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Why aqueous alteration in asteroids was isochemical: High porosity ≠ high permeability

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ABSTRACT

Carbonaceous chondrite meteorites are the most chemically primitive rocks in the solar system, but the most compositionally pristine have experienced pervasive aqueous alteration, apparently within asteroid parent bodies. Unfractionated soluble elements suggest minimal flow of liquid water, consistent with data from oxygen isotopes, and meteorite petrography. However, numerical studies persistently predict large-scale water transport in model asteroids. These models have tended to use permeabilites in the range 10^{-13} to 10^{-11} m^2. We show that the permeability of plausible chondritic starting materials lies in the range 10^{-19} to 10^{-17} m^2 (0.1 – 10 μD): around six orders-of-magnitude lower than previously assumed. This is largely a result of the extreme fine grain size of primitive chondritic materials. Applying these permeability estimates in numerical models, we predict very limited liquid water flow (distances of 100’s μm at most), even in a high porosity, water-saturated asteroid, with a high thermal gradient, over millions of years. Isochemical alteration, with minimal fluid flow, is not a special circumstance. It is inevitable, once we consider the fundamental material properties of these rocks. To achieve large-scale flow would require average matrix grain sizes in primitive materials of 10’s – 100’s μm – orders of magnitude larger than observed. Finally, in addition to reconciling numerical modelling with meteorite data, our work explains several other features of these enigmatic rocks, most particularly, why the most chemically primitive meteorites are also the most altered.

KEYWORDS

meteorite; carbonaceous chondrite; asteroidal alteration; isochemical alteration; permeability

1. Introduction

The first studies of carbonaceous chondrites (CCs) (e.g. Daubrée, 1867) recognised that their mineralogy was highly altered, with many samples containing clay minerals, magnetite, and carbonates etc. Initially, the abundant magnetite found in some CCs was thought to be a nebula condensate (Larimer and Anders, 1967), or, together with clay minerals, the product of aqueous alteration that occurred in the solar nebula by the reaction of water vapour and anhydrous minerals (Grossman and Larimer, 1974). However, a range of evidence now suggests that aqueous alteration occurred within CC parent asteroids (Brearley, 2003).

1.1. Chemistry

Curiously, extensive alteration within asteroids did not modify CC chemistry. CI-type carbonaceous chondrites have compositions that are within 10% of the solar photosphere for most elements (Palme and Jones, 2003). Their similarity to photosphere abundances, and the fact that CIs can be analysed in the lab, mean that they are frequently used to refine photosphere abundances. Defining an ‘average’ solar system composition is also a property which has made them the geochemical standard against which all other terrestrial rock analyses are normalised. Yet CIs are also the most aqueously altered CCs. Why the most chemically primitive rocks should have experienced the most alteration has always been a paradox. Early analyses of other CC groups showed uniform volatility-controlled depletions compared to CI (Kallemeyn and Wasson, 1981). These rocks also experienced varying degrees of aqueous alteration. To preserve solar abundances in elements which are easily mobilised in fluids it was postulated that aqueous processing in asteroids was isochemical (McSween, 1979), occurring in a closed system (Kerridge et al., 1979). It is noteworthy that recent estimates of solar chemical composition not only confirm the excellent agreement between photospheric and CI chondrite abundances (Grevesse & Asplund 2007), they also show that there is no evidence for fractionation of soluble elements. Ignoring light elements, and elements where photospheric abundances are poorly constrained, the mean difference between photospheric and CI abundances is 0.014 ± 0.06 dex (Grevesse & Asplund 2007). Selecting the most soluble elements from this group (25% of the total), the mean difference between photospheric and meteoritic abundances is 0.016 ± 0.06 dex ie. there is no significant difference between photosphere / meteorite compositions in the overall dataset, and photosphere / meteorite compositions in the soluble element subset. Finally, isochemical / closed-system alteration requires the flow of liquid water to be essentially zero (Brearley, 2003). More recent chemical analyses of small (10-100 mg) bulk chondrite aliquots (Wulf et al., 1995) show remarkable reproducibility between samples, and no evidence for element mobility. Similarly, analyses of the trace element composition of fine-grained materials in CCs (Bland et al., 2005) indicate minimal aqueous mobility.

1.2. Petrography

Geochemical analysis is broadly in agreement with chondrite petrography, where evidence for metasomatism is generally restricted to distances of less than a few 100 µm. Aqueous mobility over ~100 µm appears to have occurred in some unequilibrated ordinary chondrites, with 'bleaching' of chondrules and zonation in moderately volatile elements and Ca (e.g. Grossman et al., 2000, 2002; Grossman and Alexander, 2004). Similarly, evidence for iron-alkali aqueous metasomatism is found around chondrules and CAIs in CV chondrites (Krot et al., 1995). In the CR2 chondrites, zones of alteration are observed around chondrules, where aqueous elements have exchanged with surrounding rim or matrix material. In the CR2s these zones are 50-100µm thick (Burger and Brearley, 2004, 2005). A number of studies have investigated metasomatism in the CM chondrites. Chizmadia and Brearley (2004) and Brearley and Chizmadia (2005) have studied the behaviour of Fe, S, Ni, Ca, Na, K and P during aqueous alteration of CM2 carbonaceous chondrites. Chondrule mesostasis, in even the least altered of these meteorites, is completely replaced by phyllosilicates. But although chondrule glass is replaced, and a proportion of the aqueous elements within it dissolved in the altering fluid, element mobility is restricted to a layer ~10 µm in thickness occurring at the chondrule-rim boundary (in the case of less altered CM2s), or more homogenous distribution within the rim (Brearley and Chizmadia 2005), up to 25 µm from the chondrule (in the case of more altered CM2s). In the CI chondrites, in an extensive study, Morlok et al. (2006) found that the variable level of alteration observed in these meteorites was consistent with a very low mobility of materials in solution during aqueous alteration, probably on a scale of only 100 µm: closed system alteration was favoured (Morlok et al.. 2006). From this review it is apparent that even in rocks that have experienced substantial aqueous alteration, such as the CM2 chondrites, elemental mobility was largely restricted to comparatively narrow zones around chondrules. In the majority of cases, evidence for metasomatism over length scales >>100's µm is lacking.

1.3. Alteration timescales

Mn-Cr carbonate data can be interpreted as suggesting extended timescales for continual aqueous alteration within C1 and C2 parent bodies (e.g. Hoppe et al., 2007; de Leuw et al., 2009; Petitat et al., 2009; Tyra et al., 2009). However, thermodynamically (given fine-grained reactants) this argument is difficult to support. Rather, as noted by Hoppe et al. (2007), episodic alteration possibly resulting from impact heating and remobilisation of fluids is also consistent with the existing Mn-Cr dataset. A plausible scenario would involve the bulk of aqueous alteration occurring soon after asteroid accretion (when heat generated from decay of short-lived radionuclides was available for melting water), and being relatively brief, as alteration reactions are exothermic, and anhydrous reactants are fine-grained (Wood and Walther, 1983); Olsen et al. 1988; Zolotov and Mironenko, 2008). Heating and re-mobilisation of fluids following periodic impact, with minor additional alteration, would be superimposed on this initial major phase.

1.4. Oxygen isotope models

Popular oxygen isotope hydration models postulate simple exchange between a static fluid and anhydrous phases in a closed system (Clayton and Mayeda, 1984), where water:rock (*w/r*) ratios were relatively high (Clayton and Mayeda, 1984, 1999; Leshin et al., 1997). However, there are alternatives to closed-system oxygen models. Briefly, the suggestion is that CC meteorites vary in their mineralogies and oxygen isotope ratios as a result of having come from different regions of a hydrological flow system (Young et al. 1999; Young, 2001a). The Young et al. open-system oxygen model defines a dramatically different conceptual framework for understanding CC oxygen isotopes. However, based on existing data, the jury is still out – open-system and closed-system models are debated, with both types of models apparently capable of reproducing CC oxygen isotopes.

Although the Clayton and Mayeda (1984, 1999) and Leshin et al. (1997) models take static exchange as a starting point, it has recently been suggested that the *w/r* ratios derived from these models (and literature porosity data for CCs) make large-scale fluid flow inevitable (Grimm, 2007). This is based on the assertion (Grimm, 2007) that measured (post-reaction) porosities in the most altered CCs (CI and CM-type carbonaceous chondrites) average only 11-12%. Given estimates for *w/r* ratio (and equivalent pre-reaction porosity) from oxygen isotopes (Clayton and Mayeda, 1984, 1999; Leshin et al., 1997) this would indicate a shortfall in porosity for CCs ie. >1 pore volume of water would be needed to satisfy oxygen isotope constraints, thus requiring flow. There are a number of problems with this argument, the most important being that there is no shortfall in porosity: CI and CM porosities do not cluster over a narrow 11-12% range. CIs have measured porosities of 35% (Consolmagno and Britt, 1998), CMs have porosities up to 28% (Britt and Consolmagno, 2003), and Tagish Lake (an intermediate CI/CM) has a calculated porosity of 41% (Bland et al., 2004). In addition, there is ample evidence for remobilisation of sulphates in the terrestrial environment acting to reduce porosity (Gounelle and Zolensky, 2001). Measured porosities therefore represent minimum pre-terrestrial values. Put simply, there is no shortfall in porosity, so water pore volumes >1 are not required.

1.5. Numerical modelling

In contrast to geochemical, petrographic, and oxygen isotope studies of meteorites, all numerical models of asteroidal aqueous and thermal alteration have predicted large-scale flow of liquid water (Grimm and McSween, 1989; Cohen and Coker, 2000; Coker and Cohen, 2001; Young et al., 1999, 2003; Young 2001a, 2001b, 2004; McSween et al., 2002; Travis and Schubert, 2005; Grimm, 2007). In many cases, fluid transport occurs over 10's km (Grimm and McSween 1989; Young et al. 1999; Coker and Cohen 2001; Travis and Schubert 2005). Travis and Schubert (2005) found that the timing of the onset of convection may vary, but that convective flow was observed in every scenario that was considered, with convection patterns occurring as plumes or broad ridges of upwelling. These authors concluded that convection should occur for a range of porosities, parent body sizes, and radiogenic element concentrations, and that it should last for several Ma. Young et al. (1999) proposed a model for the evolution of carbonaceous chondrite oxygen isotopes based on down-temperature fluid flow within a 50km diameter body. In this situation, flow occurs as a single pass, 'exhalation' flow, driven by internal gas pressure, rather than convective circulation. Young et al. (2003) confine convective flow to bodies >80km diameter, but note that 'exhalation' flow should occur in smaller objects. McSween et al. (2002) observe this type of flow in bodies as small as 18km in diameter. But whether flow is observed as a single pass 'exhalation' (Young et al., 1999), or in convecting cells (Grimm and McSween, 1989; Cohen and Coker, 2000; Coker and Cohen, 2001; Young et al., 1999, 2003; McSween et al., 2002; Travis and Schubert, 2005), the movement of liquid water through rock should fractionate aqueous species. The modelling results appear to directly contradict the meteorite data – the one indicates that alteration was open system, with large-scale flow; the other, that alteration was isochemical, with minimal flow.

It is the goal of this study to resolve these paradoxes: why the most chemically primitive rocks are the most aqueously altered, and why numerical modelling studies indicate large-scale flow, which in general is not predicted by meteorite geochemistry, petrography, and isotopic data.

2. Estimating permeability of asteroid parent bodies

For fluid flow to occur, a rock must be permeable. Permeability is a fundamental control on fluid flow in porous media: it is the property of the media that determines the flow rate which results from an applied pressure gradient. A rock can exhibit permeability at the grain scale, or in fractures, or both. In C1 and C2 chondrites, pervasive, homogeneous alteration at the grain scale clearly indicates that grain scale permeability was important. Initial permeability estimates of 10^{-13}–10^{-11} m^2 for chondritic asteroids (Grimm and McSween, 1989) were based on what were felt to be appropriate terrestrial analogue materials and measurements of lunar soil (Mercer et al., 1975; Lambe and Whitman, 1969; Costes and Mitchell, 1970). This range of permeabilities was adopted in all subsequent numerical modelling studies (Grimm and McSween, 1989; Cohen and Coker, 2000; Coker and Cohen, 2001; Young et al., 1999, 2003; McSween et al., 2002; Travis and Schubert, 2005), but are these values appropriate for CC precursors?

Few measurements of meteorite permeability are available. Sugiura et al. (1984) measured gas permeability for three chondrites, and Corrigan et al. (1997) extended the permeability dataset to include more chondrites. However, it is not clear how subsequent aqueous and thermal alteration, and impact shock, may have modified chondrite permeability away from the primordial value that we are seeking. Therefore, in order to constrain permeability (and therefore flow), what we require is a constraint on permeability in the least altered, most primitive extraterrestrial materials currently available.

It is well known that permeability scales with pore-size, which in turn is related to grain size, sorting, grain shape, grain packing, and the degree of cementation (Dullien, 1992). So to extend our knowledge of meteorite permeability, we have investigated both grain- and pore-size distributions in the most texturally and mineralogically primitive chondrite, and derived a permeability range based on this analysis. The aim is to constrain the relevant physical properties of the anhydrous precursor material, at the point when alteration begins. A similar approach is taken in studies of flow in prorous media in the terrestrial environment. From our perspective, this will essentially define a grain-scale permeability for chondrites. Following that, we will discuss the possibility of fracture permeability.

Chondrites are composed of two principal components: chondrules (spherical igneous inclusions with diameters ranging from 100's μm to mm's), and a fine-grained compositionally primitive matrix. The permeability of the matrix is the key control on fluid flow, because this is the most abundant component in aqueously altered CCs (on average, matrix in CMs accounts for >70% of the rock (McSween, 1979)), and water would initially have been present in matrix pore spaces, or as ice rinds on matrix grains.

Despite the complexity of the relationship between permeability, pore size, grain size and other textural properties, we can place a quantitative constraint on permeability using the simple Carman–Kozeny equation, in which it is assumed that the porous medium can be modelled as a series of equivalent conduits with a constant cross-sectional area of complex shape. Permeability is then given by:

$$k = \frac{\varepsilon r_H^2}{h_{ck}} \qquad (1)$$

where ε is the porosity, r_H is the hydraulic radius of the equivalent conduits and h_{ck} is a constant which depends upon the pore geometry, and which has been found experimentally to lie in the range $4.5 < h_{ck} < 5.5$ for unconsolidated porous media (Dullien, 1992). The Carman–Kozeny equation successfully describes the permeability of many porous media, particularly when there is a narrow range of grain-size and the grains are close to spherical (Figure 3). For some consolidated porous media, the value of h_{ck} may be considerably larger, with values up to 150 required to fit measured data (Dullien 1992).

The hydraulic radius r_H can be written as:

$$r_H = \frac{\varepsilon}{S(1-\varepsilon)} \qquad (2)$$

where S is the specific surface area of the porous medium. Defining d to be the diameter of the sphere which has the same specific surface area yields:

$$k = \frac{\varepsilon^3 d^2}{36 h_{ck} (1-\varepsilon)^2} \qquad (3)$$

which is a more familiar form of equation (1). The diameter d is often associated with grain-size, although this is correct only for packed beds of spheres.

We estimate permeability based on the hydraulic radius determined from measured pore-size distributions and equation (1). The relationship between porosity and permeability is illustrated graphically, for a range of grain sizes in Figure 2 (in this case an intermediate h_{ck} value of 50 was chosen).

In summary, although the individual accretionary setting for an asteroid varied, the fundamental physical properties of matrix (grain size, sorting, grain shape etc.) may have remained relatively constant. Defining those properties, and determining their significance for permeability and fluid flow, are our initial goals. We use a combination of nuclear magnetic resonance cryoporometry and transmission electron microscopy to constrain pore- and grain-size distribution, and from that, permeability for a chondritic precursor using the Carmen-Kozeny equation. Finally, using these derived permeabilities, we investigate implications for fluid flow using a variety of asteroidal alteration models.

3. Samples and Methodology

In terms of its mineralogy and petrography, Acfer 094 is arguably the most primitive carbonaceous chondrite in existence (Greshake, 1997; Nuth et al., 2005; Bland et al., 2007). It provides a window on what chondritic materials may have looked like prior to aqueous and thermal alteration. We have studied the matrix of this meteorite using transmission electron microscopy (TEM) and nuclear magnetic resonance (NMR) cryoporometry (Mitchell et al., 2008). TEM analysis was performed on two focussed ion beam (FIB) lift-outs of portions of Acfer 094 matrix.

3.1. Cryoporometry

Measurement of pore-size distributions by NMR cryoporometry (for details see Mitchell et al., 2008) were performed at the University of Kent. A probe liquid was imbibed into the sample and NMR transverse (T_2) relaxation was used to monitor the phase changes as a function of temperature, by studying changes in the dynamics of the liquid. This information was then interpreted by application of the Gibbs-Thomson equation (constant-pressure analogue of the Kelvin equation), whereby melting point is depressed inversely with pore diameter.

3.2. Microscopy

FIB sample preparation and TEM observations were performed at both Imperial College London and the University of Glasgow. At Glasgow, an ‘in situ’ FIB method was used: the foil was cut from the thin section using a FEI Nova 200 Dualbeam FIB instrument. It was then welded on to a Cu support and diffraction-contrast images were acquired using a FEI T20 TEM operated at 20 kV. At Imperial College London, an 'ex-situ’ preparation method was used. The TEM section was prepared from the solid in the vacuum. It was transferred outside the vacuum onto a TEM Cu grid covered with a thin carbon support film. Analysis was performed using a JEOL JEM-2010 microscope operated at an accelerating voltage of 200 kV. An ISIS EDS system by Oxford Instruments was employed to analyse the chemical composition of the samples and electron diffraction patterns used to identify amorphous and crystalline phases. Importantly for this work, there is no differential thinning in FIB preparation, so we can be confident that any porosity is primary, and not enlarged by ion bombardment (as commonly occurs with 'normal' Ar ion milling for TEM sample preparation).

3.3. Numerical modelling

We use a number of asteroidal alteration models (e.g. Grimm, 2007; Young et al., 2003; Cohen and Coker 2000). Updated input parameters for Cohen and Coker (2000) (in addition to those discussed in the text) include more recent evaluations for the vapour pressure over ice, the thermal conductivity of ice, the thermal conductivity, viscosity, and surface tension of liquid water, the heat capacity of minerals, the density of minerals and bulk rock, and the thermal expansion coefficient and bulk modulus of minerals. We also include dehydration (not relevant to models presented here), a porosity dependent permeability (as discussed here), and the instantaneous venting of water vapour when the vapour pressure exceeds the lithostatic pressure.

4. Results

4.1. Grain- and pore-size distribution

It is apparent from our own TEM analysis, and previous work (Nuth et al., 2005), that there are abundant grains in the 50-100 nm size range (Figure 3) – over the total area of our FIB sections (~110 μm^2) we observe only a single grain of ~1 µm in size. Defining a lower limit on grain-size is difficult, since the smallest grains have a diameter much less than the thickness of a FIB section, but a size range of 20-200 nm would appear to be appropriate for the bulk of Acfer 094 matrix grains. This is consistent with the grain-size distribution in IDPs (Rietmeijer, 1993). Even CO chondrites, which have experienced varying degrees of thermal metamorphism, can exhibit similar root-mean-square (rms) matrix grain-sizes (e.g. Kainsaz has an average rms grain-size of 249 nm (Brearley, 1996)).

Cryoporometry results are consistent with the TEM data. Figure 4 shows a pore-volume distribution for Acfer 094 derived using NMR cryoporometry. The majority of the porosity in Acfer 094 is at extremely low pore diameters. From 200 nm down to the lowest pore diameter currently measurable by this technique in these meteorites (~20 nm), the pore volume distribution averages 3 ± 1 $\mu l.nm^{-1}.g^{-1}$, with no sign of a drop-off at low pore diameters. Estimating the mean pore-size is difficult, as the smallest pores in Acfer 094 are below the resolution of our measurements. However, a geometric mean pore-size in the range 10–100 nm is reasonable based on the available data.

4.2. Defining permeability for a chondritic precursor

As noted previously, permeability is based on the hydraulic radius, which is derived from measured pore- and grain-size and equation (1). The hydraulic radius is related to the geometric mean pore-size by $r_H = d_P / \xi$, where ξ depends upon the pore shape (Paterson, 1983; Brace, 1977). For circular pores $\xi = 4$, while for slot-shaped pores of high aspect ratio $\xi = 2$. Using a geometric mean pore-size of order 10–100 nm, and assuming a value of $\xi = 4$, yields estimates for hydraulic radius of order 2.5-25 nm (consistent with grain sizes in the range 20-200 nm), and a corresponding permeability of order 10^{-19}–10^{-17} m^2 (0.1–10 μD) for a porosity of 40% (eqn 1; Figure 1; Figure 2). A porosity of 40% is consistent with *w/r* ratios derived from oxygen isotope studies (Clayton and Mayeda, 1984, 1999; Leshin et al., 1997). Lower porosity estimates yield lower values of estimated permeability, but the principle control on permeability is the hydraulic radius, and hence the mean pore- and grain-size (eqn. 1).

Also shown in Figure 3 are permeability data for siltstones and claystones, which are terrestrial analogues with comparable pore-sizes to meteorite samples. These lithologies exhibit permeabilities of order 10^{-21}–10^{-19} m^2 (1-100 nD; see Figure 1), and act as barriers to flow over millions of years. A permeability of 10^{-19}–10^{-17} m^2 for a chondritic precursor is six orders-of-magnitude lower than that used in earlier numerical modelling studies. As noted previously, the grain-sizes observed in Acfer 094 matrix are consistent with literature data, both for this meteorite, and for other primitive extraterrestrial materials (e.g. chondrites, IDPs, Stardust sample-return etc). Since grain-size is a fundamental control on permeability, the observation of consistently small grain-sizes (10's – 100's nm) in the most primordial materials supports the contention that permeability in chondritic precursors was very low.

4.3. Modelling fluid flow

Using a gravity flow model (Grimm, 2007) as a starting point, and a permeability range of 10^{-19}–10^{-17} m^2, we can derive approximate lengthscales for liquid water transport as a function of distance from the centre of a canonical asteroid (Figure 5). Given alteration timescales of order 1Ma, and standard material properties (Young et al., 2003), this model predicts buoyancy-driven water transport of 10's to 100's μm – consistent with petrographic studies (Krot et al., 1995; Grossman et al., 2002), and indicating essentially isochemical alteration (Figure 5). Moreover, we can take a similar approach to that of Young et al. (2003) to investigate the extent to which convective flow could occur, given these new permeability values. Young et al. (2003) employed the standard 10^{-13}–10^{-11} m^2 permeability range, and showed that convection would have occurred in bodies >80km in diameter (Figure 1 from Young et al. (2003)). However, if we take the permeability range of 10^{-19}–10^{-17} m^2 derived from our chondrite data, then even for bodies with 40% porosity, and canonical $^{26}Al/^{27}Al$, it is apparent that much larger objects are required before large-scale convective water flow could be established (Figure 6): in this case, asteroid diameters above ~440km.

In addition, we have taken a detailed numerical model of asteroidal alteration (Cohen and Coker, 2000), and modified the input parameters, based (in part) on the above analysis. The scenario shown in Figure 7 is for a 10 km diameter asteroid accreting at 3 AU, 1.25 Myrs after CAI formation. The figure illustrates peak temperature as a function of radius. In this model the asteroid composition is 20% forsterite, 15% enstatite, 40% water ice, initial voids of less than 1%, and the remainder inert rock. Previously, using higher permeabilities, flow occurred over several km's, and temperatures in most scenarios substantially exceeded those expected for CC alteration (Cohen and Coker, 2000). Using these updated input parameters, the same model (Cohen and Coker, 2000) shows essentially zero fluid flow (less than a single cell size – consistent with the predictions of the gravity (Grimm, 2007) and convection (Young et al., 2003) models when updated input parameters are used (e.g. Figure 5)). In addition, we observe moderated peak temperatures throughout the centre of the asteroid: while still high, temperatures are closer to those expected from oxygen isotope studies of CC alteration (Clayton and Mayeda, 1984, 1999; Leshin et al., 1997). We are currently exploring the additional effect of varying parameters such as composition, size, timing of accretion, percentage void space, and surface boundary temperature, on peak internal aqueous alteration temperatures.

5. Discussion

5.1 Model permeability estimates compared to literature data

Thermal, aqueous, and impact processing of meteorites can all act to modify porosity (and potentially permeability), as can atmospheric entry and storage in the terrestrial environment. With those caveats in mind, how do our permeability estimates for precursor materials compare with measurements from carbonaceous chondrites? Sugiura et al. (1984) measured gas permeabilities for three chondrites. One of these was a sample of Allende, which was found to have a permeability of c. 10^{-16} m^2. Another recorded a permeability of c. 10^{-15} m^2 (1 mD; 1 D = 9.87×10^{-13} m^2); the permeability of a third was too small to detect ($<10^{-21}$ m^2 or 1 nD). Unfortunately no description of the pore-space morphology of the samples was provided. Corrigan et al. (1997) significantly expanded the permeability dataset for chondrites, also measuring gas permeabilities. The average permeability for all the meteorites analysed was 8.3×10^{-17} m^2. CM chondrites – included in this average – proved to be an analytical challenge due to the presence of cracks caused by desiccation while on Earth (Corrigan et al., 1997 and pers. comm..), occasionally resulting in relatively high permeabilitites. For the most mineralogically primitive meteorites in their suite (samples of Vigarano, Efremovka, Leoville – all reduced CV3s), Corrigan et al. (1997) found an average permeability of 2.5×10^{-18} m^2. Both the Sugiura et al. (1984) and Corrigan et al. (1997) measurements likely represent an upper limit for liquid water permeability, as the experiments were in the low pressure, molecular gas flow domain. It is therefore encouraging that these data (particularly the permeabilities from the least aqueously and thermally altered, most mineralogically primitive samples) appear to be in excellent agreement with our estimated permeabilities of 10^{-19}–10^{-17} m^2 for a chondritic precursor material.

5.2. Constraining fracture permeability

We have constrained permeability based on the material properties (grain size, porosity, pore-size distribution etc) of primitive meteorites. This is essentially a grain-scale permeability, taking the rock as a homogeneous whole. As C1 and C2 chondrites are distinguished by pervasive, homogeneous alteration of matrix at the grain-scale, the implication is that grain-scale permeability was important. But what constraints can we impose on the relative importance of fracture permeability?

In studying meteorites we are hampered by the obvious fact that we have hand samples – we cannot go to the outcrop. Therefore, any discussion of the possible contribution of fracture, or microfracture permeability is unlikely to lead to a definitive conclusion. However, it is possible to impose some constraints. The nearest meteoriticists can get to an outcrop-scale is in large CC falls. In the case of the CMs, the largest fall is Murchison (>100kg). There are large numbers of hand specimens of this meteorite, but at the hand specimen scale, abundant alteration-filled fractures are not observed (Figure 8a). At finer scales, veins filled by secondary minerals have occasionally been described. However, these features appear to be exceedingly rare. Figure 8b shows an energy dispersive element map for a section of Murchison – alteration phases dominate matrix, but they are not associated with veins. More detailed studies have come to a similar conclusion: veins appear to be rare or absent in CM chondrites (Benedix et al. 2003; Tyra et al. 2009). The same is true for CI chondrites. Carbonate and sulphate veins in Orgueil were once considered to be evidence for aqueous flow. However, it is now clear that these features are a product of terrestrial alteration (Gounelle and Zolensky, 2001).

The work of Corrigan et al. (1997) is also relevant here. The samples measured for permeability were all relatively large hand samples, ranging in size from ~3 cm upto ~15 cm in some cases (Corrigan pers. comm.). Despite this, low permeabilities – consistent with the grains-scale permeability derived above – were the norm. To summarise the observational data, there does not appear to be evidence for pervasive interconnected fractures at scales ranging from 10’s-μm to 10’s-cm in CCs, either from studies of chondrite petrography, or permeability measurements.

Finally, the pervasive, homogeneous nature of alteration in C1 and C2 chondrites is actually quite remarkable, and places its own constraint on the importance of fractures as focussed conduits for flow. For the CM2s this homogeneity has recently been quantified through X-ray diffraction measurements of modal mineralogy (Howard et al., 2009). Even including meteorites that are considered highly altered (e.g. Nogoya and Cold Bokkeveld), modal total phyllosilicate only varied by a few % amongst CM2 falls (the total range was

73–79%). If flow had occurred through focussed conduits, it would not result in homogeneous alteration of matrix. Rather, alteration would be concentrated around fractures. In addition, significant interchange of fluid between the fracture network and enclosed blocks would be severely limited by the low permeability of matrix – once again, grain-scale permeability imposes a fundamental constraint. As we have shown, grain-scale permeability in CCs is low, so transport lengthscales are short (100's μm at most, even given alteration timescales of 1Ma). To produce uniform wholesale alteration of the bulk rock, the density of microfractures would need to be of this order. The only remaining possibility for significant fracture permeability would be a much larger scale fracture network, effectively decoupled from individual meteorite-sized blocks. This is not excluded by our data, however, we note that (by definition) it is not testable through meteorite sample analysis. In addition, it does not appear to be required (in terms of stabilising parent body temperatures) based on numerical modelling (Figure 7).

5.3. Implications for fluid flow

The permeability estimates that we derive for a chondritic precursor are in agreement with permeability measurements from mineralogically primitive chondrites (Corrigan et al. 1997). Given that we do not see compelling evidence for a pervasive interconnected fracture network at length scales that would be effective at producing homogeneous alteration of CCs, our conclusion is that grain-scale permeability is dominant, and that permeability values in the range 10^{-19}–10^{-17} m^2 for a porosity of 40% are appropriate for chondritic asteroids at the onset of aqueous alteration. Applying these permeability estimates in numerical models, we find buoyancy-driven water transport over distances of 100's μm at most, and similar short distances for element mobility via diffusion. We note that these predicted flow / transport distances are in excellent agreement with distances for metasomatism from petrographic observations of chondrites. Fundamentally, isochemical alteration, with minimal fluid flow, is not a special circumstance. It is unavoidable, once we consider the material properties of these rocks. The extreme fine grain size of chondritic matrix results in very low permeability, which results in minimal flow, even where *w/r* ratios were high. Clearly this imposes constraints on a number of existing models.

5.4. Why are the most chemically primitive rocks also the most altered?

Fine-grained condensates would have hosted volatile elements, including water. Assuming that water was initially present as ice rinds on matrix grains, then water-saturated porosity in the bulk rock would scale with matrix abundance. The most primitive rocks are 100% fine-grained matrix. They will be the most altered, but permeabilities are such that fluid flow would be minimal, even when calculated *w/r* ratios are high (Clayton and Mayeda, 1984, 1999; Leshin et al., 1997). The greater the proportion of fine-grained material in a chondrite, the closer it is to a primitive 'solar' composition, but also the more water was available for alteration.

6. Conclusions

We have studied the pore- and grain-size distribution in a carbonaceous chondrite that has experienced minimal thermal and aqueous alteration. Both datasets are consistent, indicating a geometric mean pore-size of 5–50 nm, a hydraulic radius of order 2-20 nm (consistent with grain sizes in the range 20-200 nm), and a permeability in the range 10^{-19}–10^{-17} m^2 (for a porosity of 40%). We note these grain sizes are also consistent with previous studies of other primitive extraterrestrial materials (primitive chondrites, IDPs, and Stardust sample-return materials), and that our estimated permeability range is consistent with measurements from primitive chondrites (Corrigan et al., 1997). Assuming these new permeability values are representative of a ‘canonical’ chondritic precursor material, our modelling shows that, even at the upper permeability bound (10^{-17} m^2, for matrix grain sizes of order 100’s nm), convective flow would not occur unless the asteroid diameter exceeded ~440km. In addition, we show that lengthscales for liquid water transport are in the range 10’s - 100’s µm (even with alteration timescales of order 1Ma), indicating essentially isochemical alteration, and consistent with meteorite petrography, geochemistry, and a closed-system oxygen isotope model (e.g. Clayton and Mayeda, 1984, 1999; Leshin et al., 1997).

In summary, our analysis resolves the paradox of how primitive chemistry can co-exist with altered mineralogy; how we can have oxygen isotopic evidence for high *w/r* ratios despite minimal evidence for flow; why previous numerical modelling studies have produced results that appear to be at odds with meteorite chemistry and petrography; and why the most primitive rocks are also the most altered.

Acknowledgements

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Figure captions

Fig. 1. Permeability / porosity as a function of hydraulic radius. Data are shown for a variety of terrestrial porous media (Doyen, 1988; Bourbie and Zinszner, 1985; Paterson, 1983; Mortensen et al., 1998; Hildenbrand et al., 2002), and predicted using the Carman-Kozeny equation (1). The solid line denotes the predicted permeability for a value of h_{ck} = 5, appropriate for unconsolidated and some consolidated porous media. The dashed line denotes the predicted permeability for a value of h_{ck} = 100, appropriate for some consolidated sandstones and siltstones. The shaded region denotes the estimated hydraulic radius for meteorite samples.

Fig. 2. Permeability vs. porosity for a range of grain sizes, and a value of h_{ck} = 50 (intermediate between some unconsolidated media and consolidated siltstone (Figure 1)). This figure illustrates graphically the relationship between permeability, porosity, pore geometry, and grain size defined in eqn. (3).

Fig. 3. A selection of transmission electron microscopy (TEM) images showing the matrix of Acfer 094. TEM analysis followed focussed ion beam lift-out of two portions of fine-grained matrix material from the carbonaceous chondrite Acfer 094. FIB sections covered a total area of ~110 μm^2. A variety of matrix grains can be distinguished (in these images, coherent features which have a relatively constant grey-scale), set against a background of amorphous and finer-grained material. The great majority of grains are smaller than 200 nm in diameter.

Fig. 4. Nuclear magnetic resonance cryoporometry data. Pore volume distribution vs pore diameter for the carbonaceous chondrite Acfer 094. The data are consistent with the TEM results, and indicate that the bulk of the porosity in Acfer 094 is at very low pore diameters.

Fig. 5. Modelling asteroidal alteration. Transport lengthscale as a function of distance from a model asteroid centre. Results for permeabilities (k) of 10^{-17} m^2 and 10^{-19} m^2 are shown, using a gravity flow model (Grimm, 2007), standard material properties (Young et al., 2003), a porosity of 40%, and assuming an alteration timescale of 1Ma. The shaded region denotes the approximate lengthscale for metasomatism observed in altered carbonaceous chondrites, derived from a range of petrographic studies.

Fig. 6. Modelling asteroidal alteration. The range of critical radii for the onset of convective flow, for a spherical asteroid with 40%, given heat production from radionuclide decay (the 'canonical' initial Solar System $^{26}Al/^{27}Al$ value is taken to be ~5×10^{-5}). Contours are for permeabilities of 10^{-19} and 10^{-17} m^2, derived from our analyses of the primitive chondrite Acfer 094. It is apparent that, even at the upper permeability bound of 10^{-17} m^2, objects with diameters in excess of 440km are required before large-scale convective water flow could be established.

Fig. 7. Modelling asteroidal alteration. Temperature as a function of distance from a model asteroid centre. Results are shown for permeabilities based on eqn. 3, using a 1000-cell Cohen and Coker (2000) type model, and including the effects of gas venting. The heat source is radionuclide decay, and the model uses a Cassen-based time-dependent surface temperature boundary condition (Cassen, 1994). Additional details can be found in Cohen and Coker (2000).

Fig. 8. Murchison CM2 chondrite, in hand specimen and thin section. (a) A hand specimen, ~10cm across (courtesy Linda Welzenbach. Smithsonian Institution). (b) Energy dispersive element map of a thin section of Murchison. These images illustrate the homogeneous nature of alteration in CM chondrites. Neither here, nor in the literature do we find evidence for a pervasive fracture network.

Figure 1

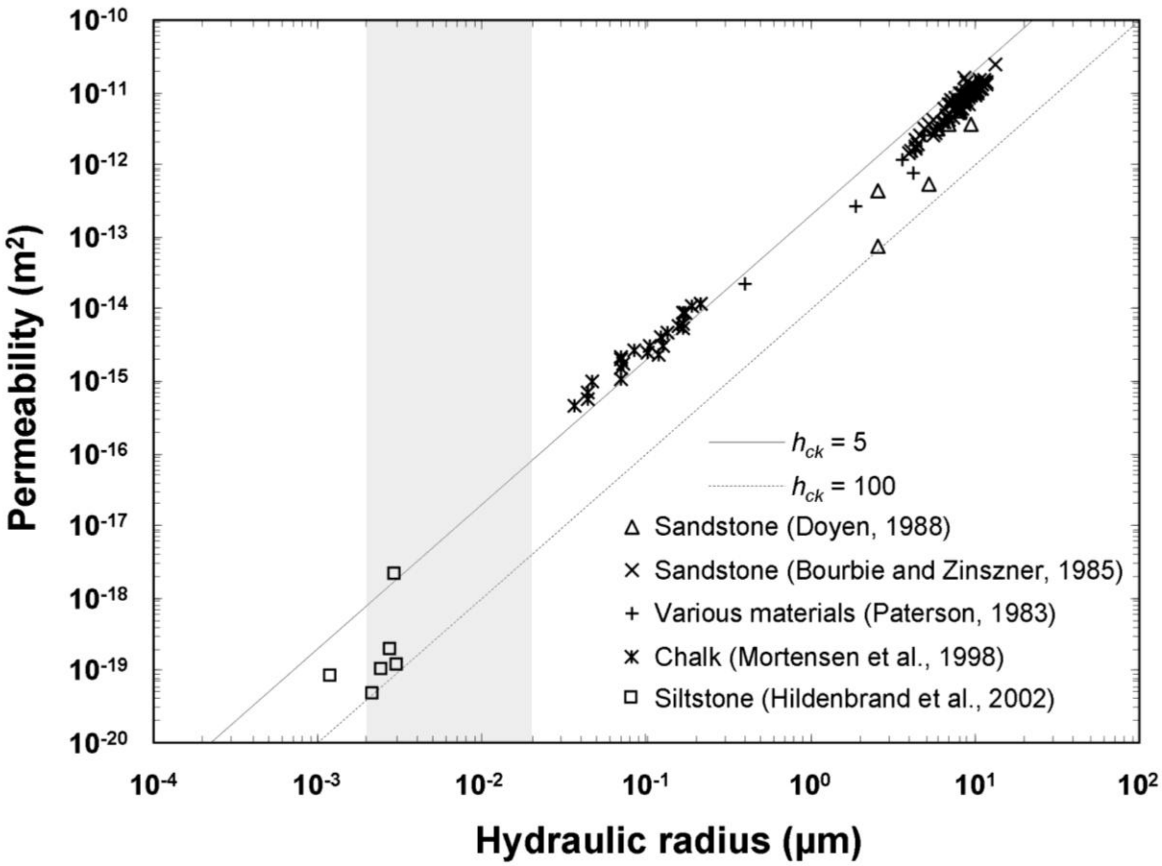

Permeability (m²)
Hydraulic radius (μm)
10^-10
10^-11
10^-12
10^-13
10^-14
10^-15
10^-16
10^-17
10^-18
10^-19
10^-20
10^-4
10^-3
10^-2
10^-1
10^0
10^1
10^2
h_ck = 5
h_ck = 100
Sandstone (Doyen, 1988)
Sandstone (Bourbie and Zinszner, 1985)
Various materials (Paterson, 1983)
Chalk (Mortensen et al., 1998)
Siltstone (Hildenbrand et al., 2002)

Figure 2

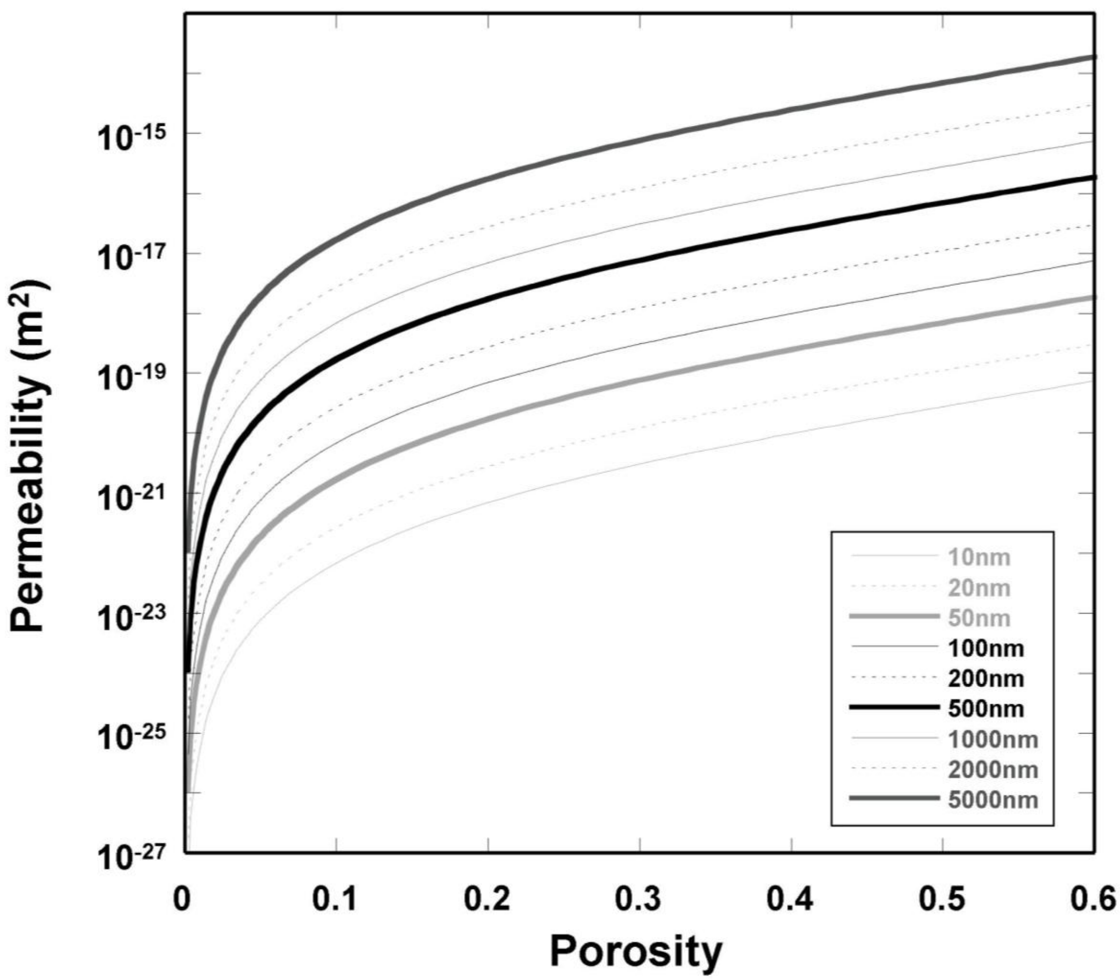

Permeability (m²)
10-15
10-17
10-19
10-21
10-23
10-25
10-27
0
0.1
0.2
0.3
0.4
0.5
0.6
Porosity
10nm
20nm
50nm
100nm
200nm
500nm
1000nm
2000nm
5000nm

Figure 3

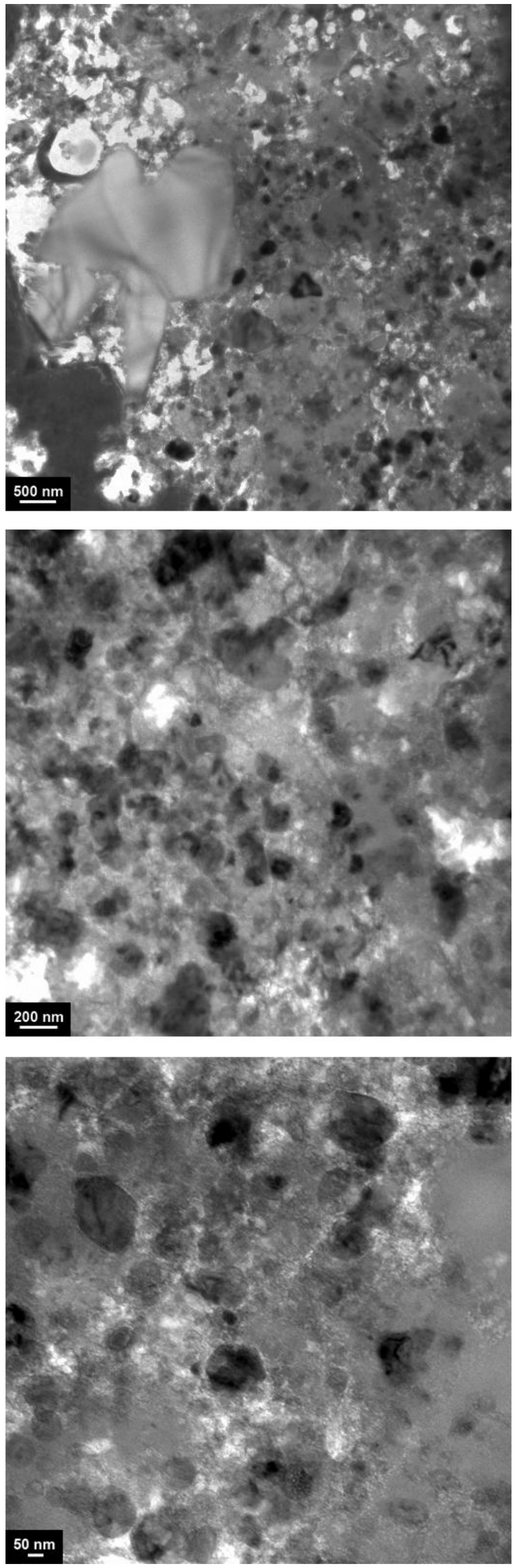


Figure 4

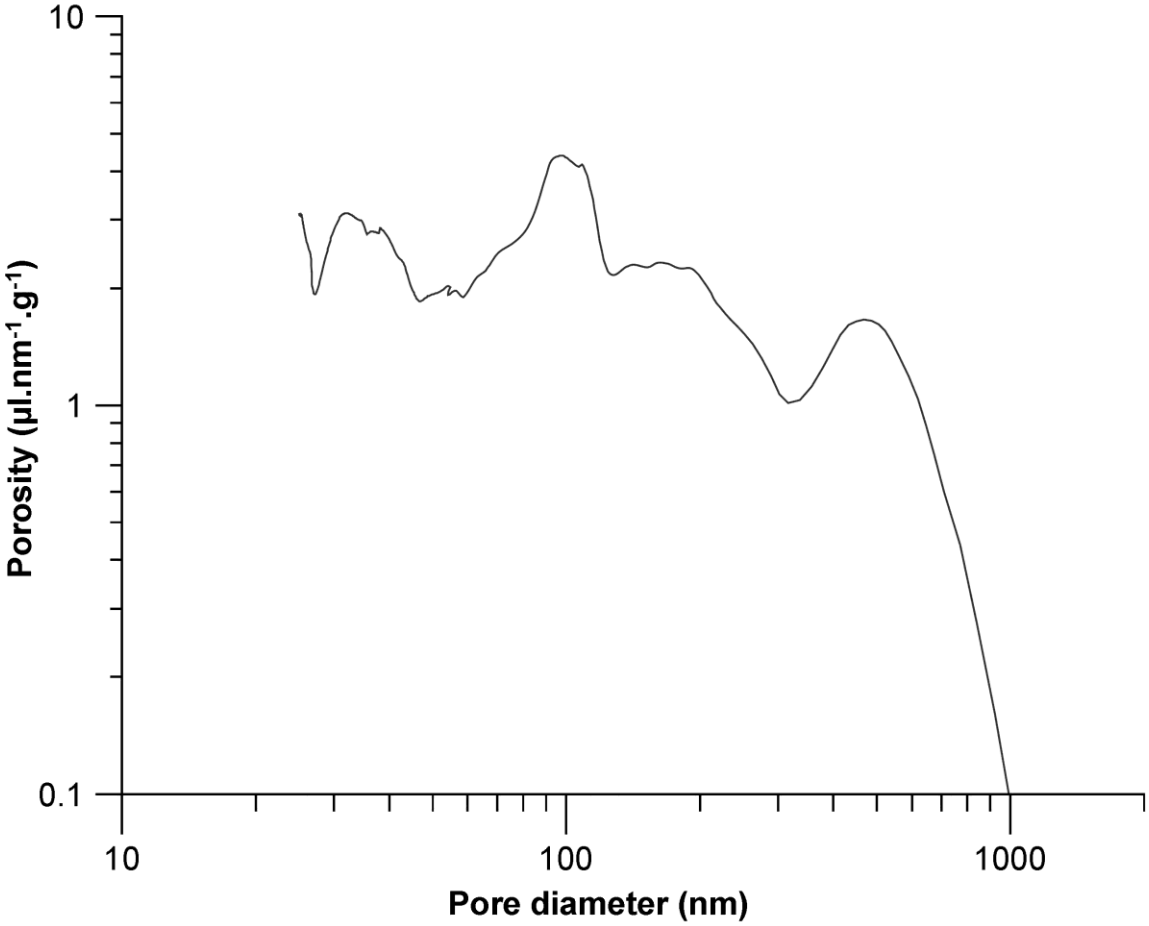
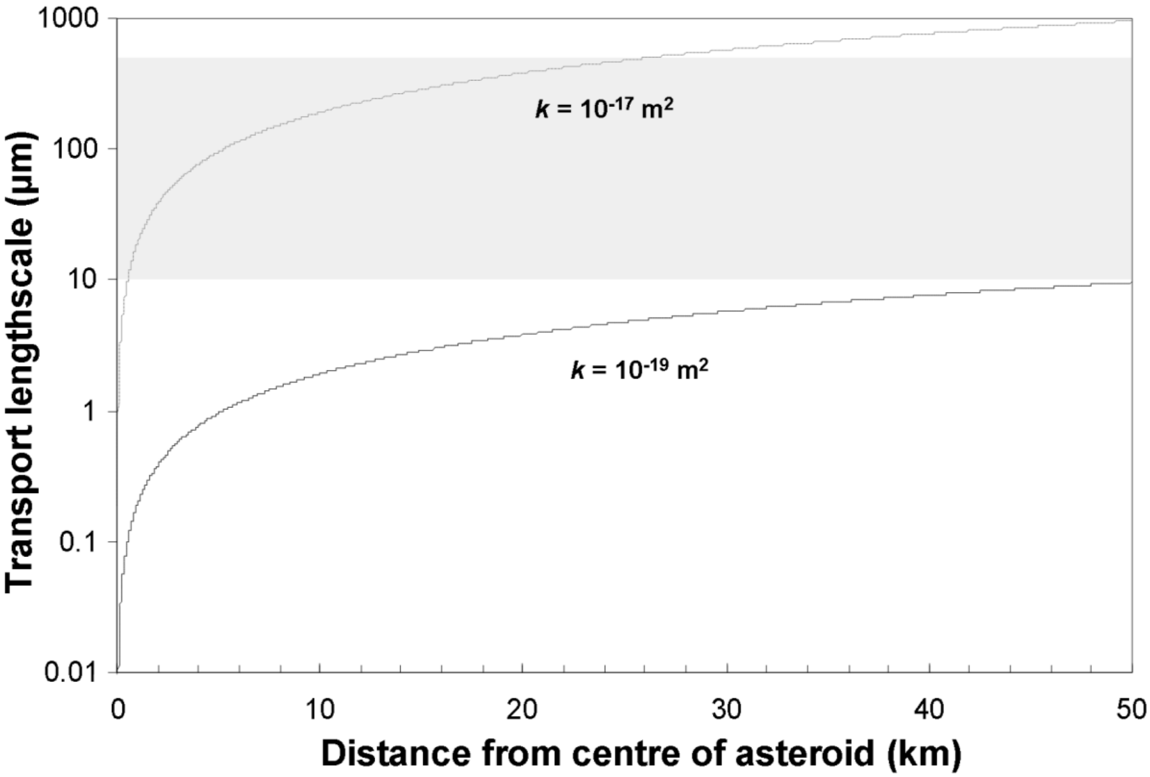
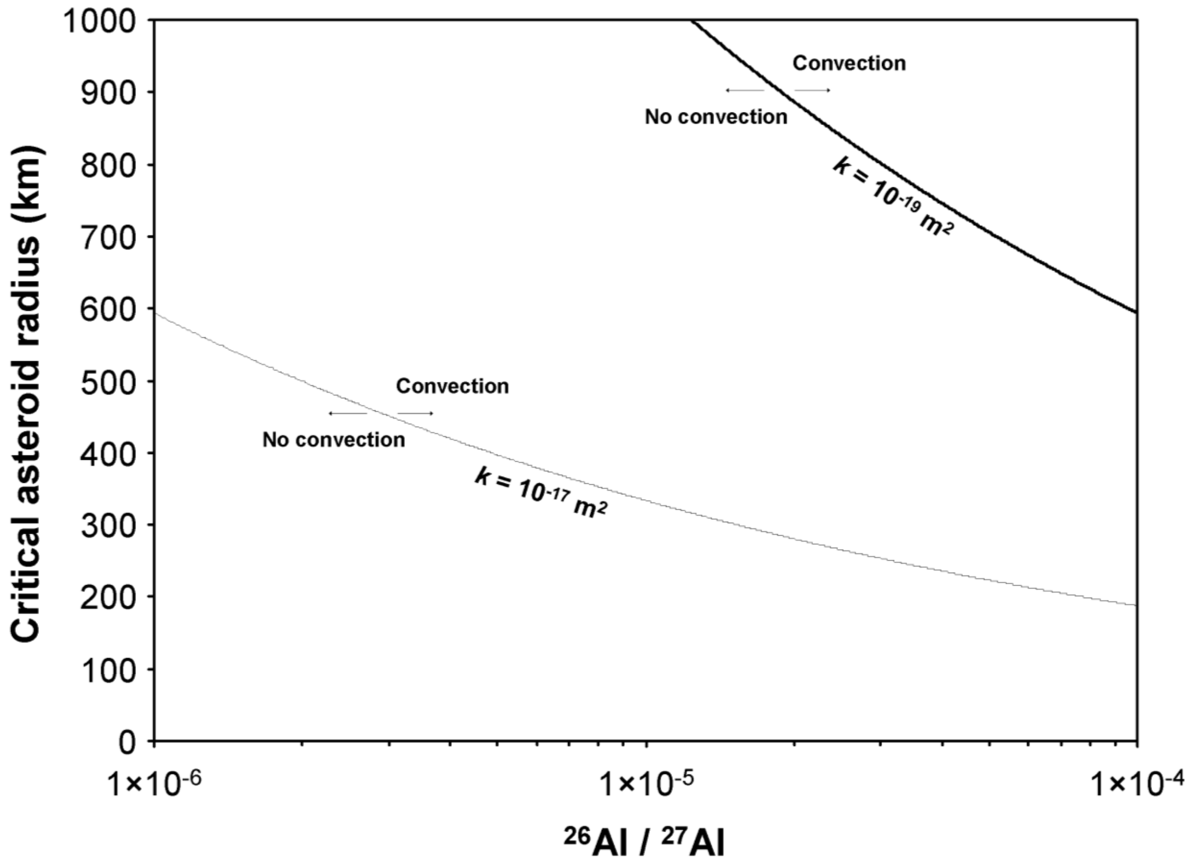


Figure 5



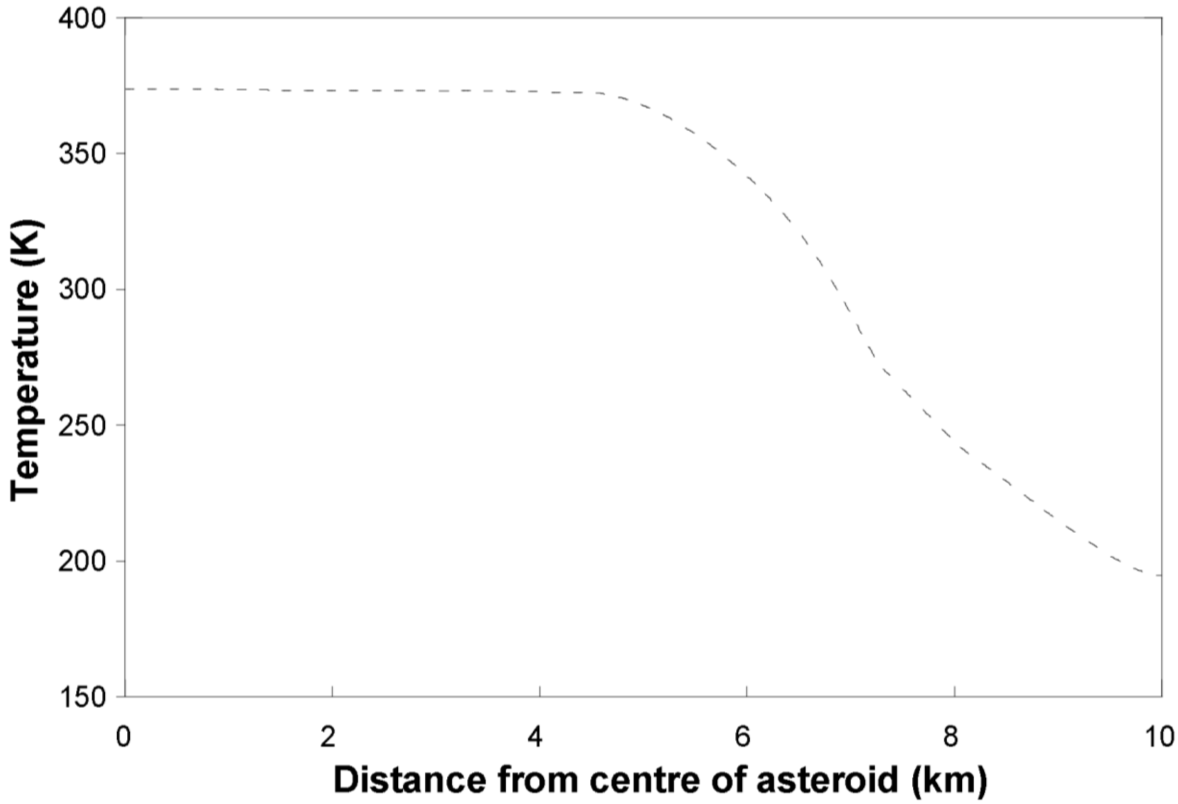
1000
100
10
1
0.1
0.01
k = 10-17 m2
k = 10-19 m2
Transport lengthscale (µm)
0
10
20
30
40
50
Distance from centre of asteroid (km)

Figure 6



1000
900
800
700
600
500
400
300
200
100
0
Critical asteroid radius (km)
1×10-6
1×10-5
1×10-4
26Al / 27Al
Convection
No convection
k = 10-19 m2
Convection
No convection
k = 10-17 m2

Figure 7



400
350
300
250
200
150
Temperature (K)
0
2
4
6
8
10
Distance from centre of asteroid (km)

1 **Figure 8**

2