



Laboratory simulation of terrestrial meteorite weathering using the Bensour (LL6) ordinary chondrite

Martin R. LEE^{1*}, Caroline L. SMITH^{1†}, Sarah H. GORDON^{1‡}, and Mark E. HODSON²

¹Department of Geographical and Earth Sciences, University of Glasgow, Gregory Building, Lilybank Gardens, Glasgow G12 8QQ, UK

²Department of Soil Science, School of Human and Environmental Science, The University of Reading,
Whiteknights, Reading RG6 6AB, UK

[†]Present address: Department of Mineralogy, The Natural History Museum, Cromwell Road, London SW7 5BD, UK

[‡]Present address: Department of Earth Science and Engineering, South Kensington Campus, Imperial College, London, UK

*Corresponding author. E-mail: Martin.Lee@ges.gla.ac.uk

(Received 16 June 2005; revision accepted 29 May 2006)

Abstract—Laboratory dissolution experiments using the LL6 ordinary chondrite Bensour demonstrate that meteoritic minerals readily react with distilled water at low temperatures, liberating ions into solution and forming reaction products. Three experiments were performed, all for 68 days and at atmospheric fO_2 but using a range of water/rock ratios and different temperatures. Experiments 1 and 2 were batch experiments and undertaken at room temperature, whereas in experiment 3, condensed boiling water was dripped onto meteorite subsamples within a Soxhlet extractor. Solutions from experiment 1 were chemically analyzed at the end of the experiment, whereas aliquots were extracted from experiments 2 and 3 for analysis at regular intervals. In all three experiments, a very significant proportion of the Na, Cl, and K within the Bensour subsamples entered solution, demonstrating that chlorapatite and feldspar were especially susceptible to dissolution. Concentrations of Mg, Al, Si, Ca, and Fe in solution were strongly affected by the precipitation of reaction products and Mg and Ca may also have been removed by sorption. Calculations predict saturation of experimental solutions with respect to Al hydroxides, Fe oxides, and Fe (oxy)hydroxides, which would have frequently been accompanied by hydrous aluminosilicates. Some reaction products were identified and include silica, a Mg-rich silicate, Fe oxides, and Fe (oxy)hydroxides. The implications of these results are that even very short periods of subaerial exposure of ordinary chondrites will lead to dissolution of primary minerals and crystallization of weathering products that are likely to include aluminosilicates and silicates, Mg-Ca carbonates, and sulfates in addition to the ubiquitous Fe oxides and (oxy)hydroxides.

INTRODUCTION

Over the last twenty years, large numbers of meteorites have been recovered from the Earth's surface, principally from hot deserts, including the Sahara and Nullarbor (Australia), and cold deserts, mainly Antarctica. These finds include rare and scientifically important meteorites from asteroidal and planetary parent bodies, but all will have been mineralogically, chemically, and isotopically modified during their exposure at the Earth's surface of up to 10^6 years. For some uses, such as describing the petrology of chondrules or isolation and analysis of presolar grains, terrestrial weathering will be of little significance. However, for other applications, such as investigating processes and products of

aqueous alteration within asteroidal or planetary parent bodies, terrestrial weathering is potentially very important because its products may be difficult to distinguish from minerals formed by preterrestrial alteration (Gooding 1986; Lee et al. 2003).

Despite the clear importance of understanding the deleterious effects of terrestrial weathering, it has received relatively little attention. Most of the previous work has used chemical, isotopic, and spectroscopic techniques to quantify the accumulation of weathering products within a group of meteorite finds, usually ordinary chondrites whose terrestrial age is known, for example Holbrook (Bland et al. 1998a) and Tatahouine (Barrat et al. 1998), or has been determined by isotopic techniques (Bland et al. 1998b, 2000; Lee and Bland

2004). This work has shown that ordinary chondrites weather rapidly, even in arid environments (Bland et al. 1998a). The most abundant and easily characterized weathering products are Fe oxides and (oxy)hydroxides, which form by oxidation and hydration of Fe-rich metal, sulfide, and silicates, but sulfate minerals (e.g., gypsum) and carbonates (calcite, magnesite) can also be produced (Marvin 1980; Jull et al. 1988; Velbel 1988; Velbel et al. 1991; Barrat et al. 1998). The relative degrees of weathering of these finds may be determined directly by quantifying the volumes of reduced and oxidized iron within the rock (Bland et al. 1998b), or indirectly by measuring changes in the bulk chemical or isotopic composition of the meteorite resulting from acquisition of ions from the Earth's atmosphere or from soils on which the meteorite lies (Langenauer and Krähenbühl 1993; Shinonaga et al. 1994; Ash and Pillinger 1995).

The only previous study that has attempted to simulate terrestrial meteorite weathering in the laboratory was by Bland et al. (1997), who undertook experiments on powders of the H5 ordinary chondrite Allegan. Results showed that Fe,Ni metal and troilite reacted to produce akaganéite (FeOOH), but that olivine and pyroxene also underwent dissolution. Two other studies have described results of hydrothermal experiments on the CV3 fall Allende that were designed to mimic asteroidal aqueous alteration, but their results are relevant to the present work. Duke and Brearley (1998) reacted millimeter-size cubes of Allende with deionized water at 100–200 °C and for 7–60 days. They found that meteoritic minerals including olivine and troilite had altered to yield a range of reactions products including Na, Mg, and Ca sulfates; Ca carbonates; and Fe hydroxides. Brearley and Jones (2002) found that in the high temperature/short duration experiments (200 °C/30–90 days), olivine had been replaced by phyllosilicates, Fe oxides, and amorphous silica, whereas in lower temperature/longer duration experiments (100 °C/180 days), reaction products were limited to amorphous silica that had precipitated along grain boundaries, some of which had subsequently recrystallized to phyllosilicates.

The experiments described above have investigated aqueous alteration of meteorites mainly by characterizing solid reaction products. The aim of the present study is to identify the minerals that participate in dissolution reactions and, if possible, determine their relative susceptibilities to natural weathering by measuring the concentrations of ions in solution. We have used subsamples of the LL6 ordinary chondrite Bensour, a fall that was recovered in 2002 and has experienced negligible terrestrial alteration. A number of different experiments were performed, solutions were chemically analyzed, and the meteorite subsamples were characterized before and after the experiments. Results of this work demonstrate that chlorapatite and feldspar are especially susceptible to dissolution, but most constituents of ordinary chondrites will react readily with distilled water at low

temperatures. The implications of this study are that the impact of early terrestrial weathering on meteorites is not limited to alteration of metal and sulfide, but includes dissolution of phosphates and silicates, some of the solutes from which will be available to form (alumino)silicate, carbonate, and sulfate reaction products.

MATERIALS AND METHODS

Dissolution Experiments

This study has used subsamples of Bensour (LL6 ordinary chondrite), >45 kg of which was recovered from the region of the Moroccan-Algerian border in February 2002 (Gattacceca et al. 2003; Russell et al. 2004). This meteorite was collected shortly after its fall to Earth and we obtained a dozen fusion-crust stones, each a few g in size, from a private collector. The stones had been stored in small sealed polythene bags. Just before starting the experiments, each stone was mechanically broken to produce subsamples, ~0.3–1.4 g in weight, whose external surfaces comprise a mixture of fusion crust and freshly exposed meteorite interior. These subsamples were then ultrasonically cleaned in ultrapure deionized water (UDIW) to remove any particles adhering to grain surfaces prior to oven-drying at 30 °C.

Three sets of dissolution experiments were undertaken, all at atmospheric fO_2 , comprising two batch experiments (hereafter termed experiments 1 and 2) and one using a Soxhlet extractor (experiment 3). Experiment 1 was a single batch experiment consisting of a 0.3834 g subsample of Bensour that was left untouched in 0.019 L of UDIW within a PPE bottle in a shaking water bath at 25 °C. After 68 days, the meteorite subsample was removed, rinsed in UDIW, and oven-dried at 30 °C overnight. Solution from the opened bottle was divided into 3 aliquots. The pH and temperature of one was measured. The other two aliquots were then passed through 0.2 µm filters; one was acidified to 5% HNO₃. Both filtered aliquots were kept in the dark at 5 °C until analysis, the unacidified sample for SO₄²⁻ and Cl⁻ by ion chromatography and the acidified sample for Na, Mg, Al, Si, K, Ca, Mn, Fe, and Ni by inductively coupled plasma-optical emission spectroscopy (ICP-OES). Analytical precision, based on repeat analyses of randomly chosen solutions, was ±5%. Accuracy, based on analysis of an in-house aqueous standard, was ±10%. A blank experiment comprising a PPE bottle containing 50 mL UDIW that was sampled after 68 days was also run and the solution was analyzed as above. Reported concentrations in the text and tables have been blank-corrected by subtracting concentrations determined in the blank solution from those determined in the nonblank.

Experiment 2 comprised three batch experiments (2a, 2b, and 2c), all 68 days in duration, which used different 1.2–1.4 g subsamples of Bensour. Each subsample was placed in a 60 mL PPE bottle containing UDIW (using a ratio of 1 g

meteorite to 50 mL UDIW) and the three bottles were left in a shaking water bath at 25 °C. The starting pH values for experiments 2a, 2b, and 2c were 7.7, 7.6, and 7.7, respectively. In this experiment, 20 mL of solution were removed from each of the bottles after 1, 3, 7, 15, 21, 28, 35, 49, 54, and 68 days, then treated and analyzed as described above. After each aliquot of solution was removed, 20 mL of fresh UDIW were added to the bottles and the experiments continued. It was assumed that dissolution of the meteorite would be sufficiently rapid that periodic removal of solution and replenishment with fresh solution would have a relatively minor impact on dissolution kinetics, especially when sampling was carried out at intervals of one week or more. Blank experiments were run in two PPE bottles, each containing 50 mL UDIW that were sampled on the same dates as solutions from experiment 2 and analyzed as above. All reported concentrations were again blank-corrected.

The Soxhlet extractor (Hodson 2002) was used in experiment 3 to more closely mimic conditions under which meteorites are weathered in hot deserts. The apparatus is designed so that UDIW at 34–46 °C drips onto the meteorite subsamples that are contained within a cellulose thimble 1.4 cm in diameter. Pure water is recirculated through the extractor via boiling and condensation, and solutes accumulate in the water reservoir at the base of the apparatus for sampling and chemical analysis. Prior to starting the experiments, the extractor was run for two days with the thimble in place but without the meteorite samples. It was then turned off, allowed to cool, and rinsed in UDIW. The reservoir at the bottom of the extractor was then filled with a fresh 200 mL of UDIW, four Bensour subsamples (each weighing between 1.6 and 4.7 g, a total mass of 12.2 g) were placed in the thimble, and the extraction was started. Twenty mL of solution were retrieved from the reservoir after 1, 2, 3, 7, 15, 21, 28, 35, 49, 55, and 68 days and chemically analyzed using the same procedure as described above. Following removal of each aliquot of solution, 20 mL of fresh UDIW were added. Note that blanks were not run in parallel with this experiment since only one extractor was available, but a subsequent 7-day blank experiment was undertaken whereby the apparatus was run under the same conditions, but containing only the cellulose thimble with UDIW. This experiment was designed to provide an assessment of the potential leaching of ions from the walls of the apparatus, which is made of borosilicate glass (Pyrex 7740) with a composition of 81% SiO₂, 13.0% B₂O₃, 4.0% Na₂O, 2.0% Al₂O₃, and 0.5% K₂O. Modeling of the saturations of solutions from experiments 1, 2, and 3 before blank correction was performed using PHREEQC (Parkhurst and Appelo 1999) and the WATEQ4F database (Ball and Nordstrom 1991), assuming equilibrium with atmospheric CO₂ and O₂. For a given phase, a saturation index greater than 1 indicates saturation, and in theory, that phase should precipitate, although this does not take into account reaction kinetics or nucleation energies, which may hinder crystallization.

Petrographic and Mineralogical Characterization

Unaltered subsamples of Bensour and pieces of the meteorite that had been recovered from the dissolution experiments were mounted in epoxy resin, polished, and coated with carbon. Backscattered electron (BSE) images and qualitative X-ray chemical analyses of the polished samples were acquired using a FEI Quanta 200F field-emission scanning electron microscope (SEM) equipped with an EDAX Pegasus 2000 energy-dispersive X-ray microanalysis system (EDS). High-resolution secondary electron images of gold-coated surfaces of fresh and reacted meteorite subsamples were acquired using the same SEM. Quantitative chemical analyses of minerals within the polished thin sections were obtained using a Cameca SX50 electron probe operated at 15 kV/10 nA for feldspar analyses and 20kV/20 nA for other minerals. Mineral standards were used and data were quantified using an on-line PAP correction procedure.

The bulk chemical composition of a small, unweathered subsample of Bensour was determined by ICP-AES (Varian Vista Pro, axial) at the Natural History Museum, London. The instrument was calibrated using synthetic standards checked against international standard rock samples. Major elements, Cr, and Ni were determined on a LiBO₂ fusion of a 0.0994 g subsample dissolved in 250 mL of 4% HNO₃. For S and trace elements, a further 0.1040 g subsample was dissolved using HF/HClO₄/HNO₃, and made up to 10 mL in 5% HNO₃. The relative abundances of minerals within Bensour were determined by point counting using the SEM. The microscope stage was moved in equal steps over the sample and the material in the middle of the field of view at each point was imaged and then identified by qualitative chemical analysis using EDX. The weight percent of each mineral in Bensour was subsequently calculated from the volumetric abundance of each mineral and its typical density.

RESULTS

Mineralogy and Chemical Composition of the Unweathered Meteorite

Bensour is a very fine-grained monomict breccia composed of olivine, orthopyroxene (OPX), clinopyroxene (CPX), plagioclase feldspar, Fe,Ni metal, troilite, and chromite (Russell et al. 2004). Gattacceca et al. (2003) described evidence for shock, including twinning and deformation of troilite and kamacite grains, which is indicative of shock stage S3. Figure 1 is a BSE image with corresponding X-ray maps of a typical area of the interior of the meteorite. Individual grains are ~20–150 μm in size and between and within them are interconnected, vein-like networks of pores (Fig. 1a). The mean porosity of Bensour, determined by SEM point counting, is 11.0 vol%. The Mg, Si, and Fe X-ray maps show that olivine and OPX are the most

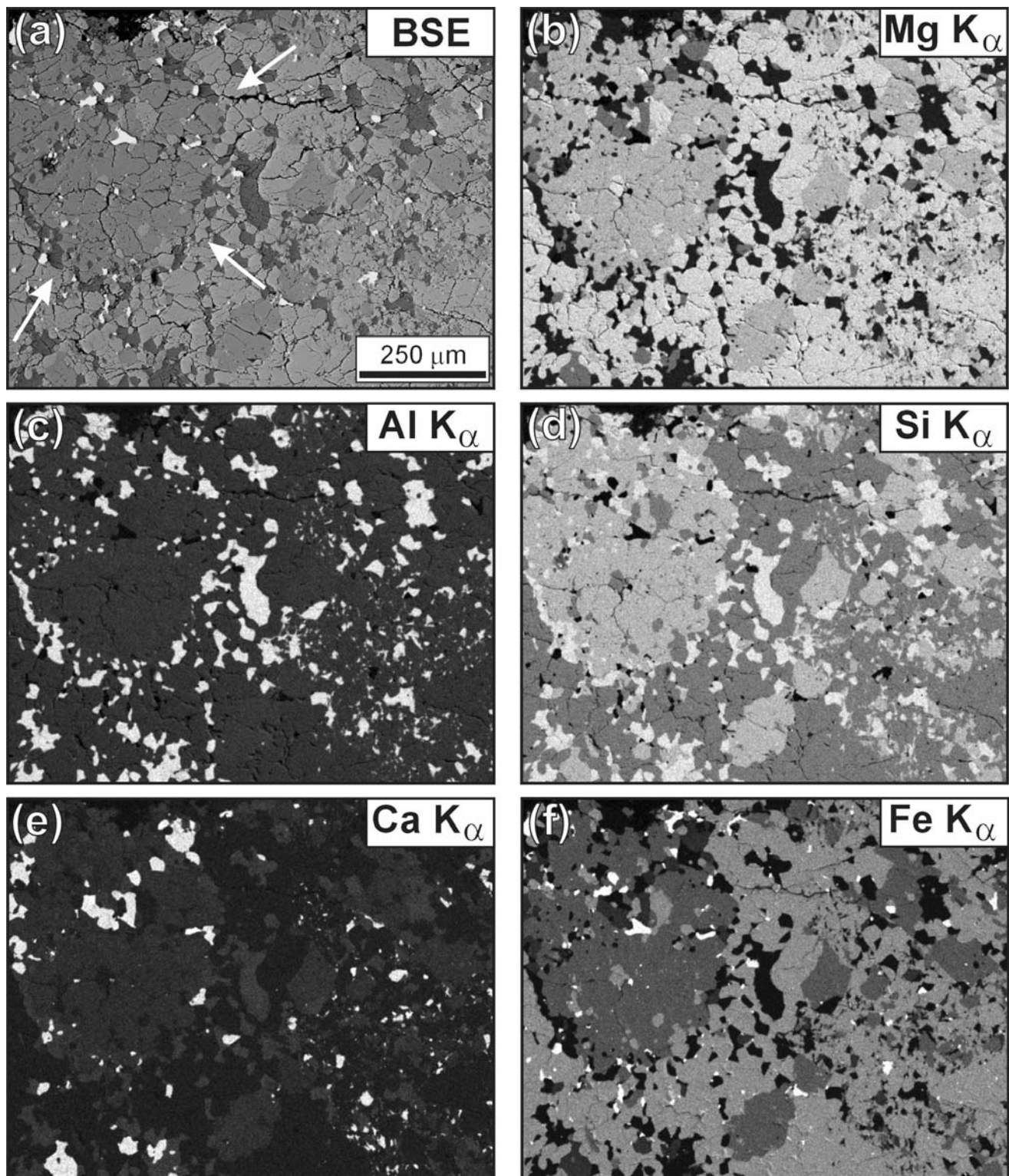


Fig. 1. Images of a fresh sample of Bensour. a) A backscattered electron image showing that most of the minerals have a similar mean atomic number apart from feldspar (medium gray) and metal/sulfide grains (white). Vein-line networks of pores (black) follow grain boundaries and cross-cut grain interiors. A former chondrule is indicated by arrows. Images (b–f) are corresponding X-ray maps. Areas of high counts (light gray) in the Mg K_α map (b) show the location of olivine, OPX, and CPX. White areas in the Al K_α map (c) are feldspar, whereas the Si K_α X-ray map (d) distinguishes feldspar (white) from OPX (light gray) and olivine (darker gray). Areas with high counts in the Ca K_α map (e) are CPX whereas the Fe K_α map (f) shows the location of troilite and Fe,Ni metal (both small white grains) and helps to distinguish olivine (light gray), from OPX and CPX (medium gray) and feldspar (black).

Table 1. The modal abundances of minerals in Bensour compared with Saint-Séverin (LL6).

Mineral	Abundance of solids (wt%)					
	Bensour area 1	Bensour area 2	Bensour area 3	Bensour area 4	Bensour mean	Saint-Séverin
Olivine	46.57	41.05	57.75	62.44	51.89	52.2
OPX	25.79	28.45	18.11	13.61	21.53	20.9
CPX	6.72	5.01	4.01	5.42	5.29	7.08 ^a
Feldspar	11.77	11.73	12.68	13.47	12.41	10.50
Fe,Ni metal	3.10	1.18	0.00	1.21	1.37	1.82
Troilite	5.61	11.65	5.51	3.11	6.50	6.16
Chromite	0.44	0.00	1.19	0.75	0.59	1.06
Ca phosphate	0.00	0.92	0.75	0.00	0.42	0.34 ^b
Total	100.0	100.0	100.0	100.0	100.0	100.1
<i>n</i>	306	193	243	218	960	–

^aPigeonite plus diopside.

^bDescribed as whitlockite by Gastineau-Lyons et al. (2002).

The material at each point was chemically analyzed by SEM-EDX and data exclude porosity that comprises 11.0 vol% of the whole rock. *n* = the number of points counted in each area. Saint-Séverin data are from Gastineau-Lyons et al. (2002).

abundant constituents of Bensour (Figs. 1b, 1d, and 1f) and ~0.5 mm-size aggregates of OPX grains represent former chondrules (Fig. 1a). Feldspar is also abundant (Figs. 1c and 1d), but grains are only a few tens of micrometers in size. The main Ca-bearing mineral is CPX, which is again fine-grained but considerably less abundant than feldspar (Fig. 1e) and neither Fe,Ni metal nor troilite is common (Fig. 1f).

The modal abundances of minerals found within four areas of one subsample are listed in Table 1. Most minerals vary in abundance over a relatively narrow range but olivine and OPX show the greatest variation, part of which may reflect occasional difficulties in distinguishing olivine and OPX in X-ray spectra. We recognize the problems in obtaining representative mineral abundances from a relatively small sample of such a heterogeneous material (Keil 1962; Gastineau-Lyons et al. 2002), but the mineralogy of Bensour compares reasonably well with the modal mineralogy of Saint-Séverin (LL6) determined by Gastineau-Lyons et al. (2002) (Table 1). The chemical compositions of the main Bensour silicate minerals are listed in Table 2. Olivine and OPX compositions determined here fall between previous analyses of these minerals in Bensour by Cole and Sipiera (2003) (olivine $\text{Fa}_{30.7}$, OPX $\text{Fs}_{25.4}$) and Russell et al. (2004) (olivine $\text{Fa}_{31.6}$, OPX $\text{Wo}_{3.5}\text{Fs}_{24.3}$). Gattacceca et al. (2003) found that most of the metal in Bensour comprises taenite (average composition Ni 45.23%, Co 1.87%) with a tetrataenite rim whereas kamacite (average composition Ni 4.11%, Co 8.21%) is rare.

The bulk chemical composition of Bensour determined by ICP-AES is listed in Table 3 (column 1). The Bensour subsample used for this analysis is far smaller than the 10 g that Keil (1962) and Jarosewich (1990) recommend for a representative analysis of an ordinary chondrite but even if a bulk chemical composition determined from 10 g of Bensour were available, it would almost certainly differ from the ~1 g-size subsamples used in experiments 1 and 2. Also listed

in Table 3 is a bulk composition for Bensour calculated using the abundance and chemical composition of each mineral (listed in Tables 1 and 2). As analyses of Bensour Ca phosphate, troilite, and chromite were unavailable, stoichiometric chlorapatite, $\text{Ca}_5(\text{PO}_4)_3\text{Cl}$; troilite, FeS; and chromite, FeCr_2O_4 have been assumed for these calculations. The bulk chemical compositions for Bensour obtained by the two methods agree fairly well with each other and are also similar to the bulk chemical composition of Saint-Séverin (LL6) listed in Jarosewich (1990) (Table 3).

The mineralogy and bulk chemical compositions described above do not take into account the fusion crust that was present on subsamples used in the dissolution experiments. Genge and Grady (1999) showed that the outer parts of fusion crusts of ordinary chondrites contain newly formed olivine; Na-, Al-, and Ca-rich aluminosilicate glass; and Ni-rich sulfide droplets. As a result, the crusts typically have lower Mg/Si and higher Na/Si, Al/Si, and Ca/Si values than the bulk meteorite. However, fusion crusts from three LL6 meteorites listed in Genge and Grady (1999) do not differ significantly in Si-normalized Na, Al, Mg, and Ca values to bulk Bensour. The influence of fusion crust on relative abundances of elements released during the experiments is therefore inferred to have been minor in comparison to mineral grains in the interior of each subsample.

Dissolution Experiment 1

The concentration of ions in solution after 68 days of immersion of a 0.3 g subsample of Bensour in distilled water are listed in Table 4. To express these data more meaningfully, they have been used to calculate the total mass of each element that entered and remained in solution as a percentage of its initial mass within the subsample of Bensour used in the experiment (Table 5). These data show that a total of 32.5% of

Table 2. The chemical compositions of the main silicate minerals in Bensour.

Element	Mean chemical composition (wt%) ^a			
	Olivine	OPX	CPX	Feldspar
SiO ₂	37.45	54.59	53.41	64.64
TiO ₂	0.00	0.13	0.28	n.a.
Al ₂ O ₃	0.00	0.23	0.57	21.80
FeO	28.02	16.27	7.51	n.a.
MnO	0.36	0.34	0.21	n.a.
MgO	34.85	26.81	16.51	0.07
CaO	0.01	1.54	20.34	2.48
Na ₂ O	0.01	0.04	0.63	9.87
K ₂ O	0.00	0.00	0.00	0.80
Total	100.7	99.95	99.46	99.66
<i>n</i>	152	61	5	16
	Molecular proportions			
	Fa = 31.1	En = 72.3	En = 46.7	An = 11.7
		Wo = 3.0	Wo = 41.3	Ab = 83.9
		Fs = 24.7	Fs = 12.0	Or = 4.4

^aDetermined by electron probe microanalysis. n.a. = not analyzed.
n = number of analyses used to determine the mean.

the initial Cl, 2.7% of the K, and 1.8% of the Na within Bensour entered solution over the 68 days of the experiment but less than 1% of the Mg, Al, Si, S, and Ca were liberated.

The experiment produced no detectable change in macroscopic color of the subsample and no evidence for etching of primary minerals could be found by secondary electron SEM imaging although some grains have a very finely crystalline coating that is interpreted to be composed of an Fe(oxy)hydroxide, probably goethite (Figs. 2a and 2b). On a small number of grains the Fe (oxy)hydroxides are overlain by euhedral crystals, 1–2 μm in size, of an unidentified layered Mg-silicate (Figs. 2a and 2b). Calculations using PHREEQ indicate that solutions were saturated with respect to several Fe oxides and (oxy)hydroxides; Al hydroxides and hydrous aluminosilicates; and Na-, Mg-, K-, Ca-, and Fe-bearing hydrous aluminosilicates. Although the blank solutions were undersaturated with respect to all phases, the solubility of the Al hydroxides and Fe (oxy)hydroxides is such that even hypothetical solutions with concentrations set at the detection limits quoted at the bottom of Table 4 and pH in the range 7–9 (i.e., that of the experimental solutions) are calculated to be saturated with respect to these phases. Concentrations of Al and Fe in solution are therefore inferred to have been controlled by precipitation of reaction products.

Dissolution Experiment 2

The three batches of experiment 2 yielded Na, Mg, Al, Si, and Ca in measurable concentrations, but SO₄²⁻, Cl⁻, and Fe were at or close to detection limits in both blank and experimental solutions (Table 4). Potassium, Mn, and Ni were

Table 3. The bulk chemical composition of Bensour compared with Saint-Séverin (LL6).

Element	Bulk chemical composition (wt%)		
	Bensour ^a	Bensour ^b	Saint-Séverin
Si	18.23	19.65	18.98
Ti	0.07	0.03	0.07
Al	1.10	1.47	1.25
Cr	0.39	0.28	0.40
Fe	20.28	19.31	20.17
Mn	0.27	0.21	0.25
Mg	15.56	14.92	15.20
Ca	1.39	1.39	1.37
Na	0.61	0.94	0.74
K	0.08	0.08	0.09
P	0.10	0.07	0.05
Ni	0.98	0.62	1.05
Co	0.06	0.03	0.04
S	2.06	2.34	2.12
Cl	n.d.	0.03	n.d.
C	n.d.	n.d.	0.02

^aFrom an ~0.1 g sample of Bensour analyzed by ICP-AES.

^bCalculated from the mineralogy of Bensour, determined by point counting, and the density of each mineral.
n.d. = not determined. Saint-Séverin data are from Jarosewich (1990), but the original analysis was in Jarosewich and Mason (1969).

sought but not found. These data have been used to calculate the total mass of each element released by dissolution of the meteorite between successive sampling intervals and are expressed as a percentage of its initial mass within the subsample of Bensour used (Table 5). Negative values indicate that ions have been removed from solution between sampling intervals by precipitation and/or absorption reactions or reflect uncertainty in the analysis. The “total” value for each element represents the net mass liberated from the subsample over the duration of the experiment (as a percentage of its initial abundance in the meteorite). This value will be greater than the total mass of element remaining in solution at the end of the experiment because solutes were removed in sampling and precipitation/absorption may also have taken place.

In common with experiment 1, the proportion of Na and Cl initially within the Bensour subsample that entered solution is considerably greater than Mg, Al, Si, S, and Ca. As the solutions were not sampled at regular intervals during the experiment, for an assessment of temporal changes in the masses of ions entering solution, it is necessary to divide values in each column of Table 5 by the duration of time in days since the previous sampling event (listed in the second column of Table 5). Results show that the greatest daily release of Na, Mg, Al, Si, and Ca to solution was at the start of the experiment and decreased abruptly over the first two weeks. The very regular decrease in liberation of Na and Ca is shown in Fig. 3, whereas the release of the other three elements fluctuates and Cl shows no clear trends. The

Table 4. The chemical composition and pH of solutions from experiments 1 and 2.

Sampling interval (days)	Concentration ($\mu\text{g L}^{-1}$)									pH
	Na	Mg	Al	Si	SO ₄ ²⁻	Cl ⁻	K	Ca	Fe	
Experiment 1 (0.3 g subsample), blank-corrected values										
68	2230	6140	47	6500	750	1580	428	1750	16	7.4
Blank values										
	115	2	d.l.	81	330	80	d.l.	19	6	4.3
Experiment 2a (1.3 g subsample), blank-corrected values										
1	324	538	29	234	d.l.	110	d.l.	360	0.5	7.7
3	416	774	30	426	d.l.	330	d.l.	384	1.7	8.2
7	512	1032	97	908	300	d.l.	d.l.	467	43	8.7
15	601	1200	120	1440	d.l.	75	d.l.	532	5	8.8
21	530	1100	119	1380	420	d.l.	d.l.	473	d.l.	8.0
28	477	1010	115	1400	200	65	d.l.	432	d.l.	6.5
35	369	1000	76	1370	d.l.	d.l.	d.l.	385	d.l.	7.7
49	451	1040	148	1850	295	85	d.l.	410	5	7.4
54	704	1070	130	2080	d.l.	d.l.	d.l.	482	d.l.	5.6
68	302	1090	126	1790	d.l.	d.l.	d.l.	375	1.4	4.8
Experiment 2b (1.2 g subsample), blank-corrected values										
1	337	623	12	195	d.l.	90	d.l.	386	5	7.6
3	418	913	39	425	d.l.	d.l.	d.l.	389	d.l.	8.6
7	494	1110	78	1000	260	d.l.	d.l.	476	6	9.0
15	609	1200	120	1300	d.l.	35	d.l.	525	d.l.	9.3
21	507	1070	133	1310	d.l.	d.l.	d.l.	464	6	8.4
28	452	990	116	1310	d.l.	d.l.	d.l.	419	d.l.	7.5
35	369	870	66	1440	160	d.l.	d.l.	373	d.l.	9.0
49	520	1170	170	1910	35	15	d.l.	475	d.l.	8.7
54	239	1070	113	1940	240	d.l.	d.l.	388	d.l.	8.4
68	371	1110	197	1890	10	580	d.l.	394	50	7.6
Experiment 2c (1.4 g subsample), blank-corrected values										
1	319	600	2	170	150	10	d.l.	204	d.l.	7.7
3	360	963	20	510	95	d.l.	d.l.	303	d.l.	8.5
7	426	1310	83	1000	d.l.	d.l.	d.l.	471	3	9.3
15	566	1550	136	1460	20	195	d.l.	577	d.l.	9.4
21	513	1430	150	1610	130	35	d.l.	535	d.l.	9.3
28	467	1390	162	1510	d.l.	45	d.l.	551	4	8.7
35	391	1190	106	1750	60	d.l.	d.l.	447	d.l.	9.2
49	491	1270	210	1990	d.l.	d.l.	d.l.	454	1	9.0
54	283	1200	166	2160	140	d.l.	d.l.	398	d.l.	8.8
68	446	1170	219	2110	480	20	d.l.	378	d.l.	8.2
Blank values										
1	319	5	17	257	d.l.	190	d.l.	16	35	4.4
3	360	6	19	669	265	160	d.l.	25	33	4.4
7	426	4	d.l.	1430	d.l.	165	d.l.	19	11	4.1
15	566	2	d.l.	129	170	125	d.l.	10	39	4.2
21	513	3	9	789	d.l.	145	d.l.	12	19	4.1
28	467	4	9	94	d.l.	135	d.l.	22	17	4.0
35	391	59	74	179	345	225	d.l.	26	82	4.3
49	491	2	d.l.	144	215	185	d.l.	19	9	4.4
54	283	13	45	d.l.	250	280	d.l.	61	89	4.3
68	446	5	d.l.	57	280	d.l.	d.l.	81	12	4.0
Detection limits ($\mu\text{g L}^{-1}$)										
–	7	0.1	6	9	150	60	152	0.2	4	–

d.l. = present in concentrations below detection limits. The detection limit for Mn was $0.3 \mu\text{g L}^{-1}$ and for Ni was $9 \mu\text{g L}^{-1}$. Some values in the table are less than reported detection limits since they are blank-corrected.

Table 5. The mass of elements released to solution during experiments 1 and 2 expressed as a percentage of their initial abundance in Bensour.

Day	Time since last sampling event (days)	Mass of element liberated since last sampling event as a percentage of its initial abundance in Bensour							
		Na	Mg	Al	Si	S	Cl	K	Ca
Experiment 1									
68	68	1.83	0.20	0.02	0.23	0.06	32.51	2.68	0.63
Experiment 2a									
1	1	0.27	0.02	0.01	0.01	0.00	2.26	–	0.13
3	2	0.16	0.01	0.00	0.01	0.00	5.23	–	0.05
7	4	0.18	0.02	0.03	0.02	0.02	–4.70	–	0.07
15	8	0.20	0.02	0.02	0.02	–0.02	1.54	–	0.07
21	6	0.09	0.01	0.02	0.01	0.03	–1.07	–	0.04
28	7	0.09	0.01	0.01	0.01	–0.01	1.34	–	0.04
35	7	0.03	0.01	0.00	0.01	–0.01	–0.92	–	0.03
49	14	0.16	0.01	0.04	0.02	0.02	1.75	–	0.05
54	5	0.32	0.01	0.01	0.02	–0.02	–1.21	–	0.07
68	14	–0.15	0.01	0.02	0.01	0.00	0.00	–	0.02
	Total	1.51	0.12	0.18	0.14	0.08	12.12	–	0.57
Experiment 2b									
1	1	0.28	0.02	0.01	0.01	0.00	1.85	–	0.14
3	2	0.16	0.02	0.01	0.01	0.00	–1.21	–	0.05
7	4	0.18	0.02	0.02	0.02	0.02	0.00	–	0.08
15	8	0.23	0.02	0.03	0.02	–0.01	0.72	–	0.08
21	6	0.09	0.01	0.02	0.01	0.00	–0.47	–	0.04
28	7	0.10	0.01	0.01	0.01	0.00	0.00	–	0.04
35	7	0.06	0.01	0.00	0.02	0.01	0.00	–	0.04
49	14	0.23	0.02	0.06	0.03	–0.01	0.31	–	0.08
54	5	–0.08	0.01	0.00	0.02	0.02	–0.20	–	0.03
68	14	0.18	0.01	0.06	0.02	–0.01	11.93	–	0.05
	Total	1.51	0.14	0.23	0.15	0.05	14.81	–	0.63
Experiment 2c									
1	1	0.26	0.02	0.00	0.00	0.01	0.21	–	0.07
3	2	0.11	0.02	0.01	0.01	0.00	–0.15	–	0.06
7	4	0.14	0.02	0.03	0.02	–0.01	0.00	–	0.09
15	8	0.22	0.02	0.03	0.02	0.00	4.01	–	0.09
21	6	0.09	0.01	0.02	0.02	0.01	–2.13	–	0.05
28	7	0.08	0.01	0.03	0.01	–0.01	0.41	–	0.06
35	7	0.05	0.01	0.00	0.02	0.00	–0.66	–	0.02
49	14	0.17	0.01	0.06	0.02	0.00	0.00	–	0.05
54	5	–0.05	0.01	0.01	0.02	0.01	0.00	–	0.03
68	14	0.20	0.01	0.05	0.02	0.03	0.41	–	0.03
	Total	1.33	0.14	0.24	0.15	0.07	4.90	–	0.55

Values have been calculated using data in Table 4 and the bulk chemical composition of Bensour determined by ICP-AES (Table 3) apart for Cl, where a value of 243 ppm, derived from point counting, has been used. A negative value indicates that the mass of element in solution was lower than that remaining after extraction of the previous aliquot of solution. Fe is not listed as it was present in concentrations above detection limits in only a few aliquots.

experiments produced no detectable change in the macroscopic color of the subsamples, and no unambiguous evidence for etching of mineral grain surfaces or crystallization of reaction products could be found by secondary electron SEM imaging.

All solutions that contained Fe in concentrations above detection limits were calculated to be saturated with respect to Fe oxides and (oxy)hydroxides with magnetite and hematite

having the highest saturation indices. After only one day of immersing the Bensour subsamples in distilled water, the solutions are predicted to have been saturated with respect to Fe oxides and (oxy)hydroxides, Al hydroxides, and hydrous aluminosilicates including kaolinite and pyrophyllite. However, as noted above, blank solutions and solutions with compositions set equal to the detection limits are calculated to be saturated with respect to Al hydroxides and Fe

(oxy)hydroxides for pH values equal to those of the nonblank experimental solutions. As successive solutions taken from any one batch of experiment 2 varied between being undersaturated and oversaturated with respect to aluminosilicates and silicates, it is by no means certain that those minerals would have precipitated.

Dissolution Experiment 3 (Soxhlet)

The chemical compositions of output solutions from experiment 3 are listed in Table 6. Data from the subsequent control experiment are not listed because concentrations of anions and cations were variable over the 7 days and low (less than 10% of totals recorded in the meteorite experiment). There was no systematic increase in solute concentrations as might be expected if the glass walls of the Soxhlet apparatus were also reacting. However, in the discussion that follows it should be borne in mind that concentrations of elements and ions released from the meteorite could be as much as 10% lower than reported. The data in Table 6 show that in addition to the Na, Mg, Al, Si, and Ca recorded in experiments 2a–c, SO_4^{2-} , Cl^- , K, and Fe were above detection limits in solutions taken at each sampling interval. Manganese and Ni were sought, but in all cases were below detection limits. These data have been recalculated to show the mass of each element released as a percentage of its initial abundance within Bensour in the same manner as for experiment 2 (Table 7). Results show that a much greater proportion of Na, Si, S, Cl, and K were liberated to solution during the Soxhlet experiment than in experiments 1 and 2. The differences in Al and Ca between the three experiments are not so marked and a considerably smaller proportion of Mg entered solution during the Soxhlet experiment than the other two. The proportion of Na, K, and Ca liberated per day is greatest at the start of experiment 3 then decreases, although it does fluctuate. The greatest rates of liberation of Si and Cl are also at the beginning of the experiment, but subsequently values vary considerably. The daily release of Al, Mg, and S shows no clear trends.

By the end of the experiment the subsamples used had all changed color from off-white to light brown. Secondary electron SEM images of the postreaction meteorite subsamples provide evidence for dissolution of some grains and precipitation of reaction products. The walls of cleavages and fractures within CPX grains have been etched to form pits that are elongate in one crystallographic direction, producing a sawtooth-like pattern on the submicrometer scale (Fig. 4a). Each tooth is ~20–35 nm in diameter at its narrowest point and is capped by an equant structure 40–90 nm in diameter (Fig. 4b). The surfaces of many mineral grains have a very finely crystalline coating composed of rod-shaped crystals ~160 × 40 nm in size (Fig. 4c) whose morphology is characteristic of goethite, or interpenetrating aggregates of disc- or plate-shaped crystals ~1–2 μm in size (Fig. 4d), which is typical of hematite. Other reaction products were

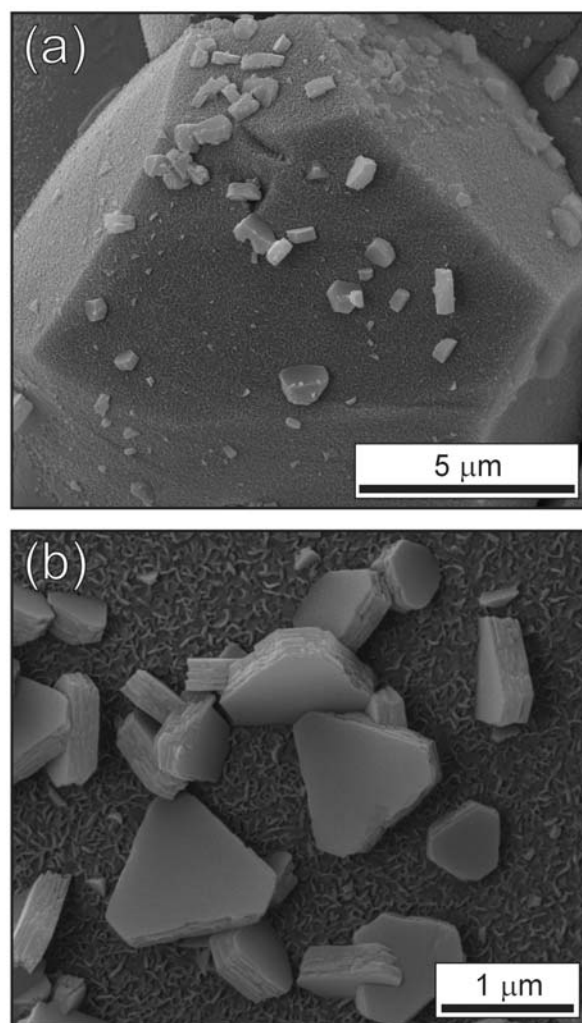


Fig. 2. Secondary electron SEM images of reaction products in the interior of a sample of Bensour recovered from experiment 1 after 68 days. a) A grain coated with a very finely crystalline reaction product, probably a Fe (oxy)hydroxide, and coarser euhedral crystals of an unidentified layered Mg-silicate. b) The layered Mg-silicate crystals, which clearly postdate Fe (oxy)hydroxides.

very scarce, although the interiors of some postreaction meteorite subsamples that had been fractured in the laboratory contained micron-sized aggregates of acicular/fibrous crystals (Fig. 4e). These crystals were too small for their chemical compositions to be unambiguously determined by EDX, but were Ni-rich. In one area, several grains had a coating of silica 2–3 μm in thickness which was itself encrusted by nanocrystalline goethite (Fig. 4f). A considerable quantity of white powder was recovered from the Soxhlet apparatus after 68 days and qualitative X-ray analysis showed it to be O- and Si-rich but also containing detectable concentrations of Na, Mg, and Al. Calculations performed using PHREEQC indicate that the solutions were saturated with respect to a variety of Al hydroxides, Fe oxides and (oxy)hydroxides, silica, and a range of hydrous silicates and aluminosilicates.

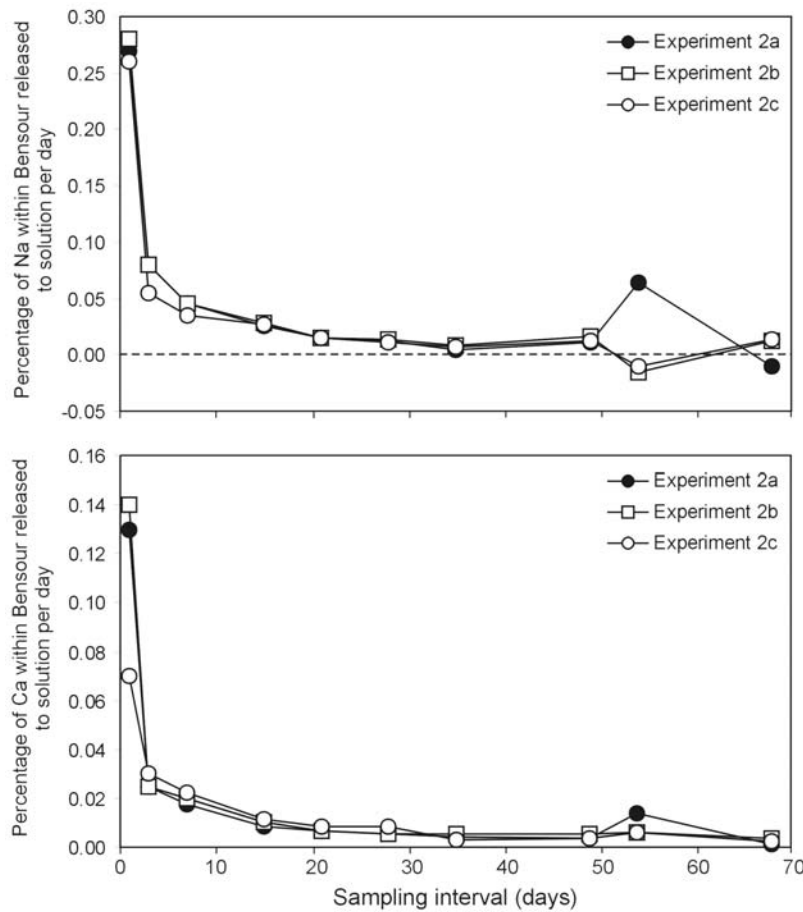


Fig. 3. Graphs illustrating the change in daily release of Na and Ca to solution during experiment 2.

DISCUSSION

Mechanisms of Water-Meteorite Interaction

The liberation of solutes and precipitation of reaction products that accompanies immersion of subsamples of Bensour in distilled water provides clear evidence for considerable water-rock interaction, even though the time scale of the experiments is orders of magnitude shorter than the duration of subaerial exposure of most meteorite finds. Interaction of the water with the subsamples of Bensour may be facilitated by the significant volume of interconnected pores within this meteorite, although such porosity and permeability is typical of many ordinary chondrite falls (Lee and Bland 2004). The reaction products that were found on the surfaces of mineral grains exposed by laboratory fracturing of the postreaction subsamples would have grown into these intergranular and intragranular pores. There are no indications of etching of the surfaces of mineral grains from the subsamples that have been immersed in distilled water at room temperature for 68 days (i.e., experiments 1 and 2), which reflects the limited volumes of minerals that have been

dissolved. However, some CPX grains recovered from experiment 3, which were exposed to lower water/rock ratios but greater temperatures, have been extensively etched to form long narrow channels with denticulated margins (Fig. 4a). This sawtooth or “cockscorb” morphology of etched grain surfaces is very common in naturally weathered pyroxenes and reflects the growth of lenticular etch pits elongated parallel to the *c*-axis of the pyroxene crystal (Berner and Schott 1982; Velbel 1993). It is possible that Fe-rich mineral grains were also etched during experiment 3, but their surfaces are now obscured by thick Fe oxide and (oxy)hydroxide coatings.

Dissolution and Relative Solubilities of Bensour Minerals

Some or all analyzed solutions from all three experiments contained detectable concentrations of Na, Mg, Al, Si, SO_4^{2-} , Cl^- , K, Ca, and Fe. The potential sources of these ions can be determined by multiplying the concentration of each ion in a given mineral by the abundance of that mineral within Bensour. Results of these calculations (Table 8) show that some ions can have only come from a

Table 6. The chemical composition and pH of output solutions from experiment 3.

Sampling interval (days)	Concentration ($\mu\text{g L}^{-1}$)									pH
	Na	Mg	Al	Si	SO_4^{2-}	Cl^-	K	Ca	Fe	
1	9690	547	402	25,600	1210	1470	1310	1230	26	8.6
2	14,800	100	660	48,800	1300	1460	2240	1800	3	8.4
3	16,000	54	818	55,700	1490	1400	2430	2250	8	8.4
7	25,100	35	788	95,100	2870	1850	3870	3910	12	8.8
15	33,800	38	380	142,000	4220	1860	5150	4960	13	9.0
21	37,600	20	224	164,000	5500	2200	5650	5500	11	8.8
28	40,400	19	109	193,000	5930	1900	5870	4920	d.l.	8.7
35	65,100	26	3121	347,000	9190	2520	9830	7830	20	8.5
49	63,000	33	781	287,000	11,000	3240	7830	3990	43	8.5
54	83,200	112	964	323,000	17,000	4760	8940	3150	10	8.5
68	55,800	46	211	147,000	10,200	3390	5260	1610	11	8.4
Detection limits ($\mu\text{g L}^{-1}$)										
–	7	0.1	6	9	150	60	152	0.2	4	–

The experiment used a total mass of 12.2 g of Bensour. Mn and Ni were all below detection limits of 0.3 and 9 $\mu\text{g L}^{-1}$, respectively. d.l. = present in concentrations below detection limits.

Table 7. The mass of elements released to solution during experiment 3 expressed as a percentage of their initial abundance in Bensour.

Day	Time since last sampling event (days)	Mass of element liberated since last sampling event as a percentage of its initial abundance in Bensour							
		Na	Mg	Al	Si	S	Cl	K	Ca
1	1	2.60	0.01	0.06	0.23	0.03	9.92	2.68	0.15
2	1	1.63	0.00	0.04	0.23	0.01	0.92	2.17	0.08
3	1	0.72	0.00	0.03	0.11	0.01	0.58	0.85	0.07
7	4	2.88	0.00	0.01	0.40	0.04	3.98	3.45	0.22
15	8	3.01	0.00	–0.05	0.51	0.04	1.32	3.42	0.17
21	6	1.93	0.00	–0.02	0.33	0.05	3.55	2.08	0.12
28	7	1.76	0.00	–0.01	0.41	0.03	–0.54	1.61	0.00
35	7	7.72	0.00	0.45	1.56	0.10	5.33	9.32	0.40
49	14	1.19	0.00	–0.30	–0.23	0.07	6.68	–2.08	–0.36
54	5	7.12	0.00	0.04	0.58	0.19	12.44	3.88	–0.05
68	14	–5.13	0.00	–0.10	–1.29	–0.14	–6.03	–5.71	–0.14
Total		30.57	0.01	0.63	4.35	0.57	44.71	29.46	1.22

Values have been calculated using data in Table 6 and the bulk chemical composition of Bensour determined by ICP-AES (Table 3) apart for Cl, where a value of 243 ppm, derived from point counting, has been used. A negative value indicates that the mass of element in solution was lower than that remaining after extraction of the previous aliquot of solution. Fe is not listed since it was present in very low concentrations or below detection limits.

single mineral, whereas others have two or more potential sources. The rate of liberation of each ion from each mineral is a function of its solubility, given the temperature, pH, and chemical composition of experimental solutions, and the access of water to mineral grain surfaces, which will depend on the porosity and permeability of each subsample and the presence or absence of reaction-product coatings on grain surfaces. The abrupt decrease in the daily rate of release of ions to solution during the first ~20 days of experiments 2 (Fig. 3) and 3 followed by a “steady state” or fluctuating rates of release is a pattern that is commonplace in batch dissolution experiments (e.g., Busenberg and Clemency 1976; Holdren and Berner 1979). The initially high rates accompany attack of the fresh mineral surfaces by aqueous solutions, and subsequent fluctuations in the accumulation of ions in

solution are inferred to be caused by saturation and chemical affinity effects (Oelkers et al. 2001).

Below we attempt to determine the relative solubilities of Bensour minerals by unraveling the competing influences of dissolution, precipitation, and absorption. Note that blank-corrected solution compositions are used throughout.

Dissolution of Ca Phosphate

Between 4.9 and 44.7% of the Cl originally contained within the Bensour subsamples was liberated during the three experiments, indicating that chlorapatite, the only Cl-bearing mineral found, was especially susceptible to dissolution. However, these values are likely to be minimum estimates because the bulk Cl concentration of Bensour used for calculating the total proportion of the element that was

liberated during each experiment (243 ppm) is almost certainly too high, as it assumes that all of the Ca phosphate found by point counting was stoichiometric chlorapatite. In fact, ordinary chondrites are known to also contain merillite and whitlockite, although the ratios of the different phosphates minerals within ordinary chondrites is highly variable (Pellas and Storzer 1974). If a value of 70 ppm Cl is assumed for Bensour, which is the mean bulk Cl concentration of LL5 and L6 falls and finds listed in Garrison et al. (2000), the mass of Cl liberated during experiment 3 corresponds to 55% more than contained within the starting material. This discrepancy suggests that the Bensour subsamples used may have contained another highly soluble Cl-rich mineral, such as halite (NaCl), which has been described from the H chondrite breccias Monahans (1998) and Zag by Rubin et al. (2002). This possibility is difficult to evaluate because such a soluble mineral is unlikely to have survived manufacture of thin sections and polished blocks and Na in the experimental solutions cannot be used to demonstrate that halite did dissolve because Na is also likely to have been liberated by dissolution of feldspar.

Dissolution of Feldspar

The presence of Na, Al, and K in the experimental solutions demonstrates that feldspar dissolved. As ~30% of the Na and K originally within the Bensour subsamples used in experiment 3 was liberated, feldspar was highly soluble under the conditions used. The total percentages of Na and K that were liberated from the subsample used in experiment 1 are much lower than in experiment 3, but still greater than all of the other ions apart for Cl. Potassium was below detection limits in experiment 2, but the proportion of Na that was released to solution is again high relative to other elements apart for Cl. The Na/K values of solutions from experiment 1 (5.2) and experiment 3 (6.5–10.6) are lower than Bensour feldspar (11.1), indicating that feldspar dissolved nonstoichiometrically or a proportion of the Na was removed from solution, probably by precipitation of reaction products. Saturation calculations confirm that a number of Na- and K-rich minerals may have crystallized during the experiments 1 and 3, including montmorillonite and illite.

Since feldspar also contains 97% of the Al within Bensour, a significant proportion of each subsample's Al should have been liberated during the experiments. However, in all three experiments, the percentage of Al released to solution was far lower than Na and K. For example, ~30% of the Al originally within the subsamples used in experiment 3 should have been released to solution (i.e., the same percentage as Na and K), but only 0.6% was found. The "missing" 29.4% corresponds to 39.5 mg of Al, which is inferred to have been removed from solution by crystallization of reaction products, presumably the Al hydroxides and aluminosilicates that are predicted to have been precipitated by calculations performed using PHREEQC.

Dissolution of Olivine, OPX, and CPX

The degree of dissolution of olivine, OPX, and CPX is difficult to determine because none of the solutes found in the three experiments are unique to any of these minerals. A proportion of the Si in solution must have been derived from feldspar in addition to one or all of olivine, OPX, and CPX, but saturation calculations also indicate that silica-rich reaction products are likely to have precipitated, and were found in reacted meteorite subsamples from experiment 3. As 28% of Bensour Ca is contained within chlorapatite and feldspar, these minerals may have supplied most of Ca in solution, but the presence of etched CPX grains in the subsamples used in experiment 3 (Fig. 4a) shows that CPX will have been the source of a proportion of the Ca. Some of the Mg in solution may also have been derived from CPX, but as olivine and OPX together contain 96% of Bensour Mg, these minerals will have been the main sources. Since a far lower proportion of Mg that was initially contained within the Bensour subsamples entered solution during experiment 3 than experiments 1 and 2, Mg must have been removed by reactions within the Soxhlet apparatus. The growth of minerals such as montmorillonite and illite may have been responsible, although none were found by SEM, or Mg may have been removed by sorption on to reaction products and divalent cations including Mg and Ca are known to be susceptible to sorption (McBride 1994).

Dissolution of Troilite and Fe,Ni metal

The presence of SO_4^{2-} in experimental solutions indicates that troilite has dissolved, although <0.6% of the S initially within the Bensour subsamples was liberated in any of the experiments. As concentrations of Fe in solution are very low in all the experiments, most of the Fe^{2+} liberated by dissolution of troilite, and probably also from olivine, OPX, CPX, and Fe,Ni metal, must have been scavenged from solution to form Fe oxides and (oxy)hydroxides. Such minerals were observed on the surfaces of subsamples used in experiments 1 and 3 and were calculated to have formed during all three experiments. These results are not surprising as the experiments were undertaken at atmospheric $f\text{O}_2$ so that Fe^0 and Fe^{2+} will have rapidly oxidized to Fe^{3+} . However, while it seems most likely that the observed Fe oxide and (oxy)hydroxide coatings were derived by dissolution of Fe-rich minerals, saturation calculations indicate that the concentrations of Fe recorded in the blank solutions would result in saturation with respect to Fe (oxy)hydroxide phases at the pH values recorded in the nonblank experimental solutions, so it is possible, albeit unlikely, that the coatings are derived from the precipitation of background Fe concentrations.

Comparison of Dissolution Rates of Bensour Minerals with Terrestrial Mineral Powders

The experimental results demonstrate that under the conditions used, chlorapatite and feldspar dissolved rapidly,

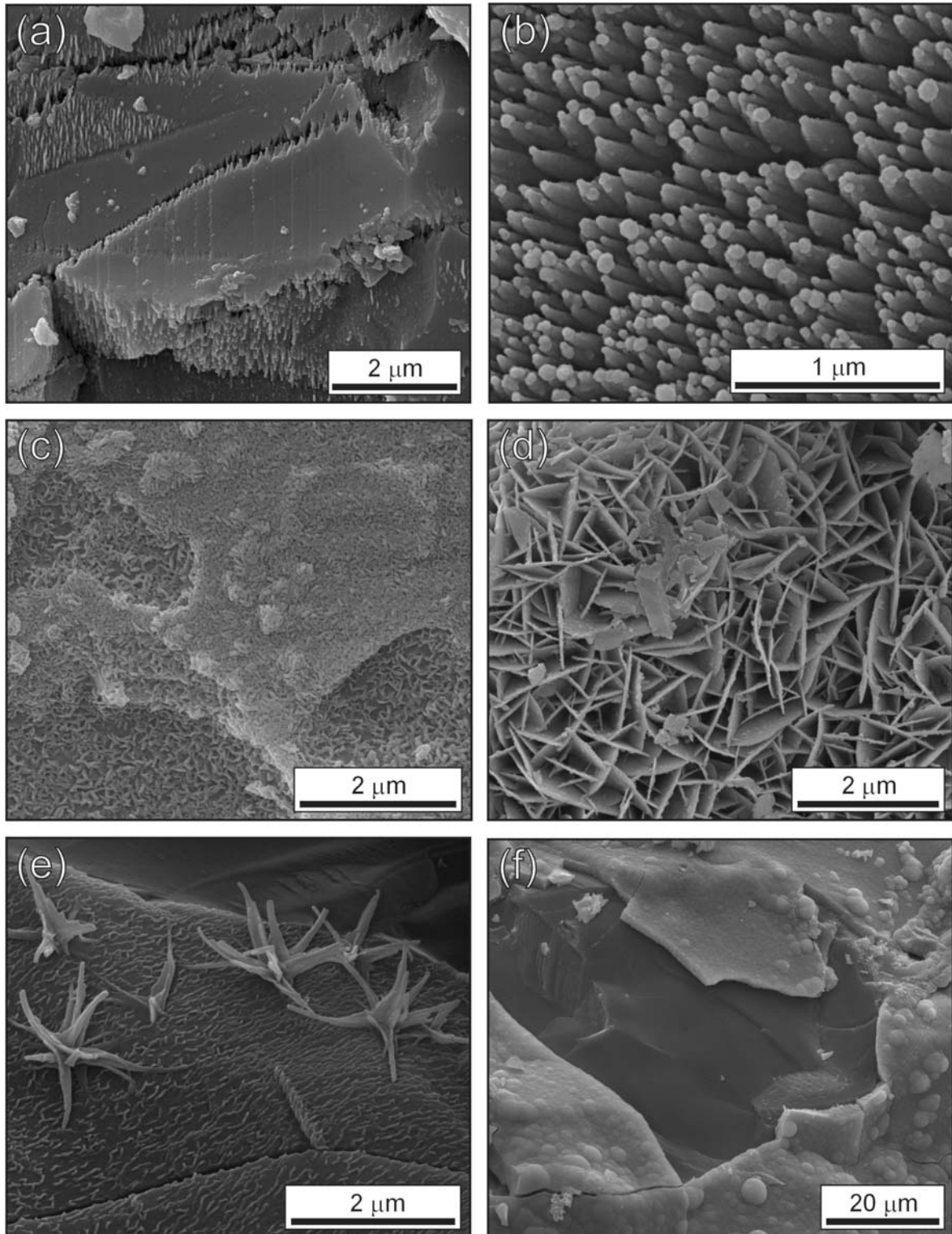


Fig. 4. Secondary electron SEM images of a subsample of Bensour recovered from experiment 3 after 68 days. a) A CPX grain showing the development of elongate etch pits on the walls of fractures. b) A heavily etch-pitted CPX surface with spires of CPX upstanding between elongate etch pits. c) The surface of a mineral grain encrusted by rod-shaped Fe-rich crystals, probably goethite. d) A grain surface encrusted by aggregates of platy Fe-rich crystals, interpreted to be hematite. e) Aggregates of fibrous Ni-rich reaction products overlying a mineral surface coated with finely crystalline goethite. f) A silica grain coating, which is itself encrusted by nanocrystalline goethite, that has partially broken from the underlying olivine/OPX grain, probably during preparation for SEM work.

Table 8. The proportions of each element that is present within the main Bensour minerals.

Element	Proportion (%)						
	Olivine	OPX	CPX	Feldspar	Troilite	Fe,Ni	Ca phosphate
Na	1	1	3	96	0	0	0
Mg	73	23	4	0	0	0	0
Al	0	2	1	97	0	0	0
Si	46	28	7	19	0	0	0
S	0	0	0	0	100	0	0
Cl	0	0	0	0	0	0	100
K	0	0	0	100	0	0	0
Ca	0	17	55	16	0	0	12
Fe	61	15	2	0	5	18	0

liberating a significant proportion of Na, Cl, and K within the starting materials. Although comparable proportions of Al must also have entered solution, this element was very effectively scavenged to form reaction products. Laboratory experiments using powders of terrestrial minerals confirm that apatite dissolves rapidly. For example, in experiments on Durango fluorapatite ($\text{Ca}_{10}(\text{PO}_4)_6\text{F}_2$) undertaken at 25 °C, 1 atm, and pH 7.3–8.45, Guidry and MacKenzie (2003) recorded a dissolution rate of $\sim 10^{-10}$ moles m^{-2} sec^{-1} . However, terrestrial feldspar (albite) dissolves at a much slower rate than fluorapatite ($\sim 6.3 \times 10^{-13}$ moles m^{-2} s^{-1}) (Brantley 2003). Results from experiments 1 and 2 are consistent with meteoritic chlorapatite dissolving much more rapidly than feldspar, assuming that grains of these minerals within Bensour are of a comparable size (i.e., have similar surface area to volume ratios). By contrast, more comparable proportions of Na, K, and Cl entered solution in experiment 3, suggesting more equal rates of dissolution over the 68-day experiment. As outlined above, values for the proportions of Cl initially contained within the Bensour subsamples that entered solution (Tables 5 and 7) must be treated with caution owing to uncertainties about the initial Cl concentration of Bensour and the possible presence of one or more other soluble Cl-rich minerals.

The laboratory dissolution rates of terrestrial forsterite and fayalite (both 3.6×10^{-10} moles m^{-2} s^{-1}) (Brantley 2003) and enstatite (3.2×10^{-11} moles m^{-2} s^{-1}) (Brantley 2003) are close to those of fluorapatite and considerably greater than feldspar. Thus, the proportions of Mg and Si originally within the Bensour subsamples that were released to solution should have been comparable to the proportions of Cl and greater than the proportions of Na and K liberated in each experiment (again assuming comparable grain sizes within Bensour). However, the proportions of Mg and Si recorded apparently indicate that olivine and OPX dissolved at a considerably slower rate than chlorapatite and feldspar. One explanation could be that coatings of Fe oxide and (oxy)hydroxide reaction products were deposited on surfaces of the Fe-rich silicate grains, inhibiting interdiffusion of ions between mineral surfaces and solutions. Such a process has been invoked by Bland et al. (1998a, 1998b) to account for the

decrease over time in weathering rates of ordinary chondrite falls from hot deserts. However this mechanism, termed “passivation,” is unlikely to have played a significant role in influencing rates of laboratory dissolution because the thickness of reaction product coatings required to slow dissolution rates is likely to be far greater than were observed in this study. Theoretical and experimental work also indicates that Fe oxide and (oxy)hydroxide coatings are unlikely to have a measurable impact on dissolution rates of silicate minerals (Velbel 1993; Hodson 2003). It is more probable that the apparently slow rates of olivine and OPX dissolution relative to chlorapatite is simply due to the effective removal of Mg from solution by absorption and removal of Mg and Si together by precipitation of reaction products. This conclusion emphasizes the important role of removal of ions from solution to form reaction products during these experiments, despite the relatively high water/rock ratios used relative to those typical of subaerial meteorite weathering.

CONCLUSIONS

Results of this study have shown that equilibrated ordinary chondrites react readily with distilled water so that even under the relatively high water/rock ratios of the batch experiments, precipitation of reaction products had a major influence on the residence times of ions in solution. Sodium, Cl, K, and Ca released from chlorapatite and feldspar had relatively long residence times, allowing solutes to accumulate, whereas Mg, Al, Si, and Fe were more readily removed to form reaction products. As the mineralogy and volumetric abundances of reaction products were difficult to determine, a full mass-balance could not be calculated and so the relative solubilities of the different minerals was difficult to determined with accuracy. Within the much lower water/rock conditions of subaerially exposed meteorite finds the Al, Mg, Si, and Fe ions liberated during terrestrial weathering should form aluminosilicate and Fe (oxy)hydroxide weathering products within inter- and intragranular pores very close to the dissolving silicates. These minerals are likely to be accompanied by alkali-

bearing silicate, sulfate, or carbonate reaction products, possibly located further from their parent minerals within the meteorite or on its outer surfaces. It is important that further work on terrestrially weathered meteorite finds is undertaken to determine whether such secondary minerals are indeed widespread and if any petrographic or chemical criteria can be used to reliably distinguish them from the preterrestrial silicate, carbonate, and sulfate minerals that have been recorded from carbonaceous and ordinary chondrite and achondrite falls.

Acknowledgments—We are grateful to the Paneth Trust for providing funds for an undergraduate internship to Sarah (Ginger) Gordon and we are also very grateful to Rob Elliott for donating the samples of Bensour. We also thank John Gileece (Glasgow) for making the polished blocks and thin sections, Robert MacDonald (Glasgow) for assistance with the electron probe, and Gary Jones (Natural History Museum) for ICP-AES analysis of Bensour. Lastly, we would like to express our gratitude to Beda Hoffman and Lysa Chizmadia and for very prompt and exceptionally useful reviews that have greatly improved this manuscript.

Editorial Handling—Dr. Ian Franchi

REFERENCES

- Ash R. D. and Pillinger C. T. 1995. Carbon, nitrogen and hydrogen in Saharan chondrites: The importance of weathering. *Meteoritics* 30:85–92.
- Ball J. W. and Nordstrom D. K. 1991. *WATEQ4F: User's manual with revised thermodynamic database and test cases for calculating speciation of major, trace and redox elements in natural waters*. Denver, Colorado: United States Geological Survey.
- Barrat J. A., Gillet P., Lecuyer C., Sheppard S. M. F., and Lesourd M. 1998. Formation of carbonates in the Tatahouine meteorite. *Science* 280:412–414.
- Berner R. A. and Schott J. 1982. Mechanism of pyroxene and amphibole weathering II. Observations of soil grains. *American Journal of Science* 282:1214–1231.
- Bland P. A., Kelley S. P., Berry F. J., Cadogan J. M., and Pillinger C. T. 1997. Artificial weathering of the ordinary chondrite Allegan: Implications for the presence of Cl⁻ as a structural component of akaganéite. *American Mineralogist* 82:1187–1197.
- Bland P. A., Berry F. J., and Pillinger C. T. 1998a. Rapid weathering in Holbrook: An iron-57 Mössbauer spectroscopy study. *Meteoritics & Planetary Science* 33:127–129.
- Bland P. A., Sexton A. S., Jull A. J. T., Bevan A. W. R., Berry F. J., Thornley D. M., Astin T. R., Britt D. T., and Pillinger C. T. 1998b. Climate and rock weathering: A study of terrestrial age dated ordinary chondritic meteorites from hot desert regions. *Geochimica et Cosmochimica Acta* 62:3169–3184.
- Bland P. A., Jull A. J. T., and Bevan A. W. R. 2000. Ancient meteorite finds and the Earth surface environment. *Quaternary Research* 53:131–142.
- Brantley S. L. 2003. Reaction kinetics of primary rock-forming minerals under ambient conditions. In *Treatise on geochemistry*, edited by Drever J. I. New York: Elsevier. pp. 73–117.
- Brearley A. J. and Jones C. L. 2002. Aqueous alteration of FeO-rich olivine: Insights from experimental alteration of Allende (abstract). *Meteoritics & Planetary Science* 37:A23.
- Busenberg E. and Clemency C. V. 1976. The dissolution kinetics of feldspar at 25 °C and 1 atm CO₂ partial pressure. *Geochimica et Cosmochimica Acta* 40:41–49.
- Cole K. J. and Sipiera P. P. 2003. Kilabo and Bensour: A comparative study of two recent LL6 falls from Africa (abstract #1335). 34th Lunar and Planetary Science Conference. CD-ROM.
- Duke C. L. and Brearley A. J. 1998. Experimental aqueous alteration of Allende (CV3) (abstract). *Meteoritics & Planetary Science* 33:A43.
- Garrison D., Hamlin S., and Bogard D. 2000. Chlorine abundances in meteorites. *Meteoritics & Planetary Science* 35:419–429.
- Gastineau-Lyons H. K., McSween H. Y., Jr., and Gaffey M. J. 2002. A critical evaluation of oxidation versus reduction during metamorphism of L and LL group chondrites, and implications for asteroid spectroscopy. *Meteoritics & Planetary Science* 37:75–89.
- Gattacceca J., Rochette P., and Bourot-Denise M. 2003. Magnetic properties of a freshly fallen LL ordinary chondrite: The Bensour meteorite. *Physics of the Earth and Planetary Interiors* 140:343–358.
- Genge M. J. and Grady M. M. 1999. The fusion crusts of stony meteorites: Implications for atmospheric reprocessing of extraterrestrial materials. *Meteoritics & Planetary Science* 34:341–356.
- Guidry M. W. and MacKenzie F. T. 2003. Experimental study of igneous and sedimentary apatite dissolution: Control of pH, distance from equilibrium, and temperature on dissolution rates. *Geochimica et Cosmochimica Acta* 67:2949–2963.
- Gooding J. L. 1986. Clay-mineraloid weathering products in Antarctic meteorites. *Geochimica et Cosmochimica Acta* 50:2215–2223.
- Hodson M. E. 2002. Experimental evidence for mobility of Zr and other trace elements in soils. *Geochimica et Cosmochimica Acta* 66:819–828.
- Hodson M. E. 2003. The influence of Fe-rich coatings on the dissolution of anorthite at pH 2.6. *Geochimica et Cosmochimica Acta* 67:3355–3363.
- Holdren G. R. and Berner R. A. 1979. Mechanisms of feldspar weathering—I. Experimental studies. *Geochimica et Cosmochimica Acta* 43:1161–1171.
- Jarosewich E. 1990. Chemical analyses of meteorites: A compilation of stony and iron meteorite analyses. *Meteoritics* 25:323–337.
- Jarosewich E. and Mason B. 1969. Chemical analysis with notes on one mesosiderite and seven chondrites. *Geochimica et Cosmochimica Acta* 33:411–416.
- Jull A. J. T., Cheng S., Gooding J. L., and Velbel M. A. 1988. Rapid growth of magnesium-carbonate weathering products in a stony meteorite from Antarctica. *Science* 242:417–419.
- Keil K. 1962. Quantitativ-erzmikroskopische Intergrationanalyse der Chondrite. *Chemie der Erde* 22:281–348.
- Langenauer M. and Krähenbühl U. 1993. Halogen contamination in Antarctic H5 and H6 chondrites and relation to sites of recovery. *Earth and Planetary Science Letters* 120:431–442.
- Lee M. R. and Bland P. A. 2004. Mechanisms of weathering of meteorites recovered from hot and cold deserts and the formation of phyllosilicates. *Geochimica et Cosmochimica Acta* 68:893–916.
- Lee M. R., Bland P. A., and Graham G. 2003. Preparation of TEM samples by focused ion beam (FIB) techniques: Applications to the study of clays and phyllosilicates in meteorites. *Mineralogical Magazine* 67:581–592.
- Marvin U. B. 1980. Magnesium carbonate and magnesium sulfate

- deposits on Antarctic meteorites. *Antarctic Journal of the United States* 15:54–55.
- McBride M. B. 1994. *Environmental chemistry of soils*. Oxford: Oxford University Press. 406 p.
- Oelkers E. H., Schott J., and Devidal J.-L. 2001. On the interpretation of closed system mineral dissolution experiments: Comments on “Mechanism of kaolinite dissolution at room temperature and pressure: Part II. Kinetic study” by Huertas et al. (1999). *Geochimica et Cosmochimica Acta* 65:4429–4432.
- Parkhurst D. L. and Appelo C. A. J. 1999. *User’s guide to PHREEQC (Version 2)—A computer program for speciation, batch-reaction, one-dimensional transport, and inverse geochemical calculations*. U.S. Geological Survey Water-Resources Investigations Report 99-4259. 310 p.
- Pellas P. and Storzer D. 1975. Uranium and plutonium in chondritic phosphates. *Meteoritics* 10:471–473.
- Rubin A. E., Zolensky M. E., and Bodnar R. J. 2002. The halite-bearing Zag and Monahans (1998) meteorite breccias: Shock metamorphism, thermal metamorphism and aqueous alteration on the H-chondrite parent body. *Meteoritics & Planetary Science* 37:125–141.
- Russell S. S., Folco L., Grady M. M., Zolensky M. E., Jones R., Richter K., Zipfel J., and Grossman J. N. 2004. The Meteoritical Bulletin No. 88. *Meteoritics & Planetary Science* 39:A215–272.
- Shinonaga T., Endo K., Ebihara M., Heumann K. G., and Nakahara H. 1994. Weathering of Antarctic meteorites investigated from contents of Fe³⁺, chlorine and iodine. *Geochimica et Cosmochimica Acta* 58:3735–3740.
- Velbel M. A. 1988. The distribution and significance of evaporitic weathering products on Antarctic meteorites. *Meteoritics* 23: 151–159.
- Velbel M. A., Long D. T., and Gooding J. L. 1991. Terrestrial weathering of Antarctic stone meteorites: Formation of Mg-carbonates on ordinary chondrites. *Geochimica et Cosmochimica Acta* 55:67–76.
- Velbel M. A. 1993. Formation of protective surface layers during silicate-mineral weathering under well-leached, oxidizing conditions. *American Mineralogist* 78:405–414.
-