	0.95912 (18)
					f	285.8 (5.4)	181.7 (3.4)	274.8 (5.2)	0.003512 (60)	0.63570 (19)	0.96142 (55)
					fb	294.6 (8.4)	187.2 (5.2)	282.6 (7.7)	0.003406 (92)	0.63528 (34)	0.9591 (17)
LEW-Px	30.51	0.3761 (16)	0.19308 (80)	0.0841 (2)	m	672.6 (3.9)	421.7 (3.4)	574.4 (1.3)	0.001482 (9)	0.62689 (13)	0.85380 (39)
					f	674.2 (3.9)	423.2 (3.4)	577.2 (1.4)	0.001478 (9)	0.62764 (21)	0.85585 (60)
					fb	692.5 (13.2)	434.6 (8.6)	592.2 (10.4)	0.001439 (28)	0.62749 (24)	0.8549 (9)
ADOR-Px	21.89	0.7917 (25)	0.3760 (12)	0.1828 (6)	m	5113 (227)	3211 (210)	3507 (203)	0.000200 (4)	0.62107 (4)	0.67536 (5)
					f	5125 (228)	3223 (211)	3524 (204)	0.000200 (4)	0.62182 (17)	0.67698 (37)
					fb	5922 (684)	3723 (470)	4067 (495)	0.000173 (18)	0.62171 (19)	0.67629 (59)

NBS-981 Pb data are reported in Table 1d. Blank isotopic composition used and measured procedural Pb blanks are given in Tables 1d and 1c, respectively. The errors on data other than the measured isotope ratios were obtained by numerical propagation, as described in the text. (†) *m*: measured ratios. *f*: corrected for fractionation, *fb*: corrected for fractionation and blank.

Table 1b. Pb–Pb and U–Pb model ages

Sample	$^{238}U/^{204}Pb$† μ	$^{207}Pb^*/^{206}Pb^*$	Model Age (Ga)†		
			$^{207}Pb–^{206}Pb$	$^{238}U–^{206}Pb$	$^{235}U–^{207}Pb$
LEW-WR	0.6789 (36)	0.59545 (72)	4.4969 (17)	17.536 (37)	7.170 (7)
LEW-Plag	23.35 (36)	0.61187 (42)	4.5364 (10)	5.062 (54)	4.693 (15)
LEW-Ol	3.429 (33)	0.59565 (76)	4.4974 (19)	9.178 (47)	5.646 (10)
LEW-PxLt	113.0 (9.6)	0.6071 (10)	4.5251 (25)	3.78 (22)	4.280 (79)
LEW-PxDk	289.4 (9.0)	0.61993 (37)	4.5554 (9)	4.410 (56)	4.510 (18)
LEW-Px	648.0 (12.2)	0.62097 (22)	4.55784 (52)	4.652 (16)	4.5866 (50)
ADOR-Px	5557.5 (562.4)	0.62095 (18)	4.55780 (42)	4.586 (15)	4.5663 (47)

Errors shown were propagated from the data reported in Table 1a. (†) Since U abundances in Table 1a are minimum values (see text), the μ given are minima and the U–Pb model ages maxima.

separate that appeared to suffer some attack during this process was LEW-Ol.

The most striking feature of these leaching experiments is the quantity of readily soluble Pb removed by this procedure—this is equivalent to approximately 6–11 ppm Pb relative to the pre-leaching sample weights (Table 1d). In every case >96% of the total Pb present was removed. Given that only soluble surficial phases were dissolved, this implies a Pb concentration of hundreds to perhaps thousands greater in such phases. The terrestrial origin of this contamination is indicated both by the high Pb contents and by its isotopic composition, which is similar to 'modern terrestrial' (MT) Pb derived from the upper continental crust (Fig. 1). The leaches from LEW and ADOR appear to be isotopically distinct from one another (Fig. 1). In terms of the amount of Pb leached, the LEW results contrast markedly with that for the ADOR pyroxene, as can be seen in Table 1d. The Pb washed from ADOR-Px amounts to a mere ~6% of the total. Nevertheless, 6% still constitutes a significant quantity of that present, and indeed most of the ^{204}Pb in this separate.

In the leaches of both meteorites, a small radiogenic component is present, evident in the slightly elevated $^{207}Pb/^{204}Pb$ ratios relative to MT Pb (Fig. 1). The amount can be determined from Fig. 1 using the lever rule. This component is more marked in the case of the ADOR leach and is probably due to the dissolution of a small amount of merrillite during the washing procedure. In fact, using the ADOR merrillite data of CHEN and WASSERBURG (1981) it would require just ~10 μg merrillite (~0.05% of the separate) to be present in order to explain the isotopic shift.

Because of the extreme Pb contamination of LEW, one would like to be able to identify both the source of the Pb and the mechanism by which it was introduced. One clue from Table 1d is that the amount of Pb present in LEW-WR and removed from the LEW separates varies by only a factor of about two on a per mass basis. This indicates that contamination is pervasive and is not unduly determined by the local chemistry within the meteorite interior.

The Pb isotopic composition of the leachates does not correspond to the common Pb ores used in gasoline, such as the 'Broken Hill' and 'Mississippi Valley' types—these are far less and far more radiogenic, respectively. However, we note that there is isotopic overlap with some measured gasoline leads (CHOW and EARL, 1972). Although this form of contamination cannot be excluded entirely, it is doubtful whether this could result in such a pervasive interior distribution. We therefore consider it highly unlikely.

This raises the possibility that contamination was introduced inadvertently during sample processing. To address this question, grain mounts of LEW plagioclase and pyroxene were examined under the SEM and compared with ADOR pyroxene. Interior grain surfaces of the LEW minerals produced during crushing were found to be smooth. In contrast, the original inter-grain surfaces of both the plagioclase and

Table 1c. Pb blanks (pg)

Sample	Residue splits		Leach splits	
	5%	90%	10%	90%
LEW-WR	2.2	nm	—	—
LEW-Plag	3.4	27.3†	nm	nm
LEW-Ol	3.5	4.8	11.7	nm
LEW-PxLt	1.8	~8.2	nm	nm
LEW-PxDk	6.4	7.6	8.7	nm
LEW-Px	2.1	4.0	3.0	25.0
ADOR-Px	2.5	7.0	2.0	nm
Avg.‡	4 ± 3	6 ± 4		

Pb blanks in picograms of common lead; nm: not measured. (†) Probably spurious (see text). (‡) Values used for blank correction in Table 1a.

Table 1d. Leach Pb isotopic results

Sample	^{206}Pb (nmol·g^{-1})	Pb (ppm)	Washed Pb††		^{206}Pb/^{204}Pb	^{207}Pb/^{204}Pb	^{208}Pb/^{204}Pb	^{204}Pb/^{206}Pb	^{207}Pb/^{206}Pb	^{208}Pb/^{206}Pb
L-LEW-Plag	13.585 (66)	11.202 (54)	99.5%	m†	18.386 (2)	15.667 (2)	38.115 (9)	0.054389 (6)	0.85203 (2)	2.07317 (36)
				f	18.430 (10)	15.723 (13)	38.298 (42)	0.054258 (30)	0.85306 (23)	2.0781 (12)
L-LEW-Ol	8.748 (14)	7.206 (12)	98.2%	m	18.368 (4)	15.641 (2)	38.018 (7)	0.054443 (11)	0.85147 (1)	2.06967 (26)
				f	18.412 (11)	15.697 (13)	38.200 (42)	0.054312 (31)	0.85249 (23)	2.0746 (11)
L-LEW-PxDk	7.342 (7)	6.021 (6)	96.3%	m	18.578 (2)	15.759 (4)	38.176 (10)	0.053828 (7)	0.84838 (8)	2.05541 (9)
				f	18.622 (10)	15.815 (13)	38.359 (42)	0.053699 (30)	0.84940 (24)	2.0603 (11)
L-LEW-Px	8.286 (9)	6.799 (7)	97.2%	m	18.587 (6)	15.778 (6)	38.250 (16)	0.053799 (16)	0.84883 (9)	2.05792 (4)
				f	18.632 (11)	15.835 (14)	38.434 (44)	0.053670 (33)	0.84985 (25)	2.0629 (11)
L-ADOR-Px	0.0281 (16)	0.0222 (13)	5.6%	m	20.088 (32)	16.509 (27)	39.004 (61)	0.049785 (79)	0.82174 (17)	1.94143 (41)
				fb	20.156 (38)	16.582 (32)	39.197 (77)	0.04962 (9)	0.82258 (30)	1.9444 (16)
Blank‡				m	18.75 (50)	15.63 (10)	38.81 (1.00)	0.0533 (14)	0.8336 (20)	2.0699 (50)
NBS-981§				m	16.896 (8)	15.437 (11)	36.528 (36)	0.059185 (30)	0.91366 (28)	2.1619 (13)
(% AMU^{-1})					0.122 (25)	0.119 (23)	0.121 (25)	0.122 (26)	0.114 (31)	0.120 (31)

Concentrations are reported relative to the sample weights of Table 1a. (††) The fraction of the total Pb, in % by weight, removable by washing with dilute HCl. (†) Symbols given in Table 1a. (‡) Isotope ratios used for blank correction. (§) External precision (2σ) of 15 runs, 5 ng standards; bias factors are quoted relative to NBS-981 ratios of TODT et al. (1984). A value of 0.120±0.027% AMU^{-1} was used for bias correcting the data here and in Table 1a.

Table 1e. Published ADOR Pb–Pb ages

Sample[†]	Phase[‡]	$^{204}Pb/^{206}Pb$	$^{207}Pb^*/^{206}Pb^*$	Age (Ga)	Source
Angra dos Reis	wr	0.00466	0.6197 (21)	4.555 (5)	TATSUMOTO et al. (1973)
Total-A[§]	wr	0.00162	0.6171 (4)	4.549 (1)	WASSERBURG
whitlockite-A[§]	me	0.00133	0.6158 (4)	4.546 (1)	et al. (1977)[††]
whitlockite-B[§]	me	0.00452	0.6163 (4)	4.547 (1)	
whole rock[§]	wr	0.00523	0.6179 (18)	4.551 (4)	CHEN and
whitlockite[§]	me	0.00255	0.6190 (35)	4.553 (8)	WASSERBURG (1981)
ADOR-Px	px	0.000173	0.62095 (18)	4.55780 (42)	This study
LEW-Px	px	0.001439	0.62097 (22)	4.55784 (52)	

(†) Sample name as reported. (‡) Phase analysed (wr: whole rock, me: merrillite, px: pyroxene). (§) Not washed with HCl prior to dissolution. (††) Data corrected by 0.1% AMU^{-1} as outlined in CHEN and WASSERBURG (1981).

the pyroxene have a "frosted" or "mottled" appearance—this was not seen on the ADOR pyroxene grains. In addition, small ~10 μm pits were noticed in which there were euhedral platy crystals. Semi-quantitative EDS spectra were obtained on smooth surfaces and compared with the mottled surfaces and these secondary crystals. The crystals were found to contain mainly Fe > S > K; mottled surfaces also showed evidence for excess S and K compared to the smooth surfaces. Since H_2S was evolved during HCl leaching, the S is probably not present as pyrite. These results are quite consistent with the recent study of weathering in Antarctic eucrites by MITTLEFEHLDT and LINDSTROM (1991). Notably, they found ubiquitous examples of trace element anomalies in Ce, K, and halides, with secondary phases consisting of "rust," a phase rich in K and S, and occasional gypsum; even interior samples were affected. This observation strongly implies that the excess Pb in LEW is not a result of laboratory contamination, but rather terrestrial exposure. In this connection we note that the ^{81}Kr terrestrial age of LEW is quite large at 420 ± 100 ka (EUGSTER et al., 1991). Mineralogically, terrestrial weathering products appear to be pervasive within Antarctic meteorite interiors (e.g., GOODING, 1983; BUCHWALD and CLARKE, 1989).

The "Pb contamination" problem has been discussed by many workers (e.g., UNRUH, 1982; TERA, 1983). In the case of chondrites, it has been felt that this often arises during sample preparation; for iron meteorites this is demonstrably associated with sawing (GÖPEL et al., 1985). In contrast to previous investigations, therefore, these results clearly show that severe Pb contamination can occur during relatively mild, but prolonged, exposure in the terrestrial environment. This is important to bear in mind with regard to strategies for the analysis of Pb in meteorites from Antarctica. Although

R. Score noted that "oxidation is present" in the hand specimen of LEW (MASON, 1987), this alteration is not at all severe and the minerals are fresh.

One clue as to the origin of the contamination comes from the terrestrial exposure history of the ice-borne meteorites found at Lewis Cliff. HUSS (1990) has recently reviewed the relevant data for infall rates and accumulation times for many Antarctic blue ice fields. He concluded that concentration in these areas typically occurs as wind ablation erodes vertically flowing ice. For a nominal 100 g-sized chondrite, he suggested a 10 ± 5 × 10^3 year "weathering lifetime" during which time the meteorite rests on the ice surface before complete disaggregation. Processes of alteration and freeze-thaw can presumably explain the grain surface textures seen under the SEM. Based upon the photographic catalogue of YANAI and IGUCHI (1981), many meteorites from the Antarctic collections appear to be heavily weathered and oxidized in this manner. The rapidity of the weathering process, even under the largely dry Antarctic conditions, is discussed by GOODING (1983).

Despite this, the ultimate source of the Pb remains something of a mystery in the case of LEW. Uncontaminated ice has been shown to contain very little Pb (<2 pg·g^{-1}) by NG and PATTERSON (1981) and BOUTRON and PATTERSON (1983). This suggests that the bulk of the Pb was introduced during the surficial weathering process and not within the ice itself. GOODING (1983) and BUCHWALD and CLARKE (1989) have suggested that the terrestrial weathering components may be derived from the breakdown of wind-borne dusts blown onto the meteorites during surface exposure. This is quite consistent with the Pb isotopic compositions observed, which resemble "average" upper crustal Pb (e.g., ALBARÈDE and JUTEAU, 1984). However, for Pb abundances typical of
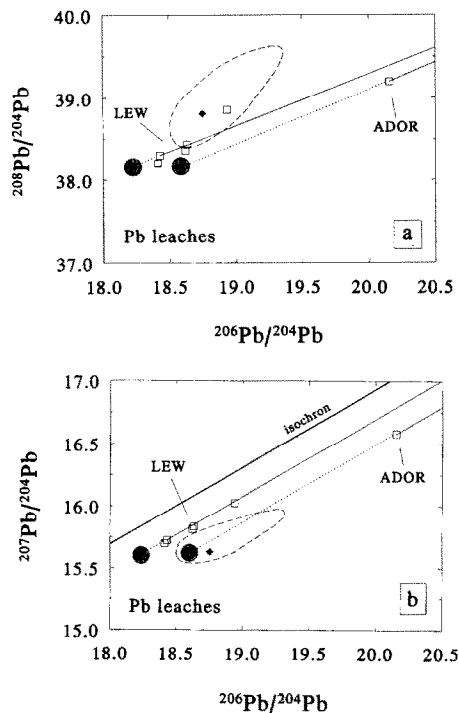
FIG. 1. Lead isotopic compositions of leaches of the LEW mineral separates, LEW-WR whole rock, and the leach of ADOR-Px. In (a) $^{208}Pb/^{204}Pb$ is plotted vs. $^{206}Pb/^{204}Pb$, and in (b) $^{207}Pb/^{204}Pb$ vs. $^{206}Pb/^{204}Pb$. In order of increasing $^{206}Pb/^{204}Pb$ the LEW leach points correspond to L-LEW-Ol, L-LEW-Plag, L-LEW-PxDk, L-LEW-Px, and then the unleached whole-rock sample LEW-WR (cf. Table 1d); the ADOR leach point is for L-ADOR-Px. The thick solid line in (b) is a Pb-Pb isochron with an age of 4.5578 Ga drawn through primordial Pb. Lines have been drawn (solid) between L-LEW-Plag and the residue LEW-Plag (cf. Table 1a), and between L-ADOR-Px and ADOR-Px. The extensions of these lines are shown dotted. The field bounded by the dashed line encompasses Pb isotopic compositions of contemporary marine sediments (BEN OTHMAN et al., 1989). The filled diamond is the zero-age Pb isotopic composition of the conformable ore Pb growth curve (ALBARÈDE and JUTEAU, 1984). Both of the above are representative of the upper continental crust. The suggested Pb isotopic compositions of the LEW and ADOR leach endmembers are drawn as stippled circles, and appear to be quite distinct from one another. Nevertheless, the leachable Pb in both cases is clearly of terrestrial origin and probably derived from the upper crust.

upper crustal rocks (~20 ppm) one would have to scavenge Pb from a dust mass that is a significant fraction of the mass of LEW to explain the degree of contamination. Whether this is feasible from this point of view is unclear.

Extraterrestrial Lead

Despite the unprecedented degree of Pb contamination of LEW, it was possible to obtain highly radiogenic Pb from this meteorite and reliable Pb-Pb and U-Pb age information. Lead data for the washed mineral separates and unwashed whole rock are given in Table 1a; model ages are reported in Table 1b. Some results are reported with errors of more than two significant digits when errors are highly correlated. The data for the washed LEW separates and ADOR-Px have been plotted in Fig. 2.

Procedural Pb blanks, run in parallel as described in the experimental methods section, are reported in Table 1c. These include blanks run together with the mineral separates as well as their associated HCl leaches. All blanks were measured on the Faraday cup to a precision of better than ±0.05 pg Pb. The data in Table 1c indicate good reproducibility and values <10 pg total Pb. The higher reported blank for the 90% split of LEW-Plag was traced to an evaporator air-supply assembly which had rusted post-filtration. Since this particular unit was used for the blank and not the sample, it is therefore considered spurious. Based upon these results, the LEW residue Pb data were corrected using 6 ± 4 pg and 4 ± 3 pg Pb for the 90% and 5% splits, respectively. Blank correction for the leach (except L-ADOR-Px), the LEW-WR, and the LEW-Ol data was unnecessary, and these data presented in Tables 1a and d have been corrected merely for instrumental fractionation.

Regrettably, the unwashed whole-rock data (LEW-WR) are dominated by the terrestrial component and are useless in regard to obtaining primary age information. The concentration of 10.3 ppm is around two orders of magnitude
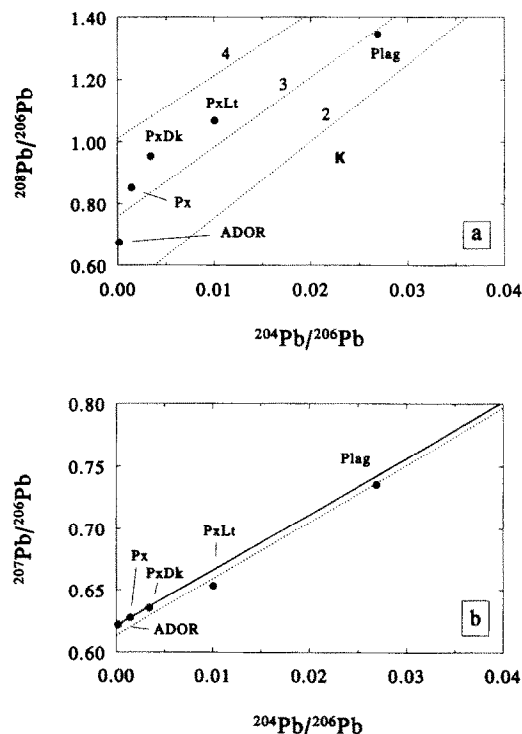

FIG. 2. Lead isotopic compositions of the LEW mineral separates and ADOR-Px after washing to remove terrestrial Pb. In (a) $^{208}Pb/^{206}Pb$ is plotted vs. $^{204}Pb/^{206}Pb$, and in (b) $^{207}Pb/^{206}Pb$ vs. $^{204}Pb/^{206}Pb$. Error bars are far smaller than the symbols shown. The thick solid line in (b) is a reference "isochron" passing through primordial Pb with an intercept corresponding to an age of 4.5578 Ga; also given for reference (dotted) is a younger 4.54 Ga "isochron." Lines of constant κ ($\equiv$ $^{232}Th/^{238}U$) for an age of 4.5578 Ga passing through primordial Pb are shown dotted in (a) for the cases κ = 2.0, 3.0 and 4.0. The low κ values for ADOR-Px and the LEW separates are unlikely to be representative of the whole rocks because of the large quantities of thorogenic Pb contained in merrillite (cf. WASSERBURG et al., 1977; CHEN and WASSERBURG, 1981).

too high to yield a reasonable mass balance with the washed minerals (Table 1a) and also has a MT isotopic composition (Fig. 1). Moreover, in both concentration and isotopic composition it is quite comparable to that found in the leaching experiments (Table 1b; Fig. 1).

Even after repeated washing, the Pb concentration for the olivine plus kirschsteinite residue (LEW-Ol) is also unreasonably high for this phase (0.13 ppm). The Pb isotopic composition is, again, dominated by MT Pb, with U-Pb ages that are extremely high and discordant (Table 1a). In this case washing did not apparently eliminate the MT Pb component completely. We suggest, albeit speculatively, that grain interiors were contaminated along the exsolution lamellae of kirschsteinite and so could not be successfully washed. The olivine leach-residue pair can be used to estimate the precontamination isotopic composition by extrapolating the "unmixing" process further. The $^{206}Pb/^{204}Pb$ ratio of the residue is more radiogenic than the leach, implying that a more radiogenic component is present. This can be specified by the intersection of the internal isochron ($^{207}Pb:^{206}Pb$ ≈ 0.621; see following) with the extrapolation of the leach-residue line in $^{207}Pb/^{204}Pb$-$^{206}Pb/^{204}Pb$ space. Both have excesses of ^{206}Pb relative to this isochron, requiring the line joining them to have a slope > 0.621. In fact, this slope is 0.608 ± 20 which places a lower limit of ~ 42 for the original $^{206}Pb/^{204}Pb$ ratio.

The plagioclase separate from LEW was the first analysed, and the first to identify the extreme MT Pb contamination present. For this reason LEW-Plag was washed less stringently than the other separates—only one HCl wash compared to 5 (LEW-Ol, LEW-PxDk, LEW-PxLt) and 10 (LEW-Px, ADOR-Px). Therefore, we are somewhat less confident that the Pb isotopic results for LEW-Plag accurately reflect indigenous Pb. Nevertheless, the Pb is highly radiogenic ($^{206}Pb/$ $^{204}Pb = 37.17$) for the U-poor phase in LEW, and far more radiogenic than MT Pb.

LEW-PxLt and LEW-PxDk are small interior and exterior separates from a much larger pool (~ 100 mg) of zoned pyroxene from LEW, represented by the separate LEW-Px. The Pb from all these separates is highly radiogenic (Table 1a) indicating that Pb is highly incompatible in pyroxene. In detail, however, LEW-Px has U and ^{206}Pb abundances intermediate between LEW-PxLt and LEW-PxDk (Table 1a). This suggests that the pyroxene zonation pattern of U from core to rim is uniform. However, the original postcrystallization zonation of Pb was less straightforward. This is seen by the observation that the Pb of LEW-Px is far more radiogenic than either LEW-PxLt or LEW-PxDk (Table 1a). In fact the concentrations of ^{204}Pb (in pmol·g^{-1}) are around 0.54, 2.25, and 1.46 in these three phases, respectively. This indicates that the original distribution of Pb was concentrated by factors of ~ 4 in the core and ~ 3 in the rim compared to the bulk pyroxene. This pattern was later dwarfed by radiogenic Pb addition according to the U distribution. The radiogenic $^{208}Pb/^{206}Pb$ ratios of LEW-Px, LEW-PxLt, and LEW-PxDk (not given in Table 1a) correspond to time-integrated $^{232}Th/^{238}U$ ratios of 3.348 ± 12, 3.516 ± 16, and 3.606 ± 7 (2σ), respectively. Taken with the U abundances reported in Table 1a, this indicates that Th by and large fol-

lows the U distribution pattern rather than that of ^{204}Pb. The high incompatibility of U (and Th) is probably the reason why the U abundance increases towards the pyroxene exterior. However, the concentration of ^{204}Pb in the pyroxene core is less simply explained. For example, this is unlikely to be related to accessory phase inclusions, as these would presumably concentrate U as well.

The data for LEW-Px and ADOR-Px presented in Table 1a are the weighted means of two runs. The ADOR-Px data ratioed to ^{204}Pb are relatively imprecise because measurements were not obtained with a multiplier. Hydrocarbon interference was monitored at mass-205 and by spectrum scans. At the start of the ADOR-Px measurements the $^{204}Pb^+$ peak had a hydrocarbon contribution <10% which decreased subsequently. Since the measurement errors are quite large no correction was applied; in any case, the age determination is limited by the precision on the $^{207}Pb/^{206}Pb$ ratio. The measurement errors on $^{206}Pb/^{204}Pb$ and $^{204}Pb/^{206}Pb$ were $\sim 5\%$, increasing to $\sim 11\%$ after blank correction. Both of these ratios are given in Table 1a, as digital filtering of the raw distributions is different. Manual filtering of the raw data for ADOR-Px produced no significant change compared to the ratios reported. ADOR-Px has an extremely radiogenic isotopic composition with a blank-corrected $^{204}Pb/^{206}Pb$ ratio of 0.000173 ± 18 (Table 1a); the ^{204}Pb in the sample amounted to $\sim 1.8 \times 10^9$ atoms.

Uranium Isotopes

The U isotopic composition of several of the LEW samples was also determined together with the U abundances given in Table 1a. Since samples were only spiked with enriched ^{236}U the applied corrections for multiplier discrimination and mass fractionation were based upon standard U measurements; a correction was also needed for contribution to $^{235}U/$ ^{238}U from the spike composition.

The results, expressed as the deviation in per mil from the normal $^{235}U/^{238}U$ ratio ($\equiv 1/137.88$), $\delta^{235}U$, are (2σ): -1.1 ± 2.1 for LEW-WR, -1.9 ± 1.8 for LEW-PxDk, -2.7 ± 3.3 for LEW-Plag, -3.0 ± 3.4 for LEW-Ol, and $+1.1 \pm 1.7$ for LEW-Px. Thus, the U isotopic composition of LEW appears to be within about $\pm 0.3\%$ of normal. This result compares favourably with the previous double-spiked U analyses of ADOR by CHEN and WASSERBURG (1981) who found $\delta^{235}U$ of -1.6 ± 4.0 and $+1.3 \pm 3.7$ for merrillite and whole rock, respectively.

Lead Model Ages

Pb-Pb and U-Pb model ages from the LEW and ADOR samples are presented in Table 1b. The errors on the data shown were derived along with those in Table 1a by the error propagation algorithm described earlier. This results, for example, in the small derived errors in the U-Pb model ages because the high covariance between the errors in μ and the $^{206}Pb/^{204}Pb$ and $^{207}Pb/^{204}Pb$ ratios (among others) has been fully taken into account (cf. Table 1b). No error was implicitly ascribed to the primordial Pb isotopic compositions, taken from TATSUMOTO et al. (1973).

One important point to reiterate is that the U concentrations reported in Table 1a were obtained by spiking after the Pb column separation with the assumption of a 100% column yield. The effect of this may be, in some cases, to slightly underestimate μ and overestimate the U-Pb model ages. In addition, we cannot fully guarantee that the leaching procedure did not introduce some disturbance into the U-Pb systematics by differential leaching, as has been noted by some workers (e.g., BROUXEL and TATSUMOTO, 1991; TATSUMOTO and PREMO, 1991). However, we consider this unlikely.

Overall, the model ages underscore the points raised in the previous section; namely, that both LEW-WR and LEW-Ol are dominated by MT Pb. This is particularly evident in the extreme U-Pb ages, which imply that large excesses of ^{206}Pb and ^{207}Pb are present relative to U. In fact, the Pb-Pb "ages" of these two samples agree within error at 4.497 Ga— a signature of the terrestrial component present. This value is within $\sim$6 Ma of the corresponding model ages of the leaches (not reported in Table 1d), and quite distinct from that of the ADOR-Px leach. Overall, this is a good illustration that MT Pb addition lowers the apparent Pb-Pb model age (provided that the primary model age is 'older' than the MT component). The accompanying effect on the two U-Pb ages (i.e., for radiogenic samples) is to raise both of them with respect to the Pb-Pb age.

In the case of LEW-Plag, the U-Pb model ages are also slightly higher than the Pb-Pb age of 4.5364 ± 10 Ga. We take this to imply that a small quantity of the dominating (99.5%) terrestrial Pb contaminant (Table 1d) was still left after one HCl leaching step. In fact, if the Pb-Pb age was to be in agreement with LEW-Px then only <0.2% of the original leachable Pb component would need to remain. Since this is quite a plausible amount, we suggest that the apparent Pb-Pb age of LEW-Plag is too young. This is only likely to be a problem for this particular separate from LEW as the others were washed far more thoroughly.

The model ages of pyroxene separates LEW-PxLt and LEW-PxDk contrast to those of LEW-WR, LEW-Ol, and LEW-Plag. In particular, their U-Pb model ages are discordant and younger than their Pb-Pb model ages (Table 1b). This effectively rules out the possibility of incomplete removal of the terrestrial Pb component as the cause. Similarly, if the U data had been affected by low column yield this would result in even lower and more discordant U-Pb ages. We can think of no acceptable reason for dismissing these analyses on grounds of contamination, inadequate washing, or an analytical artifact. Our preferred explanation is that they reflect minor re-equilibration of the zoned Pb abundance in pyroxene after intermineral closure. As noted above, the internal (LEW-PxLt) and external (LEW-PxDk) portions contain around four and three times the ^{204}Pb, respectively, than is found in the bulk pyroxene (LEW-Px). The principal effect of such a redistribution would be to diffuse some ^{204}Pb-rich lead from the core and rim into the intervening pyroxene. In partial support of this it is noted that LEW-PxLt contains the higher ^{204}Pb abundance and is correspondingly more discordant than LEW-PxDk. Nevertheless, these effects are unlikely to affect seriously the systematics of the internally closed, far larger and representative pyroxene separate LEW-Px.

The total pyroxene LEW-Px is considered to yield the most reliable crystallization age of LEW for two reasons. Firstly, it is the most radiogenic LEW sample analysed; secondly, it has nearly concordant Pb-Pb and U-Pb model ages. Moreover, due to the fact that the U abundances are minima we are confident that this separate and ADOR-Px are concordant. It can readily be shown that if the U abundance is corrected for incomplete column yield, both U-Pb model ages will decrease and converge onto the Pb-Pb model age (the Pb-Pb model age is unaffected, of course). The amounts of 'analytical U loss' required to bring all model ages into exact agreement are $\sim$2.8% for LEW-Px and $\sim$0.8% for ADOR-Px. This is well within bounds of a reasonable column yield (say >95%). It follows that these two separates do not appear to have suffered any differential U/Pb leaching during washing, of the kind observed by other workers (e.g., BROUXEL and TATSUMOTO, 1991; TATSUMOTO and PREMO, 1991). By inference, this can also be extended to the other LEW separates which were washed less extensively.

In summary, interpretation of the LEW Pb data is complicated by two factors: (1) The severe terrestrial contamination of this meteorite, rendering the LEW-WR, LEW-Ol useless and affecting LEW-Plag data to a far lesser degree; and (2) evidence for a slight postcrystallization disturbance in the case of the small interior and exterior pyroxene separates LEW-PxLt and LEW-PxDk. However, apparently concordant age systematics were obtained for LEW-Px and ADOR-Px. The Pb-Pb age of LEW is then 4.55784 ± 52 Ga with ADOR being 4.55780 ± 42 Ga (2σ; Table 1b). Lead isotopic compositions of the washed LEW separates and ADOR-Px are illustrated in Fig. 2.

Previous Work on ADOR

Lead isotopic analyses of whole-rock samples of ADOR and of merrillite separates have been reported previously by several workers (TATSUMOTO et al., 1973; WASSERBURG et al., 1977; CHEN and WASSERBURG, 1981). Procedural blanks were reduced by factors of 10 or more compared with this previous work (Table 1c). All of these previous data and the results of this study were normalized to accepted values for the NBS-981 or NBS-982 Pb standards (cf. TODT et al., 1984). Measured values for the NBS-981 isotopic standard during this study are presented in Table 1d. As discussed by CHEN and WASSERBURG (1981), the data reported by WASSERBURG et al. (1977) must be additionally corrected for multiplier bias by 0.1% AMU^{-1}, increasing the reported Pb-Pb ages by $\sim$3 Ma.

Age data from all of these studies are summarized in Table 1e. One first order point is that the data for ADOR-Px measured in this study are roughly ten times more radiogenic than had been found previously (Table 1e). It is this, accompanied by the decrease in procedural blank, that accounts for the substantial improvement in precision of the final model age. Unfortunately, taken as a whole, there appears to be quite poor agreement in the model ages found in all studies given the reported errors.

One possible source of this discrepancy might be sample treatment prior to dissolution. Both the data in this study and that of TATSUMOTO et al. (1973) were obtained on HCl-washed samples while this step was not performed (or at least not reported) in the other work. This is especially relevant when it is considered that the TATSUMOTO et al. (1973) Pb-Pb age of 4.555 ± 5 Ga agrees well with our ADOR-Px age (4.5578 ± 4 Ga) but is older than the ages reported by WASSERBURG et al. (1977) and CHEN and WASSERBURG (1981)—addition of terrestrial Pb lowers the apparent Pb-Pb age. The proportion of Pb removed from ADOR-Px by washing amounted to $\sim$5.6% of the total by weight (Table 1d) and bears a clear terrestrial isotopic signature. Although this is a miniscule quantity of Pb compared with

the LEW case, it still has a significant effect on the isotope ratios. For example, the HCl wash has approximately ten times the quantity of ^{204}Pb than is present in the residue. Reconstruction of the ADOR-Px separate prior to washing reduces the ^{206}Pb/^{204}Pb ratio from over 5000 to a value of about 535, with the Pb-Pb age reduced by $\sim$1.9 Ma to 4.5559 $\pm$ 6 Ga. Thus, this brings the ^{206}Pb/^{204}Pb ratio down into the range observed for whole rock and merrillite in the previous studies, but the age still remains higher than the adjusted data of WASSERBURG et al. (1977) but closer to those of CHEN and WASSERBURG (1981). Hence, adding back the HCl wash does not appear to fully account for the age discrepancies. In this context, three other points are worth mentioning. Firstly, if the ADOR-Px data had been undercorrected for the procedural blank contribution (Tables 1a and 1c) this would result in an even older Pb-Pb age for this sample. Secondly, the apparent 'age' difference, discussed above, is far too large to be ascribed to machine bias or interlaboratory bias effects. One point often neglected in this regard concerns U-Pb and Pb-Pb age concordancy. For highly radiogenic samples of this age, blank correction has a dramatic but correlated effect on both μ and the ^{206}Pb/^{204}Pb ratio. Although U-Pb and Pb-Pb concordancy is a necessary condition for obtaining age information, it is not a sufficient condition for believing that the lower relative error on the Pb-Pb age is of this accuracy. Thirdly, although the coincidence in the new Pb-Pb model ages of LEW-Px and ADOR-Px could have arisen by chance, the agreement is quite striking nonetheless.

Initial Lead Isotopic Composition

The extremely high μ of LEW and ADOR allows a quite precise age determination but yields relatively poor information concerning the initial Pb isotopic composition. This is very important to establish, nonetheless, for interpreting these ages with respect to the history of the angrite parent body itself.

The initial ^{207}Pb/^{204}Pb and ^{206}Pb/^{204}Pb ratios of three mineral-fraction pairs calculated using a two-stage model are illustrated in Fig. 3 together with primordial Pb (TATSUMOTO et al., 1973; GÖPEL et al., 1985). These are: ADOR-Px and LEW-Px, LEW-Px and LEW-Plag, and LEW-Px and LEW-PxDk. This calculation assumes that the ^{204}Pb present in the washed minerals is "indigenous" and not a residual contaminant. The errors on the derived initial ratios are around $\pm$5% or less and are quite strongly correlated ($\rho \approx 0.8$). Two-stage model μ_1 values for a range of reasonable ages for the solar system are within error of zero in each case although the errors are, of course, enormous. The pair involving LEW-PxDk falls just below primordial Pb. However, LEW-Plag and LEW-PxDk are slightly discordant (Table 1a); the overall effect of incomplete removal of the terrestrial component will be, in this instance, to marginally decrease the initial Pb isotope ratios (i.e., the "isochron" will be slightly too steep). Because of these slight misgivings, as discussed previously, we consider the highly radiogenic data from LEW-Px and ADOR-Px to produce the most reliable initial Pb isotopic compositions.

With this borne in mind, there is nothing in Fig. 3 to indicate that the initial isotopic composition of the angrites upon crystallization deviated significantly from primordial Pb. This appears to hold for both the internal isotope systematics of LEW and also when LEW and ADOR are considered together. This result shows that the "model" of assuming initial primordial Pb for the angrites in deriving Pb-Pb and U-Pb model ages is a valid one. More importantly, though, it enables us to place quite firm constraints on the time interval between the generation of the high μ (i.e., volatile ele-
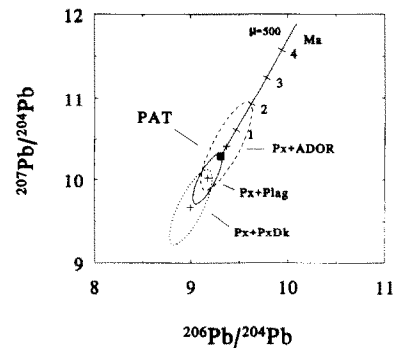

FIG. 3. Initial Pb isotopic compositions of the angrites illustrated in a plot of ^{207}Pb/^{204}Pb vs. ^{206}Pb/^{204}Pb. Three mineral pairs are considered, with the 2σ error ellipses drawn in each case. These are LEW-Px and ADOR-Px (dashed), LEW-Px and LEW-Plag (solid), and LEW-Px and LEW-PxDk (dotted). The initial isotopic compositions were obtained using a two-stage evolution model, with errors calculated by numerical propagation (cf. RODDICK, 1987). The filled square ("PAT") is the primordial Pb isotopic composition, taken from Cañon Diablo troilite (TATSUMOTO et al., 1973). The latter two pairs are expected to yield initial Pb ratios that are slightly too low for reasons discussed in the text. Therefore, the initial isotope ratios of both ADOR and LEW are considered to be indistinguishable within error from primordial Pb. This contrasts with the far more radiogenic initial ratios of many cumulate eucrites (TERA et al., 1989). This result is significant because of the extremely high U/Pb ratios of the angrites—ADOR-Px has a μ ($\equiv$ ^{238}U/^{204}Pb) of $\sim$5600, with the value for whole-rock LEW based upon modal abundances being $\sim$290. The solid line indicates the isotopic evolution calculated for times of 1 to 4 Ma at a μ of 500, which is probably reasonable for the angrite parent body as a whole. It is inferred that the Pb-Pb ages of LEW and ADOR are younger by at most $\sim$2 Ma than the volatile depletion "event" suffered by the parent body or its precursor planetesimals.

ment depleted) character of the parent body and the eventual crystallization of the angrites. The former involves increasing μ from the solar nebula value of $\sim$0.14 (ANDERS and GREVESSE, 1989) to a value of at least several hundred. From Table 1b, ADOR-Px has a μ of $\sim$5600; the corresponding value for whole-rock LEW based upon the published modal abundances of PRINZ et al. (1988) is $\sim$290.

Figure 3 illustrates the isotope ratio deviations from primordial Pb expected after several Ma have elapsed, conservatively assuming a μ of 500 for the silicate portion of the angrite parent body. It is seen that the isotope composition will fall outside the ADOR-LEW pyroxene pair error ellipse for times greater than $\sim$2 Ma. Given that the initial isotope ratios overlap with primordial Pb, this is considered an upper limit.

From this observation, we suggest (1) that the volatile element depletion probably occurred during accretion of the parent body and (2) that LEW and ADOR represent a primary episode of planetary differentiation associated with planet formation. Strictly this is not proven, however. For example, it is possible that the angrite parent body accreted from planetesimals that were already volatile depleted. Nevertheless, if this was the case, accretion of these planetesimals was completed in under 2 Ma. The fact that the two angrites share a common Pb-Pb age is highly suggestive of an "event" of some sort taking place rather than a prolonged "episode."

No matter what exactly transpired, it remains clear that

Table 2.   Rb–Sr isotopic results

Sample	Rb (ppm)	Sr (ppm)	^{87}Rb/^{86}Sr	n	^{87}Sr/^{86}Sr measured †	^{87}Sr/^{86}Sr initial ‡
LEW-Plag	0.0672	299.3	0.000649 (7)	4/9	0.699032 (10)	0.698989 (10)
LEW-WR	0.0704	115.0	0.00177 (3)	2/4	0.699149 (11)	0.699031 (11)
LEW-Ol	0.0071	7.875	0.0026 (3)	1/4	0.699131 (14)	0.698957 (20)
LEW-PxDk	0.0053	46.33	0.00034 (2)	1/3	0.699002 (15)	0.698979 (15)
ADOR # 1	0.0134	142.0	0.00027 (1)	1/4	0.698992 (16)	0.698974 (16)
ADOR # 2	0.0302	139.3	0.00063 (1)	1/3	0.698997 (16)	0.698955 (16)
ADOR # 3	0.0184	126.0	0.00042 (1)	1/2	0.698991 (14)	0.698963 (14)
MC-Plag§	0.081 §	157.6 §	0.00143 (14) §	—	0.699082 (14)	0.698986 (17)

(†)   Normalized to ^{86}Sr/^{88}Sr = 0.1194. Measured ^{87}Sr/^{86}Sr = 0.710263 (5) $\pm 2\sigma_m$
       for NBS–987 Sr (n=18). Errors are external 2σ of the population. n is the
       number of measurements spiked/unspiked aliquots.
(‡)   Assuming an age of 4.56 Ga ($\lambda_{87} = 0.0142$ Ga^{-1}).
(§)   MC - Moore County. Rb, Sr and ^{87}Rb/^{86}Sr data taken from CARLSON and
       LUGMAIR (1988).

the Pb-Pb ages of LEW and ADOR effectively date within ~2 Ma (1) the formation of their parent body and (2) the volatile depletion process, witnessed as a raising of the planetary U/Pb ratio. This is important to establish in connection with the initial ^{87}Sr/^{86}Sr, since it allows an absolute age to be attached to the reduction in Rb/Sr ratio of the parent body. This point will be returned to later on.

INITIAL STRONTIUM ISOTOPE RATIO

Due to the inherently low volatile element content of angrites, Rb/Sr ratios are extremely low, precluding obtaining Rb-Sr ages. However, this does allow determination of precise initial ^{87}Sr/^{86}Sr ratios.

Strontium isotopic compositions and rubidium and strontium concentrations of whole-rock and mineral phases from LEW together with small chips of ADOR and Moore County plagioclase (MC-Pl) are presented in Table 2. Blanks for Sr were <10 pg and negligible in all cases, while Rb blank corrections of 3 pg were applied in some instances. The only data which have to be viewed with caution are those of LEW-WR, as this sample was not washed at all and showed overwhelming effects of MT Pb contamination. As was noted from SEM measurements, the intergrain contaminant contains significant amounts of K and, by inference, Rb. All measured ^{87}Sr/^{86}Sr values for LEW samples are very low and the total spread in ^{87}Rb/^{86}Sr is only a factor ten but at very low values. This minute spread in ^{87}Rb/^{86}Sr between the phases does not allow a reliable primary age to be determined.

On first inspection, both ^{87}Sr/^{86}Sr and ^{87}Rb/^{86}Sr of most LEW samples and ADOR are comparable. The measured ^{87}Sr/^{86}Sr for MC-Pl differs by only <9 ppm from the values previously reported by LUGMAIR et al. (1983) and CARLSON and LUGMAIR (1988). The results in Table 2 show that LEW-Px in particular has extremely low ^{87}Sr/^{86}Sr ratios. To this extent these are in keeping with a low ^{87}Sr/^{86}Sr for ADOR

which attracted considerable attention originally (WASSERBURG et al., 1977). This similarity is already significant, since if LEW had been found to have ^{87}Sr/^{86}Sr resolvably different from ADOR at the very low observed ^{87}Rb/^{86}Sr, this would have precluded derivation of these meteorites from the same parent body.

The measured ^{87}Sr/^{86}Sr and ^{87}Rb/^{86}Sr ratios are plotted in an evolution diagram in Fig. 4 along with a reference 4.56 Ga isochron. This shows that the isotopic results are compatible with a low initial ^{87}Sr/^{86}Sr and only minimal radiogenic growth over this time period. However, the whole-rock sample LEW-WR falls above the reference isochron. As this sample was not washed with HCl prior to dissolution we clearly see the effects on the Rb-Sr system of terrestrial weathering which, as may be recalled, has so affected the Pb isotopes and abundances. Since most of the Sr and Rb is concentrated in phases other than olivine (Table 2), the Rb-Sr balance of LEW-WR should lie in the vicinity of LEW-Plag and LEW-Px on Fig. 4, which it manifestly does not. The clear implication is that the whole rock contains both a Rb excess and an unsupported radiogenic ^{87}Sr component. This is most easily ascribed to terrestrial addition of Rb and Sr. The effect is small in comparison with Pb but is nonetheless significant in terms of the derived initial ^{87}Sr/^{86}Sr ratio (Table 2). Excess Rb leads to overcorrection for radiogenic growth, competed against by radiogenic Sr addition. In this instance the radiogenic Sr addition appears to predominate, resulting in a spuriously high derived initial ^{87}Sr/^{86}Sr ratio. In the case of LEW-Plag, a minute amount of residual contaminant cannot be ruled out as well, as the washing of this separate was done only once.

A detailed comparison of the "washed" Rb-Sr data with the lowest initial ^{87}Sr/^{86}Sr measured for Allende CAIs ('ALL'; GRAY et al., 1973) and the canonical basaltic achondrite initial ('BABI'; PAPANASTASSIOU and WASSERBURG, 1969)

is fraught with difficulties due to interlaboratory machine bias. For this reason, repeat measurements of plagioclase from the eucrite Moore County and the NBS-987 Sr standard were done concurrently. The $^{87}Sr/^{86}Sr$ ratios presented in Table 2 represent the weighted means of multiple runs of unspiked and ^{84}Sr-spiked samples measured on both the La Jolla single- and multiple-collector mass spectrometers; the errors quoted are $\pm2\sigma$ external precisions based upon the measured distributions. Measurement reproducibility from spiked and unspiked samples as well as for the two instruments have been documented previously (LUGMAIR et al., 1989). There is no apparent bias in the Sr isotopic data collected by the various methods noted (Table 2). For comparison with future work, therefore, data are reported relative to a value of 0.710263 $\pm$ 5 ($2\sigma_m$ weighted mean of 18 runs; ±16, 2σ external) for NBS-987 Sr.

Calculation of initial $^{87}Sr/^{86}Sr$ ratios assumed an age of 4.56 Ga (λ_{87} = 0.0142 Ga^{-1}) and are given in Table 2 with 2σ external errors. Based upon these external precisions, the initial $^{87}Sr/^{86}Sr$ ratios from the different meteorites are indistinguishable. The only significant exception, LEW-WR, was discussed above. As was also noted previously, LEW-Plag may be slightly elevated because of limited cleaning of this separate.

From this data we derive a best estimate for the value of the angrite parent body initial $^{87}Sr/^{86}Sr$ of 0.698970 $\pm$ 15 (2σ external). This initial $^{87}Sr/^{86}Sr$ appears at first glance to be quite comparable to that found by other workers for basaltic achondrites (cf. TILTON, 1988, for a review). PAPANASTASSIOU and WASSERBURG (1969) determined an initial of 0.69898 $\pm$ 6 for seven basaltic achondrites (=BABI); BIRCK and ALLÈGRE (1978) found 0.69899 $\pm$ 4 for ten eucrites; and NYQUIST et al. (1986) observed 0.69896 $\pm$ 3 and 0.69894 $\pm$ 2 for matrix and clast from Antarctican eucrite Y75011. However, matters become distinctly more complicated when interlaboratory biases are also considered. For convenience, therefore, we will write deviation in parts per 10^4 from BABI (=0.69898) as ϵ_{Sr}^B.

WASSERBURG et al. (1977) found the initial $^{87}Sr/^{86}Sr$ ratio for ADOR to be distinctly lower than that of BABI. Their preferred value was 0.69884 $\pm$ 2 (ϵ_{Sr}^B = -2.1 $\pm$ 0.3). Using their Moore County plagioclase initial ϵ_{Sr}^B of -1.0 $\pm$ 0.1, then ADOR $^{87}Sr/^{86}Sr$ should be lower than Moore County by ~80 $\pm$ 30 × 10^{-6}. This we cannot confirm based upon our Sr isotopic data in Table 2. We find a difference between Moore County plagioclase and ADOR fragments #1 and #3 of 11 $\pm$ 33 and 24 $\pm$ 31 × 10^{-6} only. At present, the origin of this discrepancy remains unclear. From the data in Table 2, therefore, there does not appear to be a significant difference in initial $^{87}Sr/^{86}Sr$ ratio between the eucrites and the angrites.

NYQUIST et al. (1989) have recently measured the initial $^{87}Sr/^{86}Sr$ ratio of LEW-86010 as part of another consortium study of LEW, and also remeasured ADOR. They found low but different initial Sr ratios in the two angrites. However, they did not report having washed their samples with HCl prior to analysis. The measured $^{87}Rb/^{86}Sr$ ratios ranged from about 0.001 to 0.004 for two plagioclase separates and a whole rock, with $^{87}Rb/^{86}Sr$ in ADOR being ~0.0025. These values are considerably higher than were found in our washed samples. For this reason, we suspect that the $^{87}Sr/^{86}Sr$ initial for

ADOR reported by NYQUIST et al. (1989) of 0.698819 $\pm$ 26 is too low due to overcorrection for ^{87}Rb decay—indeed, their measured $^{87}Sr/^{86}Sr$ appears to agree quite well with our own (Table 2). Their data for LEW may suffer from terrestrial addition of both Sr and Rb during weathering as was noted in connection with our LEW-WR whole-rock sample.

$^{87}Sr/^{86}Sr$ of 0.698970 $\pm$ 15 represents the initial ratio of igneous differentiates from a putative early-formed planetary body. The lowest initial $^{87}Sr/^{86}Sr$ of all (sic) found in CAIs from Allende ('ALL'; GRAY et al., 1973) is often considered as the canonical "solar system initial" for Sr isotopes. However, we feel that the above value for ADOR, LEW, and Moore County may, in fact, be more representative for a variety of reasons:

1) CAIs are first condensates and/or have a possible presolar history, which may make them unrepresentative.
2) CAIs may record Sr isotopic compositions during stages in solar nebular condensation at varying Rb/Sr ratio, on an uncertain timescale.
3) Since the solar nebular itself was not a closed system, decay of ^{87}Rb relative to ^{86}Sr as recorded in CAIs during condensation may be poorly represented by estimates of Rb/Sr derived from chondrites.
4) The data for ADOR and LEW presented above precisely constrain the initial $^{87}Sr/^{86}Sr$ and the age at which this was applicable for angrites (4.5578 $\pm$ 4 Ga).

NEODYMIUM ISOTOPES

Sm-Nd data for whole-rock and mineral separates from LEW are presented in Tables 3 and 4. Table 3 contains data relevant to the ^{147}Sm-^{143}Nd system and Table 4 pertains to ^{146}Sm-^{142}Nd. Neodymium was measured as NdO$^+$, and samarium as the metal ion. No mass spectrometric (isobaric) interferences of any kind were observed, and thus no corrections were applied to the data other than those for mass fractionation and oxygen isotope contributions. Individual sample loads for Nd were always less than 15 ng and sometimes as small as 500 pg. In addition, blanks were negligible in all instances and no corrections were necessary (see GOODRICH et al., 1991, for typical blanks).

^{147}Sm-^{143}Nd System

The number of mineral phases and their relative Sm-Nd distribution make LEW quite a favourable sample for Sm-Nd dating. We obtained high-quality data on the whole-rock sample and on mineral separates of plagioclase, light and dark pyroxene, and olivine. $^{147}Sm/^{144}Nd$ varies from 0.1375 in plagioclase to 0.3589 in olivine with the other values fairly uniformly distributed within this range. The resulting ^{147}Sm-^{143}Nd isochron (Fig. 5) has a slope of 0.03022 $\pm$ 23 yielding an age of 4.553 $\pm$ 34 Ga with an initial $^{143}Nd/^{144}Nd$ of 0.506682 $\pm$ 49 (2σ error; λ_{147} = 0.00654 Ga^{-1}).

As the insert in Fig. 5 illustrates, all the data points fall on or close to the isochron within ±0.5 ϵ units. The only point whose error in $^{143}Nd/^{144}Nd$ does not overlap the best fit line is that of PxLt. However, this small deviation is most likely not due to a natural phenomenon or a systematic error on $^{143}Nd/^{144}Nd$. Rather, it is a consequence of the observed

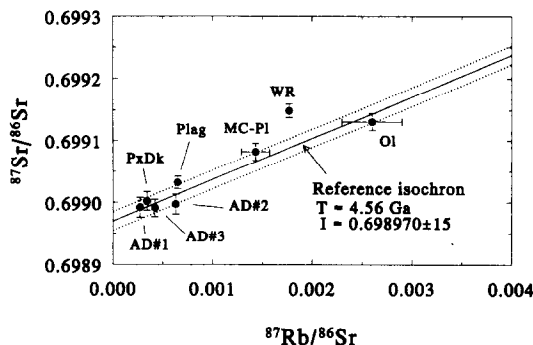FIG. 4. Initial $^{87}Sr/^{86}Sr$ of the angrites illustrated by means of a plot of $^{87}Sr/^{86}Sr$ vs. $^{87}Rb/^{86}Sr$ for LEW and ADOR. Data are shown for washed mineral separates of LEW and unwashed LEW-WR. The three samples AD#1, AD#2, and AD#3 are separate dissolutions of small fragments of ADOR (Table 2). Plagioclase from the cumulate eucrite Moore County (MC-Pl) was measured concurrently to act as an interlaboratory Sr standard. The data encompass too small a range in $^{87}Rb/^{86}Sr$ ratio (<0.003) to derive a useful age. A reference isochron has been drawn for 4.56 Ga (λ_{87} = 0.0142 Ga^{-1}) with an initial $^{87}Sr/^{86}Sr$ = 0.698970. The unwashed whole-rock LEW-WR lies above the reference isochron. Since the whole rock should actually lie in the vicinity of the LEW-Plag and LEW-PxDk points, this clearly indicates addition of both Rb and radiogenic Sr for this sample. The remaining data points for ADOR, LEW, and Moore County are quite consistent with a low initial $^{87}Sr/^{86}Sr$ of 0.698970 ± 15 (2σ) for all three meteorites followed by a very small amount of radiogenic growth since isotopic closure. Based upon the extremely high Rb/Sr ratio of the solar nebula (ANDERS and GREVESSE, 1989), this suggests that the angrite and eucrite parent bodies were formed within about ±2 Ma of each other.

$^{147}Sm/^{144}Nd$ ratio. As shown in Table 3, the uncertainty of this ratio for PxLt is rather large (0.52%) owing to the small sample split available for concentration determination. But the deviation of $^{147}Sm/^{144}Nd$ from the isochron is only +0.32% and thus easily within error. The values for the whole-rock sample $^{147}Sm/^{144}Nd$ and $^{143}Nd/^{144}Nd$ also fall very close to the CHUR data and are well within the range of values

defining CHUR in the first place (JACOBSEN and WASSER-BURG, 1984). Thus, no fractionated source relative to chondritic Sm/Nd is indicated for LEW.

The Sm-Nd age obtained for LEW of 4.553 ± 34 Ga agrees with the Sm-Nd age of 4.56 ± 0.10 Ga recently reported by NYQUIST et al. (1991) for this meteorite. It also compares very satisfactorily with the ^{147}Sm-^{143}Nd isochrons determined previously for ADOR. LUGMAIR and MARTI (1977) obtained an age of 4.55 ± 4 Ga with an initial $^{143}Nd/^{144}Nd$ of 0.50682 ± 5, whereas JACOBSEN and WASSERBURG (1984) found 4.564 ± 37 Ga. The initial $^{143}Nd/^{144}Nd$ for ADOR determined by JACOBSEN and WASSERBURG (1984) was 0.506678 ± 44 (normalized to $^{148}Nd/^{144}Nd$ = 0.241572; cf. TILTON, 1988). The slightly elevated initial reported by LUGMAIR and MARTI (1977) for ADOR stemmed from an instrumental artifact and was due to an underestimated time constant of the electrometer amplifier. Nonetheless, agreement is seen to be very good for both the ages and initial ratios of ADOR and LEW and, in this case, between two different laboratories.

Samarium-147 decay constant

The weighted mean of the three precise ^{147}Sm-^{143}Nd age determinations on ADOR and LEW is 4.554 ± 16 Ga (2σ), which includes the Student's t-factor. Based upon the more precise Pb-Pb ages for LEW and ADOR reported in Table 1 (4.5578 ± 4 Ga), the ^{147}Sm-^{143}Nd angrite ages are at best only marginally lower by −0.08 ± 0.36%. Thus, they are in agreement at the <1σ level. However, as the decay constants of ^{238}U and ^{235}U are known more precisely than that of ^{147}Sm, this gives us the opportunity of obtaining a more precise (relative) number for the latter. For example, to give a more consistent Sm-Nd age for the angrites would require decreasing λ_{147} to a value of 6.535 ± 0.024 × 10^{-3} Ga^{-1}. This is well within error of the value originally derived from a literature review by LUGMAIR and MARTI (1978) of 6.54 ± 0.10 × 10^{-3} Ga^{-1}. Since there is so little difference, it is not recommended that this new value be adopted.

Table 3. LEW-86010 ^{147}Sm–^{143}Nd isotopic results

Sample	Sm (ppm)	Nd (ppm)	$^{147}Sm/^{144}Nd$	$^{143}Nd/^{144}Nd$	$\epsilon_{Nd}^{143}(0)$
LEW-WR	2.896 (2)	8.846 (3)	0.1979 (1)	0.512664 (14)	+0.51 (0.27)
LEW-Plag	0.04878 (7)	0.2144 (10)	0.1375 (6)	0.510829 (36)	−35.29 (0.70)
LEW-Ol	0.06384 (6)	0.1075 (1)	0.3589 (4)	0.517527 (36)	+95.37 (0.70)
LEW-PxLt	3.845 (20)	10.13 (1)	0.2297 (12)	0.513602 (14)	+18.80 (0.28)
LEW-PxDk	6.185 (2)	17.182 (5)	0.2177 (1)	0.513265 (14)	+12.21 (0.27)
CHUR†	—	—	0.1966	0.512638	≡ 0

Nd isotopic data normalized to $^{148}Nd/^{144}Nd$=0.241572 (equiv. to $^{146}Nd/^{144}Nd$=0.7219). $\epsilon_{Nd}^{143}(0)$ represents the deviation in parts per 10^4 from the CHUR $^{143}Nd/^{144}Nd$ ratio today. (†) JACOBSEN and WASSERBURG (1984).

Table 4. LEW-86010 ^{146}Sm–^{142}Nd isotopic results

Sample	^{144}Sm/^{144}Nd	^{142}Nd/^{144}Nd	ϵ_{Nd}^{142} (0)
LEW-WR	0.04057 (3)	1.141834 (34)	0.00 (0.30)
LEW-Plag	0.02819 (12)	1.141735 (30)	−0.87 (0.26)
LEW-Ol	0.07357 (9)	1.142042 (94)	+1.82 (0.82)
LEW-PxLt	0.04709 (24)	1.141874 (32)	+0.35 (0.28)
LEW-PxDk	0.04462 (2)	1.141859 (40)	+0.22 (0.35)
La Jolla Nd	—	1.141834 (11)	≡ 0
CHUR†	0.04030	—	

Nd isotopic normalization factors are given in Table 3. ϵ_{Nd}^{142} (0) represents the deviation in parts per 10^4 from the terrestrial standard ^{142}Nd/^{144}Nd ratio today. (†) JACOBSEN and WASSERBURG (1984).

NUNES (1981) previously found no discrepancy at the level of ~0.8% based upon a comparison of Sm-Nd and U-Pb zircon ages from the Stillwater complex. He suggested, in fact, that λ_{147} might be reduced by −0.25 ± 0.32% (using La Jolla Nd normalization) to be consistent with the U decay constants. The above findings, however, suggest the agreement to be somewhat better.

^{146}Sm-^{142}Nd System

The ^{146}Sm-^{142}Nd systematics of LEW are given in Table 4 and illustrated in Fig. 6. The data points are plotted in terms of relative deviations (ϵ_{Nd}^{142}) of ^{142}Nd/^{144}Nd from the terrestrial normal, which is taken as the present-day solar system value and whose uncertainty is indicated by the horizontal dotted lines in Fig. 6. Because of the large spread in Sm/Nd ratios, excesses of ^{142}Nd from now-extinct ^{146}Sm (λ_{146} = 6.73 Ga^{-1}) are clearly resolved and a good isochron was obtained (Fig. 6). This yielded a slope of 0.0071 ± 17, corresponding to the ^{146}Sm/^{144}Sm ratio at the time of isotopic closure; the derived initial ^{142}Nd/^{144}Nd ratio of 1.141540 ± 71 is equivalent to ϵ_{Nd}^{142} of −2.57 ± 0.62. As noted above for the ^{147}Sm-^{143}Nd system, the whole-rock data are consistent with derivation of LEW from a chondritic Sm/Nd reservoir.

These results for LEW compare reasonably well with the original work of LUGMAIR and MARTI (1977) on ADOR, who found an initial ^{146}Sm/^{144}Sm of 0.0047 ± 23 (with ϵ_{Nd}^{142} = − 1.4 ± 0.7). However, both this ADOR value and that of LEW reported here are clearly inconsistent with the value 0.0118 ± 32 obtained by JACOBSEN and WASSERBURG (1984) for ADOR.

Large excesses of ^{143}Nd (up to 36%) and ^{142}Nd (up to 0.47%) have been found by LUGMAIR et al. (1983) in a carbon-chromite fraction from Allende which they interpreted to result from α-recoil of the decay products of parent nuclides ^{147}Sm and ^{146}Sm followed by capture in a carbon coating. In order to test this recoil model SHIMAMURA and LUGMAIR (1984) measured ^{234}U in the same samples. They observed

large excesses of ^{234}U from the decay of ^{238}U with a relative distribution among the etch fractions entirely consistent with the ^{142,143}Nd excesses, thus proving that the recoil-mechanism is responsible for the large enrichments of these radiogenic nuclides. While the effects on ^{142}Nd in the original ADOR data were only very small, these later, much larger excesses demonstrated beyond any doubt the existence of live ^{146}Sm in the early solar system. The calculated values for ^{146}Sm/^{144}Sm resulting from the three dissolution steps ranged from 0.0035 ± 1 to 0.0063 ± 5 with an average of 0.0045 ± 5. These data compare favourably with the value found previously for ADOR. Unfortunately, however, no precise time can be assigned to the formation of these putative assemblages of carbon-coated silicate grains, although it is unlikely to be much different from 4.56 Ga ago. Additionally, the geometry of these assemblages is not known; neither is the variation of Sm content of the silicate grains nor the potential artifact caused by the small differences in recoil energies/ranges of ^{142}Nd and ^{143}Nd nuclei and the etching procedures. Thus, calculation of a simple average of the available data appeared to be the most conservative approach, though it now seems that the higher value of 0.0063 ± 5 from CG-1 is closest to the value obtained from LEW.

Recent work by several laboratories on various meteorite types has only fueled the controversy as to the initial ^{146}Sm/^{144}Sm ratio of the solar system (conventionally taken to have formed at 4.56 Ga). This is unfortunate since ^{146}Sm is an important nuclide which potentially could serve as a short-term chronometer for early solar system processes and allow the mean age of the p- or γ-process in the galaxy to be derived (e.g., AUDOUZE and SCHRAMM, 1972; LUGMAIR et al., 1975). PRINZHOFER et al. (1989) presented data from two meteorites with ^{147}Sm-^{143}Nd ages of 4.46 and 4.47 Ga (Ibitira and Morristown). The measured respective ^{146}Sm/^{144}Sm ratios were 0.009 ± 1 and 0.0075 ± 11 with ϵ_{Nd}^{142} = −2.8 ± 0.4 and −2.1
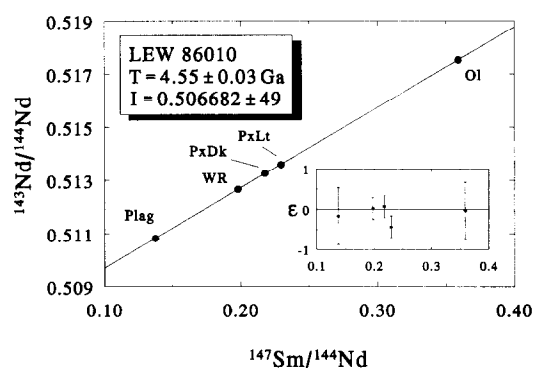

FIG. 5. ^{147}Sm-^{143}Nd evolution diagram for LEW. The measurement errors are smaller than the symbols plotted. The isochron corresponds to an age of 4.553 ± 34 Ga with an initial ^{143}Nd/^{144}Nd ratio of 0.506682 ± 49 (2σ; λ_{147} = 0.00654 Ga^{-1}). The deviations from the best-fit line in ε-units are shown in the inset figure. Although LEW-PxLt appears to lie off the isochron, this is not actually the case. The error in ^{147}Sm/^{144}Nd for LEW-PxLt is comparatively large (0.52%) due to the small size for this separate, while the deviation from the isochron is smaller at +0.32%. The ^{147}Sm/^{144}Nd and ^{143}Nd/^{144}Nd ratios of LEW-WR (Table 3) are within error of CHUR (JACOBSEN and WASSERBURG, 1984), indicating that LEW was derived from a source with an unfractionated Sm/Nd ratio with respect to chondrites.
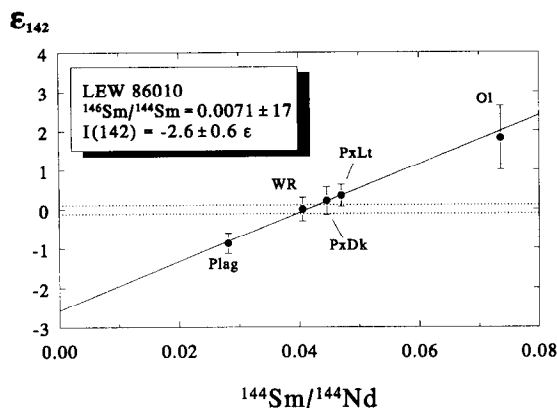
ε_{142}

FIG. 6. ^{146}Sm-^{142}Nd evolution diagram for LEW, plotted as ε_{142} vs. ^{144}Sm/^{144}Nd ratio. ε_{142} is the deviation in parts per 10^4 from the mean of terrestrial standards measured during this study. The dotted lines show the range of uncertainty (2σ) in the standard values of ± 0.10 ε-unit (Table 4). The best-fit line through the data implies an initial ^{146}Sm/^{144}Sm = 0.0071 $\pm$ 17 with an initial ε_{Nd}^{142} = -2.57 ± 0.62. For a chondritic ^{144}Sm/^{144}Nd of 0.04030 (Table 4), there is no significant deviation of the isochron in ε_{142} from the terrestrial normal. This implies, as noted in Fig. 5 for the ^{147}Sm-^{143}Nd system, that LEW is a product of a source with chondritic Sm/Nd ratio.

$\pm$ 0.5. These data, disregarding the isochron ages, are actually in very good agreement with those reported here for LEW. However, when the measured ^{146}Sm/^{144}Sm ratios are corrected for the decay of ^{146}Sm back to 4.56 Ga they become 0.015, in broad agreement with those of JACOBSEN and WASSERBURG (1984), but a factor of two to three higher than indicated by the present data and those of LUGMAIR and MARTI (1977) and LUGMAIR et al. (1983). Because ADOR and LEW have essentially identical Pb-Pb ages which correspond so closely to 4.56 Ga implies (1) that they should have initial ^{146}Sm/^{144}Sm ratios within error, and (2) that these initials should equal the solar system initial ^{146}Sm/^{144}Sm ratio and require no significant corrective term. In this regard, the agreement between the initial ^{146}Sm/^{144}Sm ratios for LEW derived in this study (0.0071 $\pm$ 17) and the earlier work on ADOR (0.0047 $\pm$ 23) by LUGMAIR and MARTI (1977) is quite satisfactory. Moreover, at least for the case of these results it strengthens the view that there were actually no ". . . experimental difficulties for some of the data," as has been suggested by PRINZHOFER et al. (1989). The results obtained in this laboratory are quite consistent and imply a solar system initial ^{146}Sm/^{144}Sm ratio near 0.006 rather than any values higher than 0.01.

Among other recently reported ^{146}Sm/^{144}Sm data are those of a eucrite clast from Bholghati (NYQUIST et al., 1990), for this meteorite, LEW 86010 (NYQUIST et al., 1991), and the unusual achondrite Acapulco (PRINZHOFER et al., 1990). Although the Bholghati clast shows some signs of disturbance in the ^{147}Sm-^{143}Nd system, an age of 4.510 $\pm$ 34 Ga was obtained for the more resilient and colinear data points. From their ^{142}Nd/^{144}Nd data the authors derived a ^{146}Sm/^{144}Sm = 0.0043$^{+39}_{-26}$ for a time 4.56 Ga ago. The data obtained for LEW by NYQUIST et al. (1991) are 4.56 $\pm$ 0.10 Ga for the Sm-Nd crystallization age and ^{146}Sm/^{144}Sm = 0.0078 $\pm$ 20 with ε_{Nd}^{142} = -2.5 ± 0.8. Acapulco exhibits a well-behaved

^{147}Sm-^{143}Nd isochron yielding an age of 4.60 $\pm$ 0.03 Ga. This age appears somewhat high considering firstly, the small uncertainty attached, and secondly, the initial ^{143}Nd/^{144}Nd which is consistent with a chondritic Sm/Nd origin. The initial ^{146}Sm/^{144}Sm reported for Acapulco is 0.0067 $\pm$ 19 with ε_{Nd}^{142} = -2.11 ± 0.76. Nonetheless, all these ^{146}Sm/^{144}Sm values are in good agreement with those reported here, if some of the aberrant data points from the Bholghati clast results and from LEW (NYQUIST et al., 1991) are omitted from the data regressions. These additional data for ^{146}Sm/^{144}Sm certainly serve to strengthen the case for a value around 0.006. But how can one reconcile this number with 0.015 of PRINZ-HOFER et al. (1989) or 0.0118 of JACOBSEN and WASSERBURG (1984)?

In a recent paper, PRINZHOFER et al. (1992) presented a quite plausible model which explains the apparent discrepancy between the young isochron ages of Ibitira and Morristown on the one hand and the actually measured values for ^{146}Sm/^{144}Sm and ε_{Nd}^{142} on the other. The latter, as was noted above, agree rather well with the data obtained for LEW and other samples. This model involves partial chemical re-equilibration of the rare earths between phosphate and plagioclase at times significantly younger than 4.46 Ga; the phosphates are left essentially unaffected while the pyroxene is totally unaffected. This scheme would account for a partial resetting of the ^{147}Sm-^{143}Nd isochrons, whereas the ^{146}Sm-^{142}Nd systems would not be resolvably affected. As a result, the true solidification ages of these meteorites may be close to 4.56 Ga. This then would resolve the conflict which had existed for some time and was noted earlier. The data for ADOR of JACOBSEN and WASSERBURG (1984), however, remain problematic.

We now can combine all the available data and consider their relative uncertainties, calculated and implicit, to formulate best estimates for the ^{146}Sm-^{142}Nd system parameters. The suggested values, at 4.56 Ga, are ^{146}Sm/^{144}Sm = 0.0070 $\pm$ 10 and ε_{Nd}^{142} = -2.5 ± 0.5 with a half-life $T_{146}^{1/2}$ = 103 Ma.

With this reasonably precise set of parameters established, it appears feasible to apply the ^{146}Sm-^{142}Nd system as an early solar system chronometer. Its usefulness is very much enhanced by the long-lived ^{147}Sm-^{143}Nd chronometer which, together, may permit further insights into the evolution of even chemically slightly disturbed samples in a manner such as pointed out by PRINZHOFER et al. (1992). As an absolute chronometer this system is now bound securely to the precise Pb-Pb ages of LEW and ADOR, as discussed earlier. However, the time resolution afforded through present-day measuring capabilities will not exceed $\sim$15 Ma in most instances.

In closing it may be worth noting that ^{146}Sm/^{144}Sm = 0.007 is entirely consistent with estimates of the p-process production ratios $P_{146/144}$ of AUDOUZE and SCHRAMM (1972) and LUGMAIR et al. (1975) if continuous synthesis and a formation interval of $\sim$10^7 years are assumed. However, $P_{146/144}$ as calculated by WOOSLEY and HOWARD (1978) for production by photodisintegration is too low by more than an order of magnitude. In a recent re-evaluation by these authors (WOOSLEY and HOWARD, 1990), considering revised (γ, n) rates for ^{148}Gd and ^{150}Gd, the calculated ^{146}Sm/^{144}Sm production ratios now range from $\sim$0.01 to 0.4. This brings them into much closer agreement with the observed values.

The effects on nucleochronological models of using the lower $^{146}Sm/^{144}Sm$ ratio of 0.007 are broadly to increase the mean age of galactic p- or γ-process nucleosynthesis, or to require a longer time interval between the cessation of p- or γ-process and the formation of the solar system (cf. LUGMAIR et al., 1975; PRINZHOFER et al., 1989).

OTHER ISOTOPIC STUDIES

Due to the extremely old age of the angrites, there is the possibility that evidence for ^{26}Al (half-life $T^{1/2}_{26} = 0.72$ Ma) might be found, as in CAIs (see WASSERBURG, 1985, and PODOSEK and SWINDLE, 1988, for a review). Indeed, if the mean Pb-Pb age of CAIs of 4.559 ± 4 Ga is correct, as reviewed by TILTON (1988), and if ^{26}Al was "live" and uniformly distributed in the early solar nebula, one should expect ^{26}Mg excesses in high Al/Mg phases in LEW given that its Pb-Pb age is 4.5578 ± 5 Ga. A previous search by SCHRAMM et al. (1970) failed to find such evidence among several meteorite classes. However, this rested largely upon analyses of (1) eucrites such as Moore County, and (2) ordinary chondrites. Moore County is now known to be far too young (Sm-Nd age 4.457 ± 25 Ga; TERA et al., 1987b) to manifest any effect due to ^{26}Al. The same problem may also be true for ordinary chondrites, many of which experienced metamorphism at times much less than 4.56 Ga (GÖPEL et al., 1989). ADOR is also not particularly useful in this regard as it consists mostly of fassaitic pyroxene with a comparatively low Al/Mg ratio. Fortunately, LEW contains plagioclase with a sufficiently high Al/Mg ratio to address this question.

Several measurements of the Mg isotopic composition were carried out on LEW-Plag and LEW-Ol with Al/Mg of ~1100 and ~0.002, respectively (CROZAZ and McKAY, 1990). No significant deviations of $^{26}Mg/^{24}Mg$ from normal were found. An upper limit of ~2 × 10^{-7} on $^{26}Al/^{27}Al$ is indicated for the time when LEW crystallized.

PRINZ et al. (1988) put forward a model by which the starting material of the angrites was a mixture of type B CAIs and the opaque matrix component of chondrites. If this were the case, then the possibility could not be excluded that resolvable traces of typical isotope anomalies as found in basically all CAIs, such as on ^{48}Ca or ^{50}Ti, could have survived in LEW. In particular, anomalies in ^{50}Ti, which are normally excesses of ~9 ε-units, would be resolvable even for a threefold dilution with isotopically normal solar system material. However, as discussed by LUGMAIR et al. (1989), all results for ^{48}Ca and ^{50}Ti obtained from various mineral fractions of LEW are normal within error. These measurements do certainly not represent conclusive tests for models such as that of PRINZ et al. (1988). Nevertheless, these negative results would suggest that any direct involvement of original CAI material (as we know it from Allende and other meteorites) in the formation of the angrite source is unlikely. We also note that CAI-type material is characterized by extremely high U/Pb ratios, allowing them to be dated radiometrically (e.g., CHEN and TILTON, 1976; CHEN and WASSERBURG, 1981). One would therefore expect to see significant deviations from primordial Pb in the initial isotopic compositions of LEW and ADOR if much CAI material was present in

their source magmas, even after only a few Ma of isotopic growth. Needless to say, this is not evident in the inferred initial Pb isotopic ratios shown in Fig. 3.

The Sm isotopic composition in LEW-PxDk was also measured. All isotopic ratios, except $^{150}Sm/^{149}Sm$, were found to be normal within error. $^{150}Sm/^{149}Sm$ shows an excess of 3.2 ± 0.6 ε-units and is taken to be due to the absorption by ^{149}Sm of cosmic-ray-produced secondary neutrons for 17.6 ± 1.0 Ma, the cosmic-ray exposure age determined by EUGSTER et al. (1991). Using the neutron cross section of σ_{149} = 5.66 × 10^4 barn from LUGMAIR and MARTI (1977), a neutron fluence of (2.0 ± 0.4) × 10^{15} n cm^{-2} or a flux of 3.6 ± 0.7 n cm^{-2}sec^{-1} are calculated. This value may be compared to 2.2 ± 0.5 n cm^{-2}sec^{-1} obtained for ADOR. The higher neutron flux of our sample of LEW either indicates a larger shielding depth or a larger pre-atmospheric size than for ADOR. This is just the opposite of what was concluded by EUGSTER et al. (1991) and HOHENBERG et al. (1991) from their $^{131}Xe/^{126}Xe$ data on LEW. To explain this conflict one is forced to resort to complicated scenarios, such as outlined by HOHENBERG et al. (1991). Because of lack of any further constraints these will not be reiterated here.

DISCUSSION

There are a considerable number of interesting aspects to emerge from the results presented above. These can be roughly divided into (1) the specific implications for the genesis of the angrites, the angrite parent body, and the nature of the precursor materials, and (2) more generalized conclusions regarding the timescale of solar nebula condensation. It may appear premature to make specific generalizations based upon just two meteorites of a class whose petrogenesis and relationship to other meteorite classes is far from being well understood. Nevertheless, the results and circumstance warrant some sort of appraisal.

Angrite Relationships

One of the principal aims of this study was to determine the exact relationship that LEW-86010 has to Angra dos Reis. Of course, it is only possible here to remark on whether the chemical similarities of the two meteorites also extend to their ages and other isotope systematics. In recent geochemical work on the petrogenesis of these two angrites it has been concluded that the two are chemically similar, but a comagmatic origin is precluded (CROZAZ and McKAY, 1990; MITTLEFEHLDT and LINDSTROM, 1990). However, the recently found LEW-87051 angrite may be petrogenetically related to LEW-86010 (MITTLEFEHLDT and LINDSTROM, 1990). JUREWICZ et al. (1991) have additionally shown that eucrites and angrites may have a common source, of carbonaceous chondrite type, where the controlling factor governing magma composition is the oxygen fugacity during melting. Since LEW and ADOR cannot be derived from the same magma batch, the question then becomes whether they come from the same or different parent bodies. This can be addressed, albeit not definitively, using the chemical history recorded by the isotope systematics, provided that this history is sufficiently unique.

In our original preliminary reports (LUGMAIR et al., 1989;

LUGMAIR and GALER, 1989), we concluded that consanguinity of LEW and ADOR could not be ruled out based upon the available data. With the aquisition of the further data reported here it is clear that LEW and ADOR possess quite remarkable chronological and isotopic similarities: (1) Pb-Pb age and initial isotopic composition, (2) initial $^{87}Sr/^{86}Sr$ ratio, (3) ^{147}Sm-^{143}Nd age and initial ratio, and (4) ^{146}Sm-^{142}Nd isotope systematics. Additionally, the oxygen isotopic data reported by CLAYTON and MAYEDA (1989) supports derivation of LEW and ADOR from a common oxygen reservoir similar to that of the eucrites (CLAYTON and MAYEDA, 1983). When taken together, these isotopic observations imply production of LEW and ADOR from similar sources which apparently experienced volatile element loss and igneous activity at similar times. Although this circumstance could have arisen on separate parental asteroids, it is highly improbable that both bodies could have experienced the same forms of chemical processing synchronously. In contrast to MITTLEFEHLDT and LINDSTROM (1990) we therefore suggest a common parent body for the angrites as more reasonable.

Condensation and Chronology of the Solar Nebula

Pb-Pb ages of angrites and CAIs

It may at first glance not seem particularly important that the Pb-Pb age of ADOR has been refined from 4.551 ± 4 Ga (CHEN and WASSERBURG, 1981) to 4.5578 ± 4 Ga—an increase of a mere 7 Ma. However, this age approaches so closely the accepted "age" of the solar system at around 4.56 Ga that it appears to offer quite a hard constraint on models of solar nebula condensation. For example, dating of Allende CAIs by the Pb-Pb method (TATSUMOTO et al., 1976; CHEN and TILTON, 1976; CHEN and WASSERBURG, 1981; ARDEN and CRESSEY, 1984) has recently been reviewed by TILTON (1988). Based mainly upon criteria of concordance, TILTON (1988) adopted 4.559 ± 4 Ga as the best estimate for the Pb-Pb ages of CAIs. As can readily be appreciated, this CAI age ostensibly overlaps ($<1\sigma$ level) with the ADOR and LEW ages. Taken at face value, this would indicate that once condensation of the solar nebula was initiated (i.e., CAIs appeared), planetary-sized bodies were formed and had differentiated to form igneous rocks like the angrites within at most a couple of million years. This conclusion will not change unless the Pb-Pb ages of CAIs can be pushed further back in time.

In this connection, the data from ARDEN and CRESSEY (1984) imply slightly older Pb-Pb model ages for Allende chondrules and inclusions of 4.567 ± 4 Ga; these ages were discordant, however. MANHÈS et al. (1987) determined internal Pb-Pb isochrons on Allende inclusions, reporting that the ages fell between 4.565 and 4.575 Ga. More recent estimates from the Paris group place the Allende inclusions at 4.566 ± 2 Ga (GÖPEL et al., 1991). So far the results of these studies have only been presented in abstract form, however. Recently, IRELAND et al. (1990) obtained U-Pb ages on tiny perovskite inclusions within CAIs from Allende and the Murchison CM2 carbonaceous chondrite by ion microprobe. Data for three such inclusions yielded concordant ages with large 2σ errors—4.565 ± 34 (1 Allende grain) and 4.569 ± 26 Ga (2 grains from Murchison). Collectively, these data

indicate that perhaps a better Pb age of CAIs is a few Ma older ($\sim$4.565 Ga) than suggested by TILTON (1988). Much of the CAI Pb work has been done on bulk CAIs which were not washed before dissolution. It has already been noted that even a small amount of terrestrial Pb addition will serve to lower the apparent Pb-Pb model ages, ADOR being a case in point.

Another issue that must be addressed at this juncture concerns ^{247}Cm, which is r-process produced and α-decays to ^{235}U with a half-life of 16 Ma. This nuclide will significantly alter both the Pb-Pb and ^{235}U-^{207}Pb model ages if two conditions are met: (1) that it was "live" in sufficient abundance relative to ^{235}U, and (2) some Cm-U fractionation occurred. (The ^{238}U-^{206}Pb model age is, of course, unaffected.) Moreover, because the angrite and CAI Pb-Pb ages in general differ by less than one half-life of ^{247}Cm, effects might conceivably occur in both angrites and CAIs (assuming homogeneous distribution of ^{247}Cm). The sense of the Pb-Pb model age shift depends upon the sense of any Cm-U fractionation: The Cm-rich phase will have an apparent Pb-Pb age that is too high. Fortunately, the combination of both effects (1) and (2) above can be directly assessed by measurement of the $^{238}U/^{235}U$ ratio today (cf. SHIMAMURA and LUGMAIR, 1981; CHEN and WASSERBURG, 1981). The results of these studies on CAIs and ADOR, together with those for LEW from this study (discussed earlier) demonstrate that CAIs and the two angrites have a normal $^{238}U/^{235}U$ ratio within about $\pm0.3\%$— that is, no excesses or deficits due to ^{247}Cm can be resolved. For a $\sim$4.56 Ga radiogenic sample, this uncertainty corresponds to a Pb-Pb age shift of around ±4.4 Ma, or $\sim$0.10% in the absolute age. Such a shift is permitted by the present precision of U isotopic determination but appears a remote possibility. However, the important point is that ^{247}Cm is, at worst, only likely to have a marginal effect on the Pb-Pb chronology of CAIs relative to angrites in the early solar system.

Recently, models of solar nebula evolution and condensation have been reviewed by WOOD and MORFILL (1988) and by WEIDENSCHILLING (1988). Although gaps in understanding still exist, the prevailing viewpoint seems to be that the time between proto-nebula collapse and the formation of asteroidal-sized planetesimals is <1 Ma. The time taken to fully accrete the terrestrial planets from such materials would appear to be somewhat longer, however (WETHERILL, 1986, 1990). This former timescale appears to be quite compatible with the Pb data for CAIs and those presented for the angrites ADOR and LEW above; using an older age for CAIs implies <10 Ma, and with the younger CAI age recommended by TILTON (1988) this becomes 1 ± 4 Ma. Although there appears to be some vestige of agreement here, it is argued in what follows that this is, in fact, decidedly not the case.

Aluminium-26

The absence of evidence for "live" ^{26}Al (half-life $T_{1/2}^{26}$ = 0.72 Ma) in LEW is quite important in this regard, for it would tend to support the slightly older CAI age. That is, if any "live" ^{26}Al existed in CAIs at the "canonical" initial $^{26}Al/^{27}Al$ ratio of $\sim5 \times 10^{-5}$ (cf. WASSERBURG, 1985) and was uniformly distributed throughout the solar nebula—which is not at all clear—it would have decayed below detection by

the time LEW had crystallized. If this was indeed the case and using this lower bound on ^{26}Al, some CAIs are >5.7 Ma older than LEW; based upon the Pb-Pb age this would imply ages > (4.5635 ± 5) Ga.

Regrettably, for this very reason this ambiguity also fails to resolve the objections by Clayton (e.g., CLAYTON, 1975) that ^{26}Al was not "live" but rather is a pre-solar remnant in CAIs. Similarly, it has still not been ascertained whether ^{26}Al distribution, like ^{16}O, was inhomogeneous in the solar nebula (cf. WASSERBURG, 1985; PODOSEK and SWINDLE, 1988). Both of these matters are likely to be settled once high-precision Pb ages and Mg isotopes become available on the same refractory inclusions. Yet another possibility is that ^{26}Al was produced *in situ* within the solar system by intense proton bombardment from the early active Sun, as discussed by WASSERBURG (1985). Presently, the only firm evidence for live ^{26}Al in the early solar system comes from a basaltic, and therefore igneously processed, chondrule in the unequilibrated LL3.0 ordinary chondrite Semarkona (HUTCHEON and HUTCHISON, 1988). Meanwhile, interstellar grains preserved in Murchison often have extreme enrichments in "fossil" pre-solar ^{26}Al (ZINNER et al., 1991).

Despite these misgivings it is possible to settle the question of whether ^{26}Al was an important heat source for moderately sized bodies in the early solar system, as has often been mooted. The precise and extremely old Pb-Pb age of LEW permits more stringent limits to be placed than in the study of SCHRAMM et al. (1970). It is clear that ^{26}Al decay did not supply significant amounts of heat for the following:

1) Differentiation of parent bodies to form the basaltic achondrites. It has been shown that LEW and ADOR were very early magmatic differentiates of the angrite parent body and LEW shows no evidence for live ^{26}Al. Because most eucrites have radiometric ages younger than the angrites but share the same initial Sr isotope composition (Table 2), this should also be true for eucrites.

2) Metamorphism of the chondrite parent bodies. GÖPEL et al. (1989, 1991) have recently reported U-Pb ages of phosphates in equilibrated ordinary chondrites consistently younger than the angrite crystallization ages (~4.551 Ga). However, if these just reflect cooling ages then ^{26}Al may be a viable heat source for the thermal metamorphism (cf. GÖPEL et al., 1991).

3) Aqueous alteration of the CI chondrites. Vein formation appears to have happened too late based upon Sr isotope studies (MACDOUGALL et al., 1984) for ^{26}Al to be responsible.

4) Heating of larger bodies such as Earth and the Moon. This arises because their accretion is expected to occur on a timescale that is far too long (WETHERILL, 1986, 1990).

The only bodies that may have been thermally affected by ^{26}Al decay are the larger chondrite parent bodies, and even then only prior to ~4.563 Ga.

Initial ^{87}Sr/^{86}Sr

Another important ingredient that must be reconciled with any relative chronology in the early solar system is evolution of initial ^{87}Sr/^{86}Sr. This has been reviewed or discussed, for example, by WASSERBURG et al. (1977), TILTON (1988), and PODOSEK et al. (1991). The 'solar' Rb/Sr ratio is ~0.30 with an associated error of around 10% (ANDERS and GREVESSE, 1989). This corresponds to a ^{87}Rb/^{86}Sr ratio of ~0.92 at 4.56 Ga ago. Therefore, at a Rb/Sr ratio characteristic of the solar nebula, the ^{87}Sr/^{86}Sr will increase at the rate of ~19 ppm·Ma^{-1} or 1.3 × 10^{-5} Ma^{-1} in the absolute ratio. The time resolution between two initial Sr ratios based upon the measurement uncertainties is then at most ±2 Ma. In the case of the eucrites and angrites, which are now extremely volatile depleted (e.g., MITTLEFEHLDT and LINDSTROM, 1990), any resolvable difference in their initial ^{87}Sr/^{86}Sr ratios would then effectively date—in a relative sense—the time of abrupt decrease of the Rb/Sr ratio in their respective sources. This, then, would plausibly date the formation of their parent bodies, or at least that of volatile-poor precursor planetesimals. [In a more speculative sense, if the latter are the result of an early solar T-Tauri stage with the concomitant transition to gas-absent accretion (cf. WOOD and MORFILL, 1988), then this "age" may record just such an event.] Because of the low Rb/Sr ratio of these bodies, this age would be very insensitive to the actual, slightly younger crystallization age. Nonetheless, given that one should expect some notional time interval between parent body accretion and igneous differentiation, this 'age' is relative and a minimum one. Our finding that the initial ^{87}Sr/^{86}Sr ratios of eucrites and angrites are indistinguishable has an important consequence. Namely, it implies that the angrite and eucrite parent bodies probably were formed within ±2 Ma of one another. As will be discussed below, there are reasons for believing that the igneous history of the angrite parent body was far less complex than that of its counterpart, the eucrite parent body. This implies that the Pb-Pb age determined for LEW and ADOR is likely to be very close to the absolute formation age of both bodies. In what follows, these considerations are turned around somewhat to examine what can be learned about the absolute chronology of CAIs from these results.

The difference in initial Sr isotope composition between Moore County plagioclase and ALL reported by WASSERBURG et al. (1977) corresponds to $\Delta\epsilon_{Sr}$ of 2.1 ± 0.4 or 150 ± 30 × 10^{-6} in the absolute ^{87}Sr/^{86}Sr ratios. Using the solar nebula Rb/Sr ratio discussed above, this would imply that the eucrite parent body is 11 ± 4 Ma younger than the condensation age of ALL. Based upon our own finding that the initial Sr ratio of eucrites and angrites is indistinguishable, this should also correspond to the formation 'age' of the angrite parent body relative to ALL within uncertainty (±2 Ma). If this is indeed correct, then the oldest Allende CAI should have an 'absolute' age (using the U decay constants as a reference) of 4.569 ± 5 Ga; this constitutes a minimum estimate. One reason for calculating this age in this circumspect manner is that Pb-Pb age determinations on the cumulate eucrite Moore County have proved unsuccessful (TERA et al., 1987a). The Sm-Nd age of 4.457 ± 0.025 Ga for Moore County (TERA et al., 1987b) is in keeping with a general trend of younger ages for the cumulate compared to the noncumulate eucrites (TERA et al., 1989). As has already been pointed out, the Sr isotope initial effectively records the drastic reduction of the Rb/Sr ratio accompanying parent body formation and is insensitive to the precise age. However,

from the standpoint of ascribing absolute rather than relative ages, determination of the oldest Pb-Pb ages for these bodies is also necessary, and this is where the simpler igneous history of the angrite parent body becomes paramount.

In this connection, it is pertinent to ask whether there are, in fact, any eucrites other than Moore County that have older Pb-Pb ages. One such is the noncumulate eucrite Ibitira. Its Pb-Pb age has been reported as 4.560 ± 3 Ga (MANHÈS et al., 1987), although the apparent Sm-Nd age is 4.46 ± 0.02 Ga and has been partially reset according to PRINZHOFER et al. (1991). But more significantly, $^{146}Sm/^{144}Sm$ and initial $^{142}Nd/^{144}Nd$ of Ibitira agree within error with those of LEW, strongly suggesting similar ages. Unfortunately, though, it is clear that the igneous history of the eucrite parent body (or bodies) was more prolonged and complicated than that of the angrite parent body. A prime example of this is the enormous values for μ_1 (>100) of many cumulate eucrites (e.g., TERA et al., 1987a, 1989). The initial $^{87}Sr/^{86}Sr$ of Moore County (Table 2), however, suggests that the eucrite parent body formed within ± 2 Ma of the angrite parent body—say, between 4.556 and 4.560 Ga.

Lastly, the minimum age for the oldest Allende CAIs of 4.569 ± 5 Ga inferred from the initial Sr isotope composition is important for another reason. From the absence of evidence for live ^{26}Al in LEW, discussed earlier, a lower limit of 4.5635 ± 5 Ga was derived for the absolute age corresponding to an initial $^{26}Al/^{27}Al$ ratio of 5×10^{-5}. Hence, taking these age constraints together there is nothing inherently contradictory in having live ^{26}Al present in some of the older CAIs and a homogeneous ^{26}Al distribution in the early solar nebula. The recent combined Mg and Sr isotopic study of CAIs by PO-DOSEK et al. (1991) also provides tentative support for live ^{26}Al in the early solar system, chronologically related, in some cases, to $^{87}Sr/^{86}Sr$ evolution. However, we stress that there may yet be further complications. For a homogeneous ^{26}Al distribution, the range of initial $^{26}Al/^{27}Al$ ratios found in CAIs would necessitate a rather extended period of condensation or thermal processing compared to the <1 Ma predicted by the nebula evolution models, as is discussed below.

Planet Building

These observations do more than simply draw to attention some of the difficulties with determining an absolute chronology of the early solar system. They imply at face value that there are serious conflicts between dynamic models of proto-nebula collapse and condensation and any reasonable assessment of the isotopic timescales involved for these processes. As has been noted, models of proto-nebula evolution and condensation suggest that after <1 Ma asteroid-sized bodies have been formed (e.g., WOOD and MORFILL, 1988; WEIDENSCHILLING, 1988). In direct opposition to this appears to be the Sr isotopic evidence which implies a timescale of $\sim 11 \pm 4$ Ma between the first CAIs and the formation of the angrite and eucrite parent bodies.

The situation is further complicated by the chemical constraints on accretion of the carbonaceous chondrite parent bodies. As reviewed by WOOD (1985), this suggests that ac-

cretion of CAIs, chondrules, and matrix occurred very swiftly, perhaps even within a few years, of the last thermal processing event witnessed by the CAIs. If one accepts this evidence, then (1) Pb-Pb ages of CAIs effectively date the growth of these parent bodies, and (2) there would consequently be a time lag between the accretion of these bodies and those of the eucrite and angrite parent bodies.

There are three possibilities for reconciling these observations. The first is that the Rb/Sr ratio in the vicinity of the angrite and eucrite parent body accretion was enhanced by a factor approaching ~ 5 compared to the solar nebula value; for example, if the solar value is seriously in error or if much refractory Sr-rich material was fractionated away in these regions. Neither of these is particularly appealing. It is highly improbable that the solar photospheric value is in error by this amount (ANDERS and GREVESSE, 1989). In answer to the second point, the angrites and eucrites are products of parent bodies remarkably impoverished in volatile elements such as Rb (e.g., MITTLEFEHLDT and LINDSTROM, 1990) rather than the other way around. The second possibility is that the nebula models are seriously in error in predicting the times of collapse and condensation of the nebula. The last possibility is that the initial $^{87}Sr/^{86}Sr$ "ages" are indeed correct. This would lead to the suggestion that the angrite and eucrite parent bodies might have a secondary origin. That is, they might postdate the main condensation and planetesimal accretion phase of the solar nebula. For example, a late collisional origin is a possibility. This option would still allow a short (<1 Ma) evolutionary timescale for the solar nebula. It would also be quite compatible with the older (>4.56 Ga) ages recently reported from H4 chondrites (GÖPEL et al., 1991).

There is presently no reason to doubt the evolutionary timescales suggested by the solar nebula models. It follows that there would appear to be at least one major episode or event of asteriod "production" significantly later ($\sim 11 \pm 4$ Ma) than the appearance of the chondrite parent bodies. This is most likely to represent the later-stage sweep up of larger planetesimals in more energetic events, as discussed, for example, by WETHERILL (1986, 1990). The angrites appear to be among the earliest differentiates of such a body, composed of material that suffered catastrophic volatile element loss at this time. The combined Rb-Sr and U-Pb systematics in LEW and ADOR also imply, as already outlined, that the Rb/Sr and U/Pb ratios of the precursor materials be relatively unfractionated from solar or chondritic ratios compared to the emergent angrite parent body.

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