ISOTOPIC STUDIES ON EVOLUTION OF EARLY SOLAR SYSTEM OBJECTS BY AN ION MICROPROBE

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CERTIFICATE

I hereby declare that the work presented in this thesis is original and has not formed the basis for the award of any degree or diploma by any University or Institution.

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Certified by:

Prof. J. N. Goswami (Thesis Supervisor) Director Physical Research Laboratory, Navrangpura Ahmedabad 380 009, INDIA Dedicated to my parents....

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CHAPTER 1

Introduction

The evolution of the solar system started with the gravitational collapse of a dense interstellar molecular cloud about 4.6 billion years ago that led to the formation of the proto-Sun at the center, surrounded by a rotating cloud of gas and dust, the so called solar nebula. The solar nebula is expected to be extremely hot initially due to the very high activity of the proto-Sun that released tremendous amount of energy into its surroundings. This is also corroborated by the presence of rare refractory objects in chondritic meteorites, termed Ca-Al-rich Inclusions (CAIs), that are considered to be the first solids to form in the solar nebula. Many of the CAIs have characteristics that suggest them to be direct nebular condensates, while others have undergone episodes of melting and recrystallization at high temperature (MacPherson et al., 2003). Chondrules, small (0.1 to a few mm-sized) spheroidal silicate objects, that are abundant in chondritic meteorites are considered to be products of high temperature transient events in the solar nebula that led to melting of pre-existing solids followed by rapid cooling resulting in their formation. In comparison to the CAIs, that are mostly present in the carbonaceous chondrites, chondrules are ubiquitous in all chondritic meteorites and constitute up to 80% by volume in unequilibrated

ordinary chondrites (UOCs). Formation of chondrules thus represents a major event in the early history of the solar system. However, several important questions such as the textural and chronological relations between CAIs and chondrules, the time and duration over which chondrule formation persisted in the solar nebula and the exact mode of formation of chondrules, particularly the source of transient heating event, are not well understood at present. Understanding the processes and timescales of chondrule formation, and its relation to CAIs can significantly advance our understanding of the solar nebula processes.

Efforts have been made by several groups to understand the time and duration of chondrule formation using the now-extinct short-lived radionuclide ²⁶Al (half life=0.72 Ma) as a chronometer. ²⁶Al decays to ²⁶Mg and the former presence of ²⁶Al in early solar system objects (e.g. CAIs) is inferred from observed excess in the abundance of ²⁶Mg in them (Lee et al., 1976, 1977). It may be noted that the inferred chronological information is valid only if we assume that these objects formed in a reservoir in which ²⁶Al was homogeneously distributed with a well defined initial ²⁶Al/²⁶Al ratio that decreased with time owing to decay of ²⁶Al. A detail analysis of ²⁶Al records in CAIs as well as time-scales inferred from studies of ²⁶Al and other now-extinct short-lived radionuclides in early solar system objects support this assumption (MacPherson et al., 1995; Halliday and Kleine, 2006; Thrane et al., 2006).

The initial Al-Mg isotope studies of meteoritic chondrules were conducted in Al-rich chondrules that contain phases with high Al/Mg ratios suitable for such a study. Most of the chondrules, however, are ferromagnesian in nature and the rare Al-rich chondrules may not be representative of chondrules in general. Several studies of ferromagnesian UOC chondrules have been carried out and at present the duration of formation of chondrules inferred from these studies varies from ~1 Ma to 4 Ma (Russell et al., 1996; Kita et al., 2000, 2005a; Huss et al., 2001; Mostefaoui et al., 2002). A time interval of 1 - 2 Ma has also been suggested

between formation of CAIs and UOC chondrules. Similar inferences have also been drawn from studies of plagioclase-rich chondrules in carbonaceous chondrites (Sheng et al., 1991; Hutcheon and Jones, 1995; Krot et al., 1999; Srinivasan et al., 2000; Hutcheon et al., 2000; Marhas et al., 2000; Amelin et al., 2002; Kunihiro et al., 2004; Kurahashi et al., 2004). The choice of appropriate meteorite samples for the study of Al-Mg isotope systematics in chondrule is also important. It is now well established that ²⁶Al was a major heat source in the early solar system (Urey, 1955; Srinivasan et al., 1999). ²⁶Al along with the now-extinct radionuclide ⁶⁰Fe (half life=1.5 Ma; Shukolyukov and Lugmair, 1993; Tachibana and Huss, 2003; Mostefaoui et al., 2005; Tachibana et al., 2006) are considered as responsible for early malting of

well established that ²⁶Al was a major heat source in the early solar system (Urey, 1955; Srinivasan et al., 1999). 26 Al along with the now-extinct radionuclide 60 Fe (half life=1.5 Ma; Shukolyukov and Lugmair, 1993; Tachibana and Huss, 2003; Mostefaoui et al., 2005; Tachibana et al., 2006) are considered as responsible for early melting of planetesimals as well as thermal metamorphism in meteorite parent bodies . The possibility of thermal metamorphism affecting the Al-Mg isotope systematics in chondrules from different UOCs that have experienced a range of thermal histories, cannot be ruled out. In general, UOCs of low petrologic grades that have undergone low degree of thermal metamorphism are considered to be suitable sample for such a study. In this study, I have carried out petrographic and Al-Mg isotope studies of a set of chondrules from nine UOCs of low petrologic grades to enhance our understanding of the time of onset and duration of chondrules formation in the early solar system. The possibility of thermal metamorphism in chondrules from UOCs belonging to low petrologic grades is also examined. Data obtained in this study are combined with the existing Al-Mg isotope data for CAIs and chondrules to infer the life time of an active solar nebula during which CAIs and chondrules have formed. In this chapter, I briefly describe the now-extinct short-lived radionuclides present in the early solar system, the nature of the early formed solar system objects, the use of ²⁶Al to obtain chronological information, the present status of studies related to chronology of chondrule formation and the role of ²⁶Al as a heat source during the early evolution of the planetesimal. Finally, I present a summary of the main objectives of this thesis work.

1.1 Short-lived radionuclides in the early solar system

The presence of the now-extinct short-lived radionuclides in early solar system objects plays an important role in our understanding of the origin and early evolution of the solar system. The discovery of ¹²⁹Xe excess in Richardton meteorite in 1960 (Jeffery and Reynold, 1961) was ascribed to the presence of the short-lived radionuclide ¹²⁹I (half life=15.7Ma) in the early solar system that was incorporated "live" into this meteorite and decayed in-situ resulting in the observed excess of ¹²⁹Xe. Since then, studies of meteorites have provided evidence for the presence of several other now-extinct radionuclides, such as ⁴¹Ca, ³⁶Cl, ²⁶Al, ⁶⁰Fe, ¹⁰Be, ⁵³Mn, ¹⁰⁷Pd, ¹⁸²Hf, ¹²⁹I, ⁹²Nb, ²⁴⁴Pu, ¹⁴⁶Sm in the early solar system. Although the presence of these nuclides is inferred from the observed excess in their daughter products (e.g. ²⁶Mg in case of ²⁶Al, ⁴¹K in case of ⁴¹Ca, ⁶⁰Ni in case of ⁶⁰Fe, ¹⁰B in case of ¹⁰Be, etc), a proof for in-situ decay of these nuclides in the analyzed samples comes from the demonstration of a correlation of the excess with the abundance of the stable parent elements (e.g. a correlation of excess in ${}^{26}Mg/{}^{24}Mg$ with ${}^{27}Al/{}^{24}Mg$). The half lives of these nuclides vary from 10^5 yrs to 10^8 yrs and are listed in Table 1.1. The preservation of evidence for prior existence of any now-extinct radionuclide requires that the analyzed sample behaved as a close system from the time when these nuclides were still extant and got incorporated into the sample. Study of short-lived radionuclide helps in constraining the time-scales and processes that have shaped the early evolution of the solar system. They serve as excellent time markers with high precision of less than a million year (Ma) for events that occurred 4.6 billion years ago and over timescales of a few Ma to a few tens of Ma. Absolute chronology based on long-lived radionuclides as chronometer cannot provide the precision needed to resolve such time difference, except in U-Pb dating in very favourable cases. Of course unlike the long-lived radionuclides, that provide absolute chronology, the short-lived now-extinct radionuclides can only provide relative chronology between events marking various episodes in the early solar system.

Several sources have been suggested for the presence of the short-lived radionuclides in the early solar system. Irradiation of gas and dust in the solar nebula by energetic particles from an active early sun as well as a stellar source injecting freshly synthesized short-lived radionuclides into the collapsing proto-solar cloud

Ta	ble 1.1: Sho	ort-lived ra	dionuclides in early sol	ar system
Short-lived	Half life	Daughter	Initial abundance*	Analyzed objects
Radionuclide	(Ma)	nuclides	(ref. nuclide)	
⁴¹ Ca	0.1	⁴¹ K	$(1.5 \times 10^{-8}) [^{40}\text{Ca}]^1$	CAIs
³⁶ Cl	0.3	³⁶ Ar	$(1.5 \times 10^{-6}) [^{35}Cl]^2$	CAIs, Chondrules
²⁶ Al	0.72	²⁶ Mg	$(5 \times 10^{-5}) [^{27} \text{Al}]^3$	CAIs, Chondrules,
		U		Achondrites
⁶⁰ Fe	1.5	⁶⁰ Ni	(~0.2-1)×10 ⁻⁶)[⁵⁶ Fe] ⁴	Chondrules, Chon-
				drites, Achondrites
¹⁰ Be	1.5	^{10}B	$(\sim 1 \times 10^{-4})[^{9}\text{Be}]^{5}$	CAIs
⁵³ Mn	3.7	⁵³ Cr	$(\sim 1.4 - 4 \times 10^{-5})[^{55}Mn]^{6}$	CAIs, Chondrules,
				Achondrites
¹⁰⁷ Pd	6.5	^{107}Ag	(~4.5×10 ⁻⁵)[¹⁰⁸ Pd] ⁷	Iron meteorites, Pall-
		Ũ		asites
¹⁸² Hf	9	^{182}W	$(10^{-4})[^{180}\text{Hf}]^8$	Achondrites
¹²⁹ I	15.7	¹²⁹ Xe	$(10^{-4})[^{127}I]^9$	Chondrules, Chon-
				drites
⁹² Nb	36	⁹² Zr	$(10^{-4})[^{93}\text{Nb}]^{10}$	Chondrites,
			. ,	Mesosiderites
²⁴⁴ Pu	82	Fission	$(7 \times 10^{-3})[^{238}\text{U}]^{11}$	CAIs, Chondrites
¹⁴⁶ Sm	103	¹⁴² Nd	$(9 \times 10^{-4})[^{147}\text{Sm}]^{12}$	Chondrites

* w.r.t. reference nuclide (e.g ⁴⁰Ca in case of ⁴¹Ca)

¹Srinivasan et al., 1994, 1996; ²Murty et al., 1997; Lin et al., 2005; ³Lee et al., 1976, 1977; ⁴Shukolyukov and Lugmair, 1993; Tachibana and Huss, 2003; Mostefaoui et al., 2005; ⁵Mckeegan et al., 2000a; ⁶Brick and Allègre, 1985; Lugmair and Shukolyukov, 1998; ⁷Chen and Wasserburg, 1990; ⁸Kleine et al., 2002; Yin et al., 2002; ⁹Jeffery and Reynold, 1961; ¹⁰Harper, 1996; Schönbächler et al., 2002; ¹¹Hudson et al., 1988; ¹²Lugmair et al., 1983.

have been proposed. If we consider a stellar source and assume a homogenous distribution of the injected short-lived radionuclides in the nebular region where formation of objects such as CAIs, chondrules and differentiated meteorites are taking place, the observed initial abundances of these nuclides provide relative time scales of formation of these objects. On the contrary, the records of solar energetic particle interactions with nebular material will depend upon the activity of the sun as well as other parameters, such as effective irradiation time and irradiation geometry, and cannot be used for inferring relative timescales of early solar system objects. However, such records will help us to understand the energetic environment in the nebula during the early evolution of the sun and the solar system.

1.2 The first solar system objects

Thermodynamical, petrological, chemical and isotopic studies of meteorite samples suggest that the following three groups of objects represent primary early solar system objects known to us at present. They are (i) Refractory objects such as corundum, hibonite and CAIs with fractionated unidentified nuclear anomalies (FUN), (ii) normal CAIs and (iii) chondrules. These objects are considered as products of high temperature nebular processes. Recent studies suggest that some of the secondary (differentiated) objects such as achondrites may have also formed very early in the solar system (Bizzarro et al., 2005a).

1.2.1 Corundum, Hibonite and FUN CAIs

Corundum, hibonite and FUN CAIs, found primarily in carbonaceous chondrites, are rare and characterized by large stable isotope anomalies such as enrichment in neutron rich isotopes, ⁴⁸Ca and ⁵⁰Ti (Fahey et al., 1987; Ireland, 1990; Marhas et al., 2001) and/or highly fractionated isotopic composition (Clayton and Mayeda, 1977; Wasserburg et al., 1977; Davis et al., 1991). If we consider an initially hot nebula (T >

 $2000 \,^{\circ}$ C) of solar composition and a pressure of $10^{-3} - 10^{-4}$ torr, thermodynamical calculations suggest that corundum and hibonite are the first condensates to form in the solar system (Grossman, 1972). Apart from large stable isotopic anomalies these objects are also characterized by enhanced rare earth and refractory trace element abundances following certain patterns. Even though these objects are considered to be the first solids to form in the solar system, they are devoid of or had extremely low abundance of the now-extinct nuclides, such as ²⁶Al and ⁴¹Ca, at the time of their formation. Several explanations have been put forward, and it appears feasible that formation of these objects may have taken place before the injection of the short-lived radionuclides from a stellar source into the solar nebula (Sahijpal and Goswami, 1998). Another possibility is that ²⁶Al was heterogeneous distributed in the solar nebula and these objects formed in a nebular region devoid of ²⁶Al (MacPherson et al., 1995).

1.2.2 Normal CAIs

CAIs are generally found in carbonaceous chondrites and are rare in ordinary or enstatite chondrites. Nonetheless, CAIs constitute a small but important component of chondritic meteorites. Chemical and isotopic properties of CAIs suggest that they are among the most primitive and oldest objects in the solar system having a Pb-Pb age of 4567.2 \pm 0.6 Ma (Amelin et al., 2002). CAIs have also preserved fossil records of short-lived radionuclides, such as ²⁶Al, ⁴¹Ca and ¹⁰Be. CAIs have liquidus temperature of about 1700 K and isotopic and petrographic studies suggest cooling rates of 2 – 50 °C/hr. Many CAIs appear to have undergone partial melting and have slower cooling rates compared to chondrules. Fossil records of now-extinct short-lived radionuclide ²⁶Al have been extensively studied in CAIs and led to the suggestion for a canonical initial solar system ²⁶Al/²⁷Al ratio of 5×10⁻⁵ (MacPherson et al., 1995). However, lower initial ²⁶Al/²⁷Al ratios are also seen in some CAIs. Petrographic studies of such CAIs suggest that they have experienced secondary

disturbances that most probably perturbed the Al-Mg isotope system leading to the lower initial values. The initial 26 Al/ 27 Al ratios in CAIs range from 4×10^{-5} and 5×10^{-5} ; this spread, if converted into time gives a duration of CAIs formation of ~0.2 Ma (MacPherson et al., 1995), although this variation may reflect analytical uncertainty as well. A recent study of high precision Al-Mg isotope systematics in CAIs from Allende meteorites suggests an even shorter time of ~30,000 years as the duration of CAI formation (Bizzarro et al., 2004). There are recent suggestion that the initial 26 Al/ 27 Al may be higher by a few tens of percent from the presently accepted value of 5×10^{-5} (Bizzarro et al., 2004, 2005b; Young et al., 2005); however, in this study the value of 5×10^{-5} is adopted for interpreting the data.

1.2.3 Chondrules

Chondrules, the most abundant constituent of chondritic meteorites, are small silicate dominated spheroids formed via melting of solid precursors followed by fast cooling. Formation of chondrules was a major event in the early history of the solar system and an understanding of the chondrule formation process and its duration is of important significance. Chondrules from all chondrite groups show a wide range in size from a few tens of microns to a centimeter. However, a vast majority of them have a mean diameter of ~1 mm. Chondrules also show a wide diversity in physical and chemical properties, mineralogy, textural type and are usually composed of silicate minerals such as olivine, pyroxene, rare plagioclase, glass, troilite, Fe-Ni metal, etc. Even though Al-rich phases are rare in ferromagnesian chondrules from UOCs, successful attempts have been made to analyze Al-Mg isotope systematics in such chondrules. These studies reveal a range in the initial ²⁶Al/²⁷Al ratio in UOC chondrules at the time of their formation, these values are much lower than the value of 5×10^{-5} commonly seen in CAIs. This difference suggests that CAI formation preceded chondrule formation by at least a million year (Russell et al., 1996; Kita et al., 2000; Srinivasan et al., 2000; Huss et al., 2001; Mostefaoui et al.,

2002). Lead-lead ages of chondrules in the carbonaceous (CR) chondrite Acfer 059 and CAIs from the (CV) chondrite Efremovka also suggest an interval of 2.5 ± 1.2 Ma between formation of the CV CAIs and the CR chondrules (Amelin et al., 2002).

Chondrules have been heated to their liquidus temperature which varies enormously with their bulk compositions. For porphyritic chondrules, the temperature experienced is considered to be little below liquidus that permitted survival of seed nuclei for the generation of crystalline texture on cooling. The melting time, originally postulated to have lasted for several hours, is now considered to be less than a few minutes to take into account retention of moderately volatile elements such as Na and S. Peak temperature inferred from comparison of laboratory simulation experiments with chondrule textures suggest a range of 1800 K to 2200 K. Cooling rates of chondrules are much faster than CAIs and estimates of linear cooling rates vary from 50 to 1000 °C/hr (Hewins et al., 1996; Jones et al., 2000). Several heating mechanisms for chondrule formation have been proposed. Although none of the models is in conformity with all the observed petrographical and chemical properties of chondrules, at present shock induced heating appears to be a plausible mechanism of chondrules formation (Connolly and Love, 1998; Desch and Connolly, 2002; Desch et al., 2005). The most widely studied chondrules are ferromagnesian i.e. Fe, Mg-rich chondrules. The present study focuses on determining the time of onset and duration of chondrule formation in the solar nebula from a study of Al-Mg isotope systematics in chondrules from various UOCs of low petrologic grade. The time starting with the formation of CAIs and the completion of chondrule formation also provide an estimate of the lifetime of the dynamically active phase of the solar nebula when these two objects formed in high temperature events in the nebula. This time scale is an important parameter that needs to be accommodated in any realistic model of the early evolution of the solar system. In this study, I have carried out Al-Mg isotopic analysis of a set of UOC chondrules. The analyzed UOCs belong to petrologic grade 3 and have experienced relatively low degree of thermal

processing in their parent bodies. The petrologic grade 3 is further sub-divided into 10 classes from 3.0 to 3.9 using various indicators of thermal metamorphism such as TL sensitivity, presence of presolar grains (diamonds), degree of heterogeneity in olivine, amount of primordial ³⁶Ar, degree of matrix recrystallization, Cr distribution in olivine, etc (Sears et al., 1980; Huss et al., 1995, 2006). The maximum temperature experienced by UOCs belonging to different petrologic grades have been inferred by using various metamorphic indicators and these data suggest that chondrules from UOCs belonging to petrologic grades 3.0 to 3.4 are useful for conducting Al-Mg isotope studies. UOCs belonging to petrologic grade 3.0 are expected to experience temperatures not more than 250 °C, while those belonging to 3.4 did not experience temperature greater than 450 °C in their parent bodies (Sears et al., 1991; Huss et al., 2006). Laboratory experiments show that such temperatures, even if they lasted for tens of million years, will not lead to Mg diffusion that may affect the Al-Mg isotope systematics in Al-rich phases like plagioclase (LaTourrette and Wasserburg, 1998). Based on previous studies reported in literature, I have included in our study chondrules from nine UOCs belonging to petrologic grade 3.0 to 3.3 for studying their Al-Mg isotope systematics. It should be noted that there is no evidence of excess ²⁶Mg due to decay of ²⁶Al in chondrules from UOC belonging to petrologic grade greater than 3.4, indicating that they are prone to possible thermal disturbance affecting their Al-Mg isotope records (Huss et al., 2001).

1.3 ${}^{26}Al$ as a chronometer

The present work involves study of Al-Mg isotope systematics and the use of the short-lived radionuclide ²⁶Al (half life=0.72 Ma), that decays to ²⁶Mg, as a chronometer for inferring the time scale of formation of chondrules relative to CAIs. The discovery of ²⁶Mg excess due to decay of ²⁶Al in Allende CAIs (Lee et al., 1976, 1977) established that CAIs have incorporated live ²⁶Al at the time of their formation that

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decayed in-situ to ²⁶Mg. In practice, one measures the Al and Mg isotope ratios and a plot of measured ²⁶Mg/²⁴Mg verses ²⁷Al/²⁴Mg should define a correlation line often called as "isochron", if ²⁶Al decayed in-situ and the Al-Mg isotope system is well behaved. The slope of this isochron yields the initial ²⁶Al/²⁷Al ratio in the object at the time of its formation. If the initial ²⁶Al/²⁷Al ratio differs for any two analyzed early solar system objects, then this difference, coupled with the half life of ²⁶Al, allow us to find the time difference between their formation. Implicit here is the assumption of a homogeneous initial ²⁶Al reservoir in the solar nebula where the formation of early solar system objects such as the CAIs and chondrules took place, and a closed system behavior of Al-Mg isotope system in them. Objects with high initial ²⁶Al/²⁷Al ratio (higher slope) represent early formed objects and those with lower ratio (lower slope) indicate late forming objects (Figure 1.1). Since the CAIs have a well defined initial ²⁶Al/²⁷Al of 5×10⁻⁵, a determination of initial ²⁶Al/²⁷Al ratios of chondrules will provide their time of formation relative to the CAIs. A spread in the initial ²⁶Al/²⁷Al, if observed amongst the chondrules, will provide the duration over which chondrules formation was taking place in the nebula. As noted earlier (section 1.1) we also assume a stellar origin of ²⁶Al that was injected into the protosolar cloud and got homogenized within the nebula. A stellar origin for ²⁶Al as well several other short-lived radionuclides is supported by several observations that include initial solar system abundances of 26 Al and 41 Ca ($t_{1/2}$ =0.1 Ma) in CAIs from the CV (Vigarano-like) and CM (Murchison-like) carbonaceous chondrites (Sahijpal et al., 1998, 2000), unequivocal detection of ⁶⁰Fe (half life=1.5Ma; Tachibana et al., 2003; Mostefaoui et al., 2005) that is a product of stellar nucleosynthesis and whose initial abundance (${}^{56}\text{Fe}/{}^{60}\text{Fe} > 10^{-7}$) rules out energetic particle production (Goswami et al., 2005). Even though the possibility of an inhomogeneous distribution of ²⁶Al is still being debated, the canonical value of initial ²⁶Al/²⁷Al present in most of the petrographically undisturbed CAIs (MacPherson et al., 1995) and the close agreement of the inferred time scales for various events in the early solar system based



Figure 1.1: Example plot of ²⁶Mg/²⁴Mg vs. ²⁷Al/²⁴Mg expected for different objects hosting ²⁶Al at the time of their formation. CAIs, the first objects to form in the solar system are characterized by the canonical initial value of 5×10^{-5} . Objects that formed later, but before ²⁶Al become extinct, will be characterized by a lower value of initial ²⁶Al/²⁷Al ratio, a line with a lower slope. Terrestrial samples formed much after ²⁶Al was extinct will have a standard reference (terrestrial) ²⁶Mg/²⁴Mg value of 0.13932.(Catanzaro et al., 1966)

on different short-lived (e.g., ²⁶Al, ⁵³Mn, ¹⁸²Hf) and long-lived (^{235,238}U) isotopes as chronometers (Halliday and Kleine, 2006, Thrane et al., 2006) argues against this proposition.

1.4 Formation of early solar system objects: the present understanding

The tightly clustered initial 26 Al/ 27 Al ratio in CAIs within a narrow range around 5×10^{-5} suggests a very small time interval (less than a few times 10^{5} years) for their

formation in the early solar system. Thus for any relative chronology of early solar system objects, the initial 26 Al/ 27 Al ratio of 5×10⁻⁵ in CAIs is treated as the reference value. Even though chondrules are ubiquitous in chondrites, and an understanding of the time and duration of their formation is important for our understanding of early solar system processes, studies of Al-Mg isotope systematics in chondrules was hampered by the low abundance of phases with high Al/Mg ratio suitable to look for excess ²⁶Mg from ²⁶Al decay. Further, even within Al-rich glassy phases (mesostasis) in chondrules, the presence of Mg-rich microcrystalites makes the measurements difficult. The first observation of excess ²⁶Mg in a chondrule like object was first reported for a clast chondrule from Semarkona meteorite (Hutcheon and Hutchison, 1989); however, the first direct evidence for the presence of ²⁶Al in chondrules was provided by Russell et al. (1996). The data for Al-Mg isotope systematics in chondrules from UOC and carbonaceous chondrite currently available (Sheng et al., 1991; Hutcheon and Jones, 1995; Russell et al., 1996; Krot et al., 1999; Srinivasan et al., 2000; Kita et al., 2000, 2005a ; Hutcheon et al., 2000; McKeegan et al., 2000b; Marhas et al., 2000, Huss et al., 2001; Mostefaoui et al., 2002; Kunihiro et al., 2004; Kurahashi et al., 2004) suggest that the highest initial ²⁶Al/²⁷Al ratio in chondrules is more than a factor of two lower than the canonical value for the CAIs. This would suggest that chondrule formation in the solar nebula took place ~1 Ma following the formation of CAIs. As noted earlier, this is also supported by Pb-Pb dating of CV CAIs and CR chondrules by Amelin et al. (2002). However models are also proposed in which the formation of CAIs and Chondrules are considered to be contemporaneous (Shu et al., 1996).

Although the presently available data do suggest a time difference between formation of CAIs and chondrules, the exact magnitude of this difference and, more importantly, the duration for which chondrules formation sustained in the solar nebula remain contentious issues. Time scales of less than a Ma to greater than 4 Ma have been proposed as the duration of chondrule formation by various groups. A

part of this spread could be due to secondary processes affecting the chondrules that can disturb the Al-Mg isotope records; however, this is both difficult to establish or to rule out conclusively. The long duration of chondrule formation exceeding 4 Ma, if true, have important implications for our understanding of the early evolution of solar system objects. I have therefore carried out a systematic study of chondrules from UOCs having petrologic grades of 3.0 to 3.3 to address the question of the onset and duration of chondrule formation in the solar nebula. Chondrules from UOCs of higher petrologic grades were avoided as Al-Mg isotope systematics in chondrules from Chainpur (LL3.4) revealed excess ²⁶Mg in only one chondrule and lack of resolved excess in six other chondrules from UOCs of this grade (Huss et al., 2001). Chondrules from UOCs with petrologic grade 3.5 and above are devoid of ²⁶Al record.

1.5 ${}^{26}Al$ as a heat source in the meteorite parent bodies

Even though there is a general consensus that chondrules from UOCs belonging to petrologic grade 3.0 to 3.3 should retain pristine Al-Mg isotopic record, our knowledge about post formation evolutionary history of the chondrules is not exact. It is not clear if the chondrules accumulate into large-sized (>km) planetesimals soon after formation or they had independent evolution either as an individual entity or as part of much smaller object (meter to kilometer) before being part of larger-sized meteorite parent bodies. Once the chondrules reside in larger size planetesimals they will experience heat generated within the parent body due to radioactive heating by ²⁶Al and also ⁶⁰Fe present in it. Because of its half life, ²⁶Al will dominate the thermal evolution of meteorite parent bodies during their early evolution, while ⁶⁰Fe will be a major source after a few Ma. The extent of heating by

these nuclides depends on the initial ²⁶Al/²⁷Al and ⁶⁰Fe/⁵⁶Fe at the time of formation of the meteroite parent bodies and their sizes. The phases (glass and rare plagioclase) analyzed for Al-Mg isotopes in UOC chondrules are typically ~10 μ m. Mg isotope records in plagioclase can be disturbed over such a scale length at a temperature of ~500 °C within a time period of about one Ma (LaTourrette and Wasserburg, 1998). Although no experimental data are available, the temperature for resetting Al-Mg isotope records in glassy phases may be lower than in plagioclase. I have therefore considered the possibility of specific pre-parent/ parent body processes that may affect the observed ²⁶Al records in chondrules from UOCs of even lower petrologic grades that has been analyzed in this study.

1.6 The scientific objective and scope of this thesis

The initial studies of ²⁶Al records in chondrules from UOCs were concentrated on Alrich chondrules that were followed by studies of the more common ferromagnesian chondrules (Hutcheon and Hutchison, 1989; Hutcheon et al., 1994; Russell et al., 1996; Kita et al., 2000; McKeegan et al., 2000b; Huss et al., 2001; Mostefaoui et al., 2002). Chondrules from UOCs belonging to LL group having petrologic grade 3.0 to 3.4 were analyzed in these studies. For the present work, I have selected nine UOCs belonging to both L and LL groups with petrologic grades ranging from 3.0 to 3.3. The particular samples analyzed are, Semarkona (LL3.0) and Bishunpur (LL3.1) that are falls, and finds from Algerian deserts Adrar-003 (L/LL3.1) and from Antarctica, namely LEW86134 (L3.0), QUE97008 (L3.05), LEW86018 (L3.1), Y-791324 (LL3.1), ALHA77176 (L3.2) and ALHA76004 (LL3.3). This work represents the first detail study of ²⁶Al record in chondrules from UOCs belonging to the L group.

Twenty nine chondrules selected out of more than two hundred chondrules from these meteorites were analyzed for Al-Mg isotope systematics to look for the presence of ²⁶Al in them at the time of their formation. The studies were carried out

using ion microprobe technique that allows isotopic analysis of microscopic phases \sim 5-10 μ m in size, the typical size of the Al-rich phases in UOC chondrules.

The scientific objectives of the present work are:

- (i) To understand the time of onset of chondrule formation in the early solar system using 26 Al as a relative chronometer. The initial 26 Al/ 27 Al ratio of 5×10^{-5} for refractory Ca-Al rich inclusions (CAIs), considered to be the first solids to form in the solar system, is treated as the reference value (MacPherson et al., 1995). This study along with data obtained in earlier work will allow a robust determination of the time of onset of chondrule formation in the solar nebula relative to the CAIs.
- (ii) To estimate the duration of chondrule formation in the early solar system from the observed spread in initial ²⁶Al/²⁷Al in chondrules from UOCs of low petrologic grades, obtained in this work as well as those reported earlier by others groups, assuming a uniform distribution of ²⁶Al in the chondrule forming regions in the solar nebula.
- (iii) To check whether the distributions in initial ²⁶Al/²⁷Al in chondrules from various groups of UOCs (L and LL) and of different petrologic grades (3.0 to 3.3) are consistent with each other and to study plausible effect of thermal metamorphism in a pre-parent or parent body environment on the observed ²⁶Al records in chondrules.
- (iv) To place a limit on the active life time of the solar nebula defined as the time spanning the start of formation of CAIs to the cessation of chondrule formation.

The thesis consists of six chapters. Chapter 2 provides details of the various instruments such as ion microprobe, electron microprobe and scanning electron microscope and the experimental approach used in the present study. Descriptions of the analyzed chondrules along with their chemical composition are given in

chapter 3. Chapter 4 includes the results of Al-Mg isotope studies of chondrules from UOCs of various petrologic grades obtained using the ion microprobe. In Chapter 5, I present a discussion of the results along with their implications towards constraining the time of onset and the duration of chondrule formation in the solar nebula. The life time of the dynamically active solar nebula and the possible role of ²⁶Al as a heat source and its possible impact on the observed ²⁶Al records in certain chondrules are also discussed. A part of the work described in this thesis has been published recently (Rudraswami and Goswami, 2007). The last chapter provides a summary and conclusions of this work and outlines the scope for future work.

CHAPTER 2

Experimental techniques: SIMS, SEM and EPMA

The experimental work carried out in this study involves characterization of chemical and mineralogical compositions of Al-rich phases in chondrules followed by determination of Al-Mg isotope composition in them. Three instruments were utilized for achieving these objectives: a Scanning Electron Microscope (SEM) for identifying Al-rich phases within chondrules, an Electron Micro Probe Analyzer (EPMA) for obtaining mineralogical and chemical composition of these phases and a high-mass-resolution Secondary Ion Mass Spectrometer (SIMS) for Al-Mg isotope studies of the microscopic Al-rich phases. A detailed description and basic working principles of the ion microprobe is presented here along with the procedures used for measurement of Al-Mg isotope composition. A brief description of SEM and EPMA, used for sample characterization, is also included.

The Secondary Ion Mass Spectrometer (SIMS) also known as ion microprobe has served as an important tool for isotopic characterization of microscopic solid and is being used extensively by geo-chemists and cosmo-chemists during the last three decades (Lovering, 1975; Shimizu et al., 1978; Shimizu and Hart, 1982; Benninghoven et al., 1987; Ireland, 1994). Of the various analytical methods developed for elemental and isotopic studies of microscopic solids during the latter half of the last century, the ion microprobe with imaging capability is probably one of the most powerful and sophisticated tool. This instrument has been used extensively for studying CAIs and chondrules. Most of the Al-rich phases present in chondrules analyzed have sizes of the order of a few tens of microns or less, and a high mass resolution ion microprobe was used for studying Al-Mg isotope systematics in these phases.

2.1 The Cameca ims-4f ion microprobe

The basic working principle of an ion microprobe involves bombardment of a solid surface with a high energy primary ion beam that results in the ejection of sputtered neutral atoms, molecular fragments, clusters and a small fraction of ionized species. The sputtered ions, also called the secondary ions, are extracted, accelerated and energy filtered prior to passing through an electromagnet for mass analysis. Intensity of ions of a given mass to charge ratio, passing through the electromagnet, are measured using either an electron multiplier or a faraday cup. Intensity of ions of various masses obtained in this manner provide quantitative data on the isotopic composition of the analyzed sample. The extremely small amount of sample, typically less than a nanogram, consumed in the process makes ion microprobe analysis nearly non-destructive that allows repeated analysis of the same domain in a microscopic solid.

Ion microprobe (or SIMS) analysis performed in the present study were carried out at the Physical Research Laboratory (PRL), Ahmedabad, India using a Cameca ims-4f ion microprobe (Cameca, France). It is a custom made instrument that may be used either as an ion microprobe or as an ion microscope. In the following, a description of the various sub-systems and working principle of the Cameca ims-4f are provided.

2.2 Primary sector

The Cameca ims-4f is equipped with two primary ion microbeam sources: a cold cathode duoplasmatron for generating ions from gaseous (e.g. O, Ar) species and a specially designed microbeam source working on the principle of thermal ionization, for providing Cs ion beam.

2.2.1 Oxygen ion source

The oxygen ion source or the duoplasmatron consists of a hollow cylindrical Ni cathode, an anode and a movable intermediate electrode called, Z-electrode. A low-pressure high purity (~99.99%) oxygen gas is introduced into the hollow cathode through a regulating knob. Plasma is generated by an arc maintained between the cathode and the anode that is kept at several hundred volts relative to the cathode (Figure 2.1). Plasma is confined by a variable magnetic field produced by



Figure 2.1: Schematic diagram of the duoplasmatron source.
a coil between the Z-electrode and anode. The duoplasmatron is kept at a high potential, relative to the extraction electrode which is kept at ground potential, that can be varied from 5 to 15 kV in 2.5 kV steps. The energetic primary ion beam emerging through an opening in the extraction electrode enters the primary column, where it is mass filtered and focused before it impinges on the sample surface. The duoplasmatron can deliver both positive (O_2^+) and negative (O^-) ion beams based on the choice of the polarity of the high voltage applied to the duoplasmatron. For obtaining negative ions, the duoplasmatron is kept at a high negative potential and the axis of discharge is displaced relative to the position for positive ion extraction by physically moving the Z-electrode so that the negative ions that are concentrated in the edge of the arc can be extracted.

2.2.2 Cesium ion source

The Cs⁺ ion beam is obtained by heating a Cs metal reservoir, usually cesium chromate or cesium carbonate (Figure 2.2). Cs vapour generated by heating the reservoir up to 400 °C is passed through a porous tungsten filament which is kept at a temperature of 1100 °C that leads to generation of Cs⁺ ions by electron transfer to tungsten atom. The Cs⁺ ions are then accelerated by the potential difference between the source kept at a high positive potential and the grounded extraction electrode.

2.2.3 Primary Column

The primary ions generated by the duoplasmatron source is generally accelerated up to 12.5 kV while in the case of the cesium ion source, it is 10 kV. The ion beam then passed through the primary beam mass filter (PBMF) whose main function is to remove the impurity ions that are present in the beam, hydride and oxide being the primary contaminants. The mass filtered primary beam then passes through the electrostatic deflectors, a set of three electrostatic einzel lenses (L1, L2, L3), an



Figure 2.2: Schematic diagram of the Cesium source.

eight-plate stigmator, double deflector, mass selection aperture and primary beam aperture before impacting on the sample surface (Figure 2.3). The lens L1 controls the intensity of the primary beam, while the lens L2 is used to maximize the beam current. The lens L3 is used to focus the primary beam on the sample surface. The deflectors are used for proper alignment of the beam which guides it to the various lenses.

The double deflector is used to control the X and Y position of the primary beam. The eight plate stigmator located before the double deflector corrects for the astigmatism, caused by off centering of the beam from the principal optical axis of various aperture and lenses. A faraday cup is provided that can be inserted into the beam path to monitor the intensity of the primary beam. The lenses, aperture, deflectors and the stigmator in the primary beam column allows to have a focused primary beam on the sample surface with spot size greater than a few microns.



Figure 2.3: A schematic diagram of primary beam source and primary sector of the Cameca ims-4f ion microprobe.

2.2.4 SAMPLE CHAMBER

The closing stage of the primary column is the sample chamber. The primary beam is made to fall on the sample surface whose normal makes an angle of 30 ° with respect to the optical axis of the primary column. The standard 25 mm diameter sample mounted on a steel holder (Figure 2.4) is located on a pivoting arm allowing the motion in X and Y direction with the help of two micrometers. A magnified image of the sample surface, illuminated by optical light, can be viewed in reflected light using a microscope fitted with a CCD camera coupled to a video monitor. Polished section of samples, potted in epoxy and coated with gold or carbon, is placed in the sample holder that is kept at a potential of +4.5 kV. Thus, the primary O⁻ ions



Figure 2.4: Photograph of sample holder, supporter for the sample and polished carbon-coated and gold-coated samples, respectively (from left to right).

incident on the sample surface have an effective energy of 17 kV. In this configuration positive secondary ions sputtered from the sample surface can be extracted using a grounded extraction plate and they enter the secondary sector. The polarity of the sample surface can be changed for extracting negative secondary ions.

2.3 Production of secondary ions

The energetic primary ions impacting on the sample surface, transfer their energy and momentum to the lattice atoms, thereby setting them free from the lattice site. These atoms in turn, transfer their energy to other neighboring atoms in binary collisions. The continuation of this process leads to a "collision cascade" with large number of atoms in motion with low energy. The consequence of this cascade process leads to ejection of surface atoms that have received adequate momentum to surmount the surface barrier from the top few atomic layers in the samples. The ejected or sputtered particles consist of neutral atoms and molecular species (e.g. oxides, hydrides, dimers etc) along with a small fraction of positive and negative ions. The secondary ions have an energy distribution of more than a hundred eV with a peak at a few eV (Figure 2.5).

The yield of secondary ions is dependent on the nature of the primary beam and the chemical composition of the matrix. Detection of positive secondary ions is generally done using ${}^{16}O^{-}$ primary beam that provides higher positive secondary ion



Figure 2.5: The energy spectrum of positive secondary ions sputtered by an ¹⁶1O⁻ from the Burmal spinel. An energy window of 25 eV was used.

yield for elements like Mg and Al. The yield of secondary ions varies from element to element and is difficult to estimate analytically, and needs to be determined experimentally using samples with known composition.

2.4 Secondary column

Secondary ions emitted from the sample surface have a wide angular and energy spread. The design of the Cameca ims-4f secondary sector provides efficient means of collection and analysis of these ions. It also provides ion imaging capability. The schematic of the secondary ion transfer optics is shown in Figure 2.6.

Sputtered secondary ions are extracted and collected using the grounded electrostatic lens called the immersion lens, positioned at a distance of a few mm from the sample surface that is kept at a potential of +4.5 kV. Positive secondary ions sputtered from the sample are accelerated toward the immersion lens and get energized and pass through the aperture (a few mm-sized opening) in the immersion lens. The secondary ions from the sample surface are characterized by two parameters, their kinetic energy E and angle of emergence as depicted in Figure 2.7. The ions accelerated by the electrostatic field leaving the sample surface (MM') from points A_i , B_i , C_i and passing through the immersion lens (NN') emerge into a region devoid of electrostatic field where they appear to come from the points A, B, C, respectively in the virtual image plane. Secondary ions leaving the sample surface and having the same energy and initial angle, appear to have come from specific point from the plane C_oP (Figure 2.7). The net effect of the immersion lens is thus to construct a virtual image of the sample surface, called the crossover, at the point C_o .



Figure 2.6: Schematic diagram of the secondary sector of Cameca ims-4f ion microprobe.



Figure 2.7: Sketch showing the trajectories of the secondary ions emitted from the sample surface.

The immersion lens system focuses the secondary ions on the deflector, which corrects for the alignment of transfer optics. This is followed by three sets of transfer einzel lenses that form a real magnified image of the cross over and the sample surface in the plane of the contrast aperture and the field aperture, respectively. The instrument can provide three discrete magnifications ($25 \ \mu m$, $150 \ \mu m$, and $250 \ \mu m$), through energizing a set of lenses at a time. In this study, I have primarily used the set of lenses that provides an imaged field of diameter 150 μm . The remaining two magnifications are used during the tuning of the instrument. The contrast aperture and the field aperture are located after the transfer lenses in the focal plane of the crossover , and before the electrostatic analyzer (Figure 2.6). Interchangeable circular apertures of sizes 20 μm , 50 μm , 150 μm and 400 μm are placed at the entrance slit plane where crossover image is formed and these are

followed by adjustable entrance slit which limits the size of the crossover, thereby reducing spherical aberration and improving the spatial resolution of the ion image. Different sizes (100 μ m, 400 μ m, 750 μ m, 1800 μ m) of field aperture can be chosen for placing at the position of the real image of the sample surface. It is possible to restrict the field of view or size and shape of the image using a combination of transfer lens, contrast aperture, entrance slit, field aperture and exit slit. The secondary ions passing through the field aperture enter the electrostatic sector.

2.4.1 Electrostatic analyzer (ESA)

The electrostatic analyzer (ESA) placed just after the field aperture (Figure 2.6) helps to reduce the momentum dispersion of the secondary ions that have an energy spread of ~150 eV. The ESA consists of two concentric spherical surfaces (electrodes) separated by 1 cm and with a mean radius of 8.5 cm. It is a 90 ° sector analyzer and the electrodes are kept at appropriate potentials (typically a potential difference of ~1050 V) such that secondary ions with energy 4.5 kV will follow the mean path, while trajectories of ions of higher/lower energy will trace above/below this path. Suitable adjustment of the energy slit following the ESA allows selection of secondary ions within a narrow energy band thus reducing the momentum dispersion of the ions selected for mass analysis using an electromagnet. The variable slits present at the entrance and exit of ESA limit the beam divergence and reduces the chromatic aberrations facilitating for high mass resolution analysis.

2.4.2 Magnetic analyzer

The secondary ions with a small energy-band width (typically ~25 eV in our study) emerging from the energy slit, is directed to the magnetic prism via the spectrometer lens (Figure 2.6). The magnetic prism produces separation of the secondary ions according to their mass to charge ratio (m/q). The magnetic prism consists of an electromagnet with a gap width of 8 mm and radius of 12.7 cm. The combination of

the magnetic and the electrostatic prism makes the Cameca ims-4f a double focusing instrument. The tuning of the spectrometer lens provides desired achromatism. The mass resolution of the instrument can be adjusted by adjusting the width of the entrance and the exit slits.

2.4.3 Detection system

The ions emerging from the magnetic prism are passed through an exit slit and projector lens and are made to incident on the ion detector using deflectors. There are two modes of ion detection, imaging and intensity, achieved by a channel plate and a fluorescent screen for ion imaging, and an electron multiplier and a faraday cup for ion intensity. At any given time one of these detecting systems can be switched on. A brief description of each is given in the following.

2.4.4 CHANNEL PLATE

Ion imaging of the sputtered sample surface emitting the secondary ions is done with the channel plate that is attached to a phosphor screen for visualization of the ion image (Figure 2.6). In this mode the ion image of the sample is transferred though the mass analyzer to the fluorescent screen where a magnified ion image of the sample is provided. The fluorescent screen image may be observed on a monitor using a CCD. The brightness of the image is proportional to the ion intensity. In this mode of operation the ion microprobe act as an ion microscope, which is very helpful for tuning the instrument to high mass resolution by ion imaging of the entrance and exit slits.

2.4.5 Electron multiplier

The electron multiplier facilitates precise measurement of secondary ion intensities. The secondary ions after passing through the exit slit can be made to fall on the dynode of the electron multiplier using a deflector. The secondary ions interacting with the activated surface (e.g. Cu/Be or Al) of the dynode produce secondary electrons. These electrons are further accelerated by a suitable voltage difference (200 - 300 V) between successive dynodes leading to amplification in the seventeen stage electron multiplier (SEV 217, Pfeiffer-Balzer) used in this study. The output from the electron multiplier is monitored in pulse counting mode using high-speed electronic counter. The dynamic background of the total counting system is typically <0.01 cps. The detection efficiency of the electron multiplier deteriorates with time and it is replaced when the efficiency falls below 70%. The basic parameters such as the background, dead time and efficiency of the counting system are monitored on a regular basis.

2.4.6 Faraday Cup

The faraday cup is positioned just adjacent to the electron multiplier. When the count rate exceeds 2×10^6 cps, the counting mode automatically switched over to the faraday cup. Faraday cups normally have significant background noise and are not suitable for precise measurement of ion intensity when the count rate is $<10^6$. As most of the isotopic results obtained in this work is based on detection of low count rates, we have not used this mode of detection for measurement of ion intensities.

2.4.7 VACUUM SYSTEM

The active volume of the ion microprobe (the ion source region, sample chamber, primary and secondary sectors and the detection system) should be kept in high vacuum. This is achieved using two rotary pumps and four turbo pumps in the primary sector and the samples chamber, and with two ion pumps in the secondary sector. The vacuum in the secondary sector of the ion microprobe is kept at $<10^{-8}$ torr and can be isolated from the sample chamber using a isolation value. There are various safety controls and values at strategic positions that monitors the vacuum

in the system.

2.5 Sample preparation

Samples to be analyzed with the ion microprobe have to be polished to have a smooth flat surface down to <1 μ m. Samples are potted in suitable epoxy within standard holders or on a glass slide for making polished section following standard procedures. The epoxy mounted polished samples are then cleaned ultrasonically using distilled water and alcohol. As the meteorite samples are usually non-conducting, a coating ($\leq 1000 \text{ A}^{\circ}$) of either pure gold or carbon is provided to avoid charging effect during analysis. The sample preparation has to be done in a clean environment to avoid possible terrestrial contamination.

The polished sections of the samples are then properly documented and studied using optical microscope followed by scanning electron microscope (SEM), equipped with energy dispersive X-ray analyzer, to identify mineral phases suitable for isotopic studies. Back-scattered electron images of the sections as well as of all the phases, considered suitable for analysis are taken. Elemental mappings of the phases as well as studies of their chemical composition are then carried out using an Electron Probe Micro Analyzer (EPMA; Cameca SX-100). A brief description of SEM and EPMA is provided here.

2.6 Scanning Electron Microscope (SEM)

The scanning electron microscope (SEM) provides high resolution images that are suitable for identifying microscopic individual phases. The electron column of the SEM consists of the electron gun, demagnifying lenses, beam defining apertures and scanning coils, and one can have micron-sized beam of electrons that can be focused on to the polished sample surface for obtaining secondary or back-scattered electron images. Non-conducting samples such as meteorite, terrestrial rock samples need

to be coated with carbon for preventing charging. The magnification can be varied over a wide range of <100 to 100,000. Images taken with back-scattered electrons also provide broad information on the composition of the phases. I have obtained back-scattered electron images at low magnification of all the polished thin sections of the meteorite samples and also high magnification images of all the chondrules within the sections. The energy dispersive X-ray analyzer attached to the SEM can be used to further probe the small areas of selected phases within chondrules for their elemental composition. A LEO-440i SEM facility available at the Facilitation Center for industrial and plasma technology (FCIPT), Gandhinagar, was used for analyzing the meteorite samples selected for this study.

2.7 Electron Probe Micro Analyzer (EPMA)

The electron probe micro analyzer (EPMA; Cameca SX-100) is an important analytical tool for determining quantitative chemical composition at microscopic scale. It allows non-destructive, in situ analysis with high spatial resolution, good accuracy and has low detection limits. The EPMA was used to determine chemical composition of the Al-rich areas (glass and plagioclase), olivine and pyroxene in chondrules selected for Mg isotope studies. The vacuum in the optical column of the EPMA including the wavelength dispersive spectrometers (WDS) is kept at ~10⁻⁴ mbar, while in the region of electron gun the vacuum is ~10⁻⁶ mbar.

Electrons emitted from the electron gun (tungsten filament) are collimated, accelerated and focused by a series of magnetic lenses and apertures resulting in approximately 1 - 2 μ m diameter focussed electron beam incident on the carbon coated polished sections of sample. Detection of characteristic X-rays emitted from the sample by the wavelength-dispersive X-ray spectrometers is based on Bragg's reflection. Various crystals such as Lithium fluoride, Pentaerythritol, Thallium acid Phythallate are used to cover the range of required wavelength. The X-rays are detected by means of a propotional counter. Quantitative electron microprobe analysis is based on a comparison of the intensity of the characteristic X-rays emitted from the sample with those from reference geological standards. The instrument parameters used for EPMA are: acceleration voltage of 15 kV, a beam current of 12 nA and a beam Diameter of ~2 μ m. Concentrations of eleven elements (Na, Mg, Si, Al, P, K, Ca, Ti, Cr, Fe, Mn) were measured. In addition Al, Mg and Ca X-ray elemental maps were also obtained for selected chondrules with a resolution of 2-3 μ m/pixel using an accelerating voltage of 15 kV, a beam current of ~50-100 nA and a spot size of ~2 μ m for identifying Al-rich areas within them.

2.8 A*l*-M*g* isotope systematics

The present work focuses on the Al-Mg isotope systematics in different phases (plagioclase, Al-rich glassy mesostasis, olivine, pyroxene) of UOC chondrules and analog terrestrial standards. The measurements of the Al-Mg isotope abundances were made using the ion microprobe operated with a primary O⁻ beam and current of 1-3 nA with a beam spot ~5-10 μ m on the sample surface. A mass resolution $(M/\Delta M)$ of ~4000 is adequate to resolve hydride (NaH^+, MgH^+) , ${}^{48}Ca^{++}$, ${}^{48}Ti^{++}$ and other molecular interferences at the masses (²⁴Mg, ²⁵Mg, ²⁶Mg and ²⁷Al) of interest (Figure 2.8). During the analysis ${}^{24}Mg^+$ count rate was kept below 10⁵ counts/s to minimize dead time correction. A typical analysis consists of twenty blocks of data with each block consisting of five cycles. Counting time for each mass is based on the observed count rate to ensure a reasonable precision in the measured isotope ratios. The counting time for ²⁴Mg was adjusted to achieve total count of $\sim 10^6$ in an analysis and count time for 25 Mg and 26 Mg was typically four times that for ²⁴Mg. Measurements were carried out in peak jumping mode by cycling the magnet through the mass sequence ²⁴Mg, ²⁵Mg, ²⁶Mg and ²⁷Al with suitable wait time (typically 1 sec) before start of ion counting at each mass.



Figure 2.8: High mass resolution spectra at masses 24, 25, 26 and 27 obtained at a mass resolution (M/ Δ M) of ~4000 showing completely resolved hydride peaks.



Figure 2.9: Slit images of ${}^{24}Mg^+(a)$, ${}^{25}Mg^+(b)$, ${}^{26}Mg^+(c)$ and ${}^{27}Al^+(d)$ taken with a CCD camera.



Figure 2.10: Ion images of ²⁷Al⁺ and ²⁴Mg⁺ of a Al-rich region in chondrule glass in UOC QUE97008 taken using a CCD camera.

Automatic mass calibration is performed after one or more blocks during an analysis. Multiple analyses are conducted on the same spot, as long as the Al/Mg ratio remained stable, to improve analytical precision. Typical shape of the peaks and the slit images taken with a CCD camera for Mg at masses 24, 25, 26 and ²⁷Al are shown in Figures 2.8 and 2.9. Images of ²⁷Al⁺ and ²⁴Mg⁺ obtained for a Al-rich spot in a glassy phase in one of the chondrules from UOC QUE97008, using a small field aperture, is shown in Figure 2.10.

Isotope ratios measured by ion microprobe usually differ from the true value due to mass fractionation introduced during production, extraction and detection of ions; the magnitude of this mass fractionation, termed as instrumental mass fractionation, can be evaluated by analyzing appropriate terrestrial analog standards.

Any departure in the measured isotope ratios of ²⁵Mg/²⁴Mg and ²⁶Mg/²⁴ with respect to reference (standard) values was obtained using the following relation:

$$\Delta^{26} Mg = \left[\frac{(^{i}Mg/^{24}Mg)_{sample}}{(^{i}Mg/^{24}Mg)_{standard}} - 1\right] \times 1000 \ per \ mil \ (\%); [i = 25, 26]$$
(2.1)

where $({}^{i}Mg/{}^{24}Mg)_{sample}$ and $({}^{i}Mg/{}^{24}Mg)_{standard}$ are the measured isotope ratios in the sample and the reference (standard) value, respectively. For Mg isotopes, the standard values are: $({}^{25}Mg/{}^{24}Mg) = 0.12663$ and $({}^{26}Mg/{}^{24}Mg) = 0.13932$ (Catanzaro et al., 1966).

A linear mass fractionation law was applied to look for any excess in ²⁶Mg in the analyzed Al-rich phases in UOC chondrules, using the relation:

$$\delta^{26}Mg = \Delta^{26}Mg - 2 \times \Delta^{25}Mg \tag{2.2}$$

For terrestrial standard $\delta^{26}Mg = 0$.

In addition to correcting measured isotope ratios of a given element for instrument mass fractionation, it is also necessary to take into account the differences in ion yields when isotope ratio involves two elements e.g. 27 Al/ 24 Mg. The relative ion yield (λ) of two elements (e.g. Al and Mg) can be obtained by comparing the ion intensity ratio (27 Al+/ 24 Mg⁺) with elemental ratio (Al/Mg) measured using electron microprobe.

$$\lambda = \left[\frac{({}^{27}Al/{}^{24}Mg)_{electron\ microprobe}}{({}^{27}Al^+/{}^{24}Mg^+)_{ion\ microprobe}}\right]$$
(2.3)

A synthetic glass standard (GB-4) with 15.57% Al_2O_3 , 0.14% MgO and 72.94% SiO_2 and Lake county plagioclase were analyzed to infer ion yields of Al and Mg in glass relative to Lake County plagioclase. The results suggest that ion yield of Al relative to Mg in glass is ~8% higher than in plagioclase under similar instrument operating conditions. In the absence of an exact analog standard for chondrule glass (mesostasis) and the small difference noted above, we have used the ion yield measured for the plagioclase standard to infer the ²⁷Al/²⁴Mg ratio, based on the measured ²⁷Al+/²⁴Mg⁺ ratio, in both glass and plagioclase in the anlyzed UOC chondrules.

CHAPTER 3

Sample description: UOC chondrules

Most chondritic meteorites have experienced thermal metamorphism on their parent bodies (asteroids) resulting in textural, mineralogical, chemical and isotopic changes. The nine Unequilibrated Ordinary Chondrites (UOCs) selected for the present work are among the least metamorphosed with petrologic grade ranging from 3.0 to 3.3 and are expected to preserve their pristine isotope records. The inferred temperature experienced by these meteorites in their parent bodies is not expected to exceed 400 °C (Sears et al., 1991; Huss et al., 2006) and at such temperatures glass/plagioclase in UOC chondrules should not be susceptible to alteration or diffusion of Mg over scale length of tens of microns (LaTourette and Wasserburg, 1998). The samples for the present study include observed falls, Semarkona (LL3.0) and Bishunpur (LL3.1), received from Geological Survey of India, and finds from Algerian desert, Adrar-003 (L/LL3.1) and from Antarctica: LEW86134 (L3.0), QUE97008 (L3.05), LEW86018 (L3.1), and ALHA77176 (L3.2), received from NASA collection of Antarctica meteorite, and Y-791324 (LL3.1) and ALHA76004 (LL3.3), received from National Institute of Polar Research (NIPR), Tokyo, Japan. All the samples were in the form of polished thin section mounted on glass disc, except for sample from Adrar-003 (L/LL3.1) that was received as a fragment and was processed to obtain epoxy mounted polished thick section. Each of these sections was studied and mapped using an optical microscope, a scanning electron microscope equipped with energy dispersive X-ray analyzer (EDX) and an electron microprobe. Most of the chondrules are ferromagnesian in nature. X-ray elemental maps of Al, Mg and Ca were also obtained for each selected chondrules. Potential high Al/Mg phases, such as glassy mesostasis/plagioclase within these chondrules were identified using EDX for Al-Mg isotope studies. Identified Al-rich areas, typically tens of microns across, were further analyzed to determine their quantitative chemical composition using an electron probe micro analyzer (EPMA). The inferred chemical composition are given in Tables 3.1 to 3.14. The composition of minerals are expressed as Fo (Mg₂SiO₄) for olivine, Wo (CaSiO₃), En (MgSiO₃), Fs (FeSiO₃) for pyroxene, and An(CaAl₂Si₂O₈), Ab(NaAlSi₃O₈), and Or(KAlSi₃O₈) for plagioclase. Some of the chondrules were also studied for determining abundance of chromium in olivine, following ion microprobe analysis of Al-Mg isotope systematics, to infer the petrologic grade of their host meteorites following the approach of Grossman and Brearley (2005).

Brief descriptions of the UOC samples selected for this study are given below by grouping them into observed falls and finds belonging to L and LL groups.

3.1 UOCs: LL group

Five UOCs belonging to LL group and of low petrologic grades were selected for the present study. These include, observed falls Semarkona (LL3.0) and Bishunpur (LL3.1), and finds from Antarctica, Y-791324 (LL3.1) and ALHA76004 (LL3.3) and from hot Algerian desert, Adrar-003 (L/LL3.1).

3.1.1 Semarkona (LL3.0)

The UOC Semarkona belonging to petrologic grade LL3.0 fell in Chhindwara district of Madhya Pradesh, India in the year 1940. Two pieces weighing 423 g and 269 g were recovered. The thin section of this meteorite analyzed in this study was provided by Geological Survey of India (GSI), Kolkota. Semarkona chondrules are the most widely studied samples for Al-Mg isotope records. Mild aqueous alteration was reported in matrix of Semarkona (Alexander et al., 1989) and in radial pyroxene and cryptocrystalline chondrules (Grossman et al., 2000), but this has not effected the chondrule mineral composition. Bulk sample of Semarkona is not expected to experience thermal metamorphism at greater than 250 °C in its parent body (Alexander et al., 1989). Optical microscope and SEM studies of the analyzed Semarkona section revealed that most of the chondrules are porphyritic in nature and Al-rich areas suitable for isotope studies are primarily glassy mesostasis. Electron microprobe studies conducted on Al-rich areas yielded SiO₂ content ranging from 60 - 75 wt% (Tables 3.1 and 3.2). The sum of major oxides in silicates is close to 100 wt% indicating absence of significant hydrous reaction. Back-scattered electron images of the four Semarkona chondrules analyzed for Al-Mg isotope systematics are shown in Figure 3.1. One of them belongs to type IIB (porphyritic pyroxene), two others belong to type IIAB (porphyritic olivine pyroxene) and the fourth is a type IB (porphyritic pyroxene) chondrule.

3.1.2 BISHUNPUR (LL3.1)

Bishunpur meteorite fell in 1895 in Mirzapur district, Uttar Pradesh, India. Four stones fell after explosion, of which two were recovered; one at Bishunpur weighing 942 gram and other weighing 97 gram at Parjabatpur more than a km away. Bishunpur belongs to petrologic grade LL3.1 and is expected to experience a maximum temperature of ~300 °C in its parent body (Huss et al., 2006) and should have a well preserved Al-Mg isotope record. A polished thin section of this meteorite, received



Figure 3.1: Back-scattered electron images of the analyzed Semarkona (LL3.0) chondrules. The encircled regions are the area of ion microprobe analysis.

from Geological Survey of India, was studied to identify Al-rich areas in chondrules that are suitable for Al-Mg isotope studies. Most of the chondrules have low Al/Mg phases and we could select only two porphyritic olivine (type IIA) chondrules for Mg isotope studies. Back-scattered electron images of these chondrules are shown

in Figure 3.2. Results from quantitative analysis of major elements in Al-rich areas (glassy mesostasis) analyzed for isotope records are given in Table 3.3.

Chondrule#2 (IIB)							
Analysis No.	1	2	3	4	5	6	7
Na ₂ O	2.95	4.74	4.14	3.7	0.01	0.06	
MgO	0.01	0.13	0.07	0.04	40.39	28.49	
SiO ₂	71.53	62.44	69.94	66.11	38.52	56.19	
Al_2O_3	22.51	26.69	21.9	23.42	0.02	0.1	
P_2O_5	0.04	0.02	0.04	0.02	0.1	0.03	
K ₂ O	0.7	1.67	0.49	2.37	0	0.01	
CaO	0.12	0.98	0.93	1.42	0.16	0.52	
TiO ₂	0.04	0.06	0.42	0.2	0	0.03	
Cr_2O_3	0.02	0	0	0	0.53	0.71	
FeO	0.3	0.6	0.43	0.41	19.98	13.65	
MnO	0	0	0.06	0	0.51	0.6	
Total	98.23	97.32	98.41	97.7	100.2	100.38	
Phase	Glass	Glass	Glass	Glass	Olivine	Pyroxer	ne
composition					Fo78	Wo1En7	78Fs21
		Cho	ndrule#2	0 (IIAB)			
Na ₂ O	2.62	1.92	2.78	2.85	2.13	0.01	0.01
MgO	0.23	0.48	0.02	0.01	0.38	43.39	26.64
SiO ₂	73.29	74.21	72.17	68.32	65.36	38.98	53.88
Al_2O_3	11.44	10.48	11.43	13.85	12.18	0.01	0.15
P_2O_5	0.07	0.06	0.07	0.05	0.06	0	0
K ₂ O	1	1.07	0.99	0.88	0.76	0	0.01
CaO	1.39	1.78	1.1	2.28	4.14	0.06	0.61
TiO ₂	0.37	0.33	0.37	0.41	0.42	0	0
Cr_2O_3	0.03	0.02	0.05	0.02	0.03	0.46	0.95
FeO	5.86	5.64	6.72	7.6	11.28	16.78	16.2
MnO	0.32	0.12	0.24	0.16	0.43	0.45	0.85
Total	96.59	96.11	95.9	96.4	97.14	100.15	99.3
Phase	Glass	Glass	Glass	Glass	Glass	Olivine	Pyroxene
composition						Fo82	Wo1En74Fs25

Table 3.1: Chemical Composition of Semarkona (LL3.0) chondrules.

	Cho	ndrule#21	(IIAB)	X	,
Analysis No.	1	2	3	4	5
Na ₂ O	3.1	7.85	4.19	2.5	0.02
MgO	0.17	0.71	0.09	0.6	45.81
SiO ₂	64.1	62.34	63.57	73.68	39.66
Al_2O_3	19.51	18.81	19.63	10.35	0.04
P_2O_5	0.23	0.25	0.12	0	0.01
K ₂ O	1.03	0.29	0.87	1.2	0
CaO	1.91	3.81	0.65	0.41	0.09
TiO ₂	0.3	0.28	0.43	0.36	0
Cr_2O_3	0.48	0.56	0.02	0	0.42
FeO	4.42	1.87	5.11	4.77	12.64
MnO	0.15	0.04	0.19	0.14	0.29
Total	95.37	96.81	94.85	94.01	98.98
Phase	Glass	Glass	Glass	Glass	Olivine
composition					Fo87
	Chondr	ule#36 (IB)			
Na ₂ O	7.79	8.78	0.01	0.01	
MgO	0.19	0.23	48.69	36.25	
SiO ₂	59.25	60.76	40.5	58.08	
Al_2O_3	24.18	22.47	0.01	0.22	
P_2O_5	0.02	0.04	0	0	
K ₂ O	0.23	0.28	0	0	
CaO	6.8	5.06	0.15	0.22	
TiO ₂	0.07	0.11	0	0.05	
Cr_2O_3	0	0.01	0.6	0.58	
FeO	0.17	0.28	8.8	4.29	
MnO	0.07	0.04	0.73	0.4	
Total	98.77	98.03	99.48	100.1	
Phase	Glass	Glass	Olivine	Pyroxene	
composition			Fo91	Wo1En93	BFs6

Table 3.2: Chemical Composition of Semarkona (LL3.0) chondrules.

3.1.3 Adrar-003 (L/LL3.1)

Adrar-003, a 287 gram stone found in southern Algeria, is enriched in presolar grains (Huss et al., 2002) and belongs to petrologic grade of L/LL3.1. We have made



Figure 3.2: Back-scattered electron images of the analyzed Bishunpur (LL3.1) chondrules. The encircled regions are the area of ion microprobe analysis.

two polished sections of this sample received from Open University, UK. We have analyzed nearly a dozen chondrules in these two sections looking for high Al/Mg phases and selected three chondrules for isotope studies. The back-scattered electron images of these chondrules are given in Figure 3.3. All the analyzed chondrules belongs to type IIA (porphyritic olivine). Table 3.4 provides data on major element composition of the spots analyzed for isotope records that includes mesostasis as well as plagioclase.

3.1.4 Y-791324 (LL3.1)

This UOC was recovered in the Yamato mountain region of Antarctica in 1979 and belongs to petrologic grade 3.1. A thin section of this sample received from NIPR did

not have many chondrules and we could find only two chondrules that are suitable for Al-Mg isotope studies. Most of the phases selected are glassy mesostasis with feldspathic composition. The elemental compositions of the analyzed phases are

Chondrule	#45 (IIA)		1	<u> </u>	
Analysis No.	1	2	3	4	5
Na ₂ O	6.99	0.02			
MgO	0.32	39.23			
SiO ₂	59.95	37.82			
Al_2O_3	25.56	0.03			
P_2O_5	0	0			
K ₂ O	0.6	0			
CaO	0.46	0.09			
TiO ₂	0.52	0.05			
Cr_2O_3	0.61	0.04			
FeO	1.33	20.99			
MnO	0.15	0.41			
Total	96.49	98.7			
Phase	Glass	Olivine			
composition		Fo77			
	Chone	drule#61 (I	IA)		
Na ₂ O	4.54	7.29	6.4	5.48	0.02
MgO	0.51	0.6	0.23	4.79	38.48
SiO ₂	62.99	59.6	61.77	59.02	37.81
Al_2O_3	24.68	23.4	25.38	23.56	0.02
P_2O_5	0	0	0	0	0
K ₂ O	0.5	1.09	0.87	0.34	0
CaO	0	0.14	0.13	0.17	0.09
TiO ₂	0.4	0.97	0.29	0.27	0.06
Cr_2O_3	0.48	0.61	0.21	0.14	0.06
FeO	1.99	1.38	0.86	3.12	21.71
MnO	0.02	0	0	0	0.47
Total	96.11	95.08	96.14	96.89	98.73
Phase	Glass	Glass	Glass	Glass	Olivine
composition					Fo76

Table 3.3: Chemical Composition of Bishunpur(LL3.1) chondrules.

given in Table 3.5. The two selected chondrules belong to type IIA (porphyritic olivine) and type IIAB (porphyritic olivine pyroxene), respectively. Back-scattered electron images of these are shown in Figure 3.4.



Figure 3.3: Back-scattered electron images of the analyzed Adrar-003 (L/LL3.1) chondrules. The encircled regions are the area of ion microprobe analyses.

Chondrule#2 (IIA)							
Analysis No.	1	2	3	4	5		
Na ₂ Ó	1.45	0.94	1.28	1.47	0.01		
MgO	0.35	0.45	0.61	0.38	43.32		
SiÕ ₂	45.21	43.98	45.23	45.59	39.15		
Al_2O_3	33.26	34.21	33.55	33.54	0.06		
P_2O_5	0.31	0.29	0.25	0.28	0.04		
K ₂ O	0.01	0.01	0	0.01	0		
CaO	17.44	18.66	17.97	17.81	0.23		
TiO ₂	0.02	0.08	0.05	0.01	0.04		
Cr_2O_3	0.07	0	0	0.02	0.28		
FeO	1.93	1.48	1.21	1.54	17.58		
MnO	0.03	0	0.03	0.01	0.27		
Total	100.07	100.07	100.16	100.62	100.98		
Phase	Plagioclase	Plagioclase	Plagioclase	Plagioclase	Olivine		
Composition	An87Ab13	An92Ab8	An89Ab11	An88Ab12	Fo81		
*	Cho	ondrule#3 (IIA	A)				
Na ₂ O	3.89	3.67	5.8	0.18			
MgO	0.76	0.5	0.62	39.17			
SiO ₂	70.09	71.84	70.16	38.24			
$Al_2\bar{O}_3$	9.59	8.24	11.64	0.5			
$P_2 \overline{O}_5$	0.33	0.32	0.4	0.07			
$\bar{K_2O}$	1.28	1.24	1.27	0.04			
CaO	1.3	0.87	0.66	0.16			
TiO ₂	0.44	0.45	0.42	0.02			
$Cr_2\bar{O}_3$	0	0.05	0.06	0.78			
FeŌ	9.31	10.21	7.52	20.5			
MnO	0.24	0.26	0.19	0.58			
Total	97.23	97.65	98.74	100.24			
Phase	Glass	Glass	Glass	Olivine(Fo78	3)		
		Chondrule	e#8 (IIA)	X			
Na ₂ O	1.66	1.78	0.63	1.94	0.03		
MgO	0.41	0.6	0.43	0.59	40.77		
SiO	46.53	46.66	43.43	47.13	38.72		
Al_2O_3	32.66	32.35	34.45	31.8	0.18		
$P_2 O_5$	0.3	0.28	0.37	0.19	0.08		
K_2O	0.01	0.01	0	0.01	0.02		
CaO	17.25	16.95	18.91	16.45	0.35		
TiO	0.01	0.03	0.01	0	0.03		
Cr_2O_2	0.02	0.05	0.07	0.02	0.28		
FeO	1.48	1.39	1.25	1.6	20.15		
MnO	0	0.02	0.04	0.05	0.23		
Total	100.32	100.09	99.59	99.75	100.84		
Phase	Plagioclase	Plagioclase	Plagioclase	Plagioclase	Olivine		
Composition	An85Ah15	An84Ah16	An94Ah6	An82Ah18	<u>Fo78</u>		
Composition	1110011010	1 110 11 10 10	1 11 / 11 10 0	1110411010	10/0		

Table 3.4: Chemical Composition of Adrar-003 (L/LL3.1) chondrules.

3.1.5 ALHA76004 (LL3.3)

Allan Hills A76004, a stony meteorite, found in a region of Victoria Land, Antarctica, in 1976, belongs to petrologic grade 3.3. More than a dozen chondrules were analyzed and four of these were selected for Al-Mg isotope studies. Back-scattered electron images of these chondrules are shown in Figure 3.5. All the four chondrules analyzed belong to type IIAB (porphyritic olivine pyroxene). Results from electron microprobe analyses of Al-rich phases in these chondrules are given in Table 3.6 and 3.7. Most of them are feldspathic glass except for one plagioclase (An₈₅Ab₁₅).



Figure 3.4: Back-scattered electron images of the analyzed Y-791324 (LL3.1) chondrules. The encircled regions are the area of ion microprobe analysis.



Figure 3.5: Back-scattered electron images of the analyzed ALHA76004 (LL3.3) chondrules. The encircled regions are the area of microprobe analysis.

Chondrule#6 (IIAB)							
Analysis No.	1	2	3	4	5		
Na ₂ O	9.83	9.72	9.03	8.83	0.03		
MgO	0.38	0.34	0.75	0.48	37.83		
SiO ₂	60.92	64.5	63.66	62.96	37.6		
Al_2O_3	15.43	15.56	14.86	15.42	0.02		
P_2O_5	0.2	0.17	0.16	0.19	0.03		
K ₂ O	0.04	0.08	0.08	0.06	0		
CaO	1.64	1.59	2.01	1.96	0.12		
TiO ₂	0.52	0.53	0.51	0.52	0.01		
Cr_2O_3	0.01	0	0.07	0.08	0.22		
FeO	7.67	5.5	7.1	6.55	23.05		
MnO	0.14	0.1	0.16	0	0.65		
Total	96.75	98.06	98.37	97.05	99.55		
Phase	Glass	Glass	Glass	Glass	Olivine		
Composition					Fo74		
	Chon	drule#3 (II	A)				
Na ₂ O	2.92	3.5	0.02				
MgO	0.59	0.63	49.16				
SiO ₂	68.42	67.15	40.35				
Al_2O_3	17.86	17.69	0.01				
P_2O_5	0.11	0.04	0.01				
K ₂ O	0.26	0.11	0.01				
CaO	0.31	0.66	0.11				
TiO ₂	0.6	0.59	0.01				
Cr_2O_3	0	0.02	0.28				
FeO	3.27	4.81	10.07				
MnO	0.05	0.1	0.43				
Total	94.39	95.26	100.44				
Phase	Glass	Glass	Olivine				
Composition			Fo90				

Table 3.5: Chemical Composition of Y-791324 (L3.1) chondrules.

Chondrule#1 (IIAB)						
Analysis No.	1	2	3	4	5	
Na ₂ O	13.55	0.03	0.01			
MgO	0.78	36.48	26.27			
SiO ₂	43.83	37.76	53.32			
Al_2O_3	36.76	0.03	2.17			
P_2O_5	0.1	0.02	0.02			
K ₂ O	0.17	0.01	0.01			
CaO	3.34	0.16	2.37			
TiO ₂	0.04	0.01	0.15			
Cr_2O_3	0.22	0.08	1.06			
FeO	0.68	25.64	13.92			
MnO	0.02	0.48	0.6			
Total	99.51	100.69	99.89			
Phase	Plagioclas	eOlivine	Pyroxene			
Composition	An85Ab15	5 Fo72	Wo5En73I	Fs22		
	Ch	ondrule#2 (IIAB)			
Na ₂ O	7.01	7.53	8.11	6.5	0.01	
MgO	1.82	5.48	2.96	1.83	40.48	
SiO ₂	67.31	65.66	64.82	67.85	38.56	
Al_2O_3	11.37	11.52	11.71	13.21	0.01	
P_2O_5	0.29	0.35	0.27	0.36	0.02	
K ₂ O	0.22	0.04	0.04	1	0.01	
CaO	3.06	1.51	2.98	1.4	0.07	
TiO ₂	0.39	0.47	0.47	0.57	0.02	
Cr_2O_3	0.02	0.07	0.02	0.02	0.04	
FeO	7.71	7.78	7.99	5.69	21.48	
MnO	0.09	0.14	0.23	0.07	0.38	
Total	99.27	100.55	99.6	98.47	101.08	
Phase	Glass	Glass	Glass	Glass	Olivine	
Composition					Fo77	

Table 3.6: Chemical Composition of ALHA76004 (L3.3) chondrules.

Chondrule#11 (IIAB)						
Analysis No.	1	2	3	4	5	
Na ₂ O	7.62	6.07	0.02			
MgO	0.54	0.76	38.43			
SiO ₂	69.36	64.18	38.15			
Al_2O_3	17.48	20.12	0.01			
P_2O_5	0.16	0.16	0.04			
K ₂ O	0.68	2.36	0			
CaO	0.36	0.23	0.05			
TiO ₂	0.59	0.46	0			
Cr_2O_3	0.02	0	0.03			
FeO	2.4	2.91	24.03			
MnO	0	0.02	0.29			
Total	99.21	97.27	101.05			
Phase	Glass	Glass	Olivine			
Composition			Fo74			
	Cho	ndrule#31 ((IIAB)			
Na ₂ O	7.82	10.8	7.93	9.34	0.03	
MgO	0.68	0.13	0.27	0.29	41.38	
SiO ₂	68.61	66.57	63.93	64.16	38.49	
Al_2O_3	16.56	20.26	19.19	20.26	0.14	
P_2O_5	0	0.04	0.43	0.31	0.02	
K ₂ O	0.09	0.05	0.95	0.32	0.01	
CaO	1.04	0.54	0.34	0.61	0.09	
TiO ₂	0.51	0.17	0.2	0.2	0.01	
Cr_2O_3	0.06	0.02	0	0.01	0.05	
FeO	3.12	1.12	1.5	2.94	19.64	
MnO	0	0	0	0	0.54	
Total	98.5	99.7	94.74	98.44	100.41	
Phase	Glass	Glass	Glass	Glass	Olivine	
Composition					Fo79	

Table 3.7: Chemical Composition of ALHA76004 (L3.3) chondrules.

3.2 UOCs: L group

The four samples of L chondrites analyzed in this study, namely LEW86134 (L3.0), QUE97008 (L3.05), LEW86018 (L3.1) and ALHA77176 (L3.2) were received from the NASA collection of Antarctic meteorites. A brief description of the samples is presented below.

3.2.1 LEW86134 (L3.0)

This sample belongs to the least metamorphosed L3.0 group. Four ferromagnesian chondrules from about a dozen chondrules were selected for isotope studies. Back-scattered electron images of these chondrules are shown in Figure 3.6. Among the analyzed chondrules two belong to type IIAB (porphyritic olivine pyroxene), one to type IIA (barred olivine) and the other is type IIB (porphyritic pyroxene). SEM and electron microprobe studies were conducted to determine chemical and mineralogical composition and to identify phases having high Al/Mg ratio suitable for isotopic analysis. Results of quantitative analysis of major element composition in analyzed glass and minerals in these chondrules are given in Table 3.8 and 3.9.

3.2.2 QUE97008 (L3.05)

This sample, initially classified as belonging to petrologic grade L3.4, has been reclassified as L3.05 by Grossman and Brearley (2005) based on Cr distribution in olivine. Based on analyses of nearly a dozen chondrules, four chondrules were selected for Al-Mg isotope studies. Back-scattered electron images of these chondrules are shown in Figure 3.7. Three of the analyzed chondrules are porphyritic olivine (Type IIA) and one porphyritic olivine pyroxene (type IIAB). Compositional data for feldspathic glass and plagioclase phases (An_{93–99.7}) in chondrules of QUE97008 are summarized in Table 3.10 and 3.11.



Figure 3.6: Back-scattered electron images of the analyzed LEW86134 (L3.0) chondrules. The encircled regions are the area of microprobe analysis.

3.2.3 LEW86018 (L3.1)

LEW86018 belongs to petrologic grade L3.1. Studies for Al-Mg isotope systematics were carried out on four ferromagnesian chondrules from thirteen selected chondrules. Back-scattered electron images of the four chondrules are shown in the Figure 3.8. Two of the analyzed chondrules are of type IIA (porphyritic olivine) and other two are of type IIAB (porphyritic olivine pyroxene). Tables 3.12 and 3.13

provides results from electron microprobe analysis of major elements concentration in Al-rich glass and plagioclase (An $_{83-88}$) analyzed for isotope records using ion microprobe.

	Chondrule#3 (IIAB)							
Analysis No.	1	2	3	4	5			
Na ₂ O	8.38	0.01	0.03					
MgO	0.88	44.41	29.96					
SiO ₂	63.28	39.38	55.54					
Al_2O_3	18.83	0.01	0.41					
P_2O_5	0.05	0.02	0					
K ₂ O	1.06	0	0.01					
CaO	1.82	0.12	0.37					
TiO ₂	0.63	0.01	0					
Cr_2O_3	0.06	0.42	0.72					
FeO	4.23	14.65	11.71					
MnO	0.16	0.32	0.36					
Total	99.38	99.35	99.11					
Phase	Glass	Olivine	Pyroxene					
Composition		Fo84	Wo1En81I	Fs18				
	Chond	rule#16 (IIB)					
Na ₂ O	5.94	7.75	7.06	0.01	0.29			
MgO	1.05	0.15	0.57	37.73	17.96			
SiO ₂	68.04	63.24	65.25	38.33	49.97			
Al_2O_3	10.79	15.01	13.21	0.02	0.7			
P_2O_5	0.03	0.15	0.05	0.01	0.02			
K ₂ O	1.08	1.13	1.1	0	0.02			
CaO	1.18	1.03	1.09	0.09	4.39			
TiO ₂	0.51	0.61	0.64	0	0.07			
Cr_2O_3	0.04	0.01	0.02	0.32	0.91			
FeO	8.49	9.65	9.29	22.47	24.47			
MnO	0.4	0.26	0.29	0.63	0.81			
Total	98.97	98.57	97.51	99.6	99.6			
Phase	Glass	Glass	Glass	Olivine	Pyroxene			
Composition				Fo75	Wo9En52Fs39			

Table 3.8. Chemical Composition of LEW86134 (L3.0) chondrules

Table 5000 enternieur Composition of 22000000 (2000) enternareis.							
Chondrule#36 (IIA)							
Analysis No.	1	2	3	4	5		
Na ₂ O	5.5	3.71	4.42	0.01	0.78		
MgO	0.51	0.26	0.28	42.9	18.69		
SiO ₂	70.36	68.54	69.3	39.41	51.83		
Al_2O_3	11.61	10.68	11.22	0.02	2.13		
P_2O_5	0.01	0.03	0.03	0	0		
K ₂ O	1.09	0.95	1	0	0.11		
CaO	0.67	0.98	0.84	0.09	5.28		
TiO ₂	0.43	0.54	0.5	0	0.12		
Cr_2O_3	0.01	0.01	0.02	0.53	1.12		
FeO	6.44	10.28	9.04	16.21	17.14		
MnO	0.39	0.46	0.33	0.5	0.87		
Total	97	96.4	96.95	99.67	98.07		
Phase	Glass	Glass	Glass	Olivine	Pyroxene		
Composition				Fo82	Wo12En58Fs30		
Cł	nondrule#3	6A (IIAB)					
Na ₂ O	3.61	0.01	0.01				
MgO	0.48	45.36	29.9				
SiO ₂	63.72	39.4	55.52				
Al_2O_3	18.7	0.07	0.43				
P_2O_5	0.01	0.01	0.07				
K ₂ O	1	0.01	0				
CaO	0.89	0.17	0.73				
TiO ₂	0.66	0.01	0.03				
Cr_2O_3	0.02	0.42	0.78				
FeO	6.19	13.81	11.6				
MnO	0.18	0.32	0.55				
Total	95.26	99.26	99.08				
Phase	Glass	Olivine	Pyroxene				
Composition		Fo85	Wo1En811	Fs18			

Table 3.9: Chemical Composition of LEW86134 (L3.0) chondrules.

3.2.4 ALHA77176 (L3.2)

This UOC have petrologic grade L3.2. Our search for identifying Al-rich phases yielded only two chondrules that have phase suitable for isotopic analysis. Back-

scattered electron image of these two chondrules are shown in Figure 3.9. Both chondrules belong to type IIAB (porphyritic olivine pyroxene). Quantitative electron microprobe data for major elements in minerals and Al-rich glassy mesostasis are summarized in Table 3.14.

The Al-rich areas analyzed for Al-Mg isotope systematics in most of the chondrules from the nine UOC samples described above are primarily glassy mesostasis



Figure 3.7: Back-scattered electron images of the analyzed QUE97008 (L3.05) chondrules. The encircled regions are the area of microprobe analysis.
with MgO content ranging from 0.01 to 0.99 wt% while the Al_2O_3 content ranges between about 10 and 25 wt%. Plagioclase, present in six chondrules, are mostly anorthositic in composition with Al_2O_3 content ranging between 32-37 wt%. Mg-

Table 3.10: Chemical Composition of QUE97008 (L3.05) chondrules.					
	Cho	ondrule#1A	(IIA)		
Analysis No.	1	2	3	4	5
Na ₂ O	0.81	0.6	0.32	0	0.42
MgO	0.71	0.9	0.58	43.91	19.28
SiO ₂	46.12	46.44	44.03	38.96	50.75
Al_2O_3	32.41	31.2	33.01	0.07	9.1
P_2O_5	0	0	0	0.01	0
K ₂ O	0	0	0.01	0	0.05
CaO	18.72	18.33	18.83	0.02	8.12
TiO ₂	0.11	0.04	0.07	0.01	0.53
Cr_2O_3	0.03	0	0	0.52	0.58
FeO	0.77	0.75	1.49	15.34	10.31
MnO	0.05	0	0	0.18	0.24
Total	99.73	98.26	98.34	99.02	99.35
Phase	Plagioclas	ePlagioclas	ePlagioclas	eOlivine	Pyroxene
Composition	An93Ab7	An94Ab6	An97Ab3	Fo84	Wo19En62Fs19
	Chondru	le#1B (IIA)			
Na ₂ O	0.61	0.03	0.37	0.01	
MgO	0.7	0.76	0.94	43.82	
SiO ₂	46.49	47.62	47.61	39.11	
Al_2O_3	32.8	32.12	31.51	0.07	
P_2O_5	0	0	0	0.01	
K ₂ O	0	0	0	0	
CaO	18.65	18.49	18.48	0.03	
TiO ₂	0.05	0.02	0.03	0.01	
Cr_2O_3	0.03	0.06	0	0.62	
FeO	0.65	0.67	0.74	15.11	
MnO	0	0.02	0	0.2	
Total	99.97	99.79	99.68	98.98	
Phase	Plagioclas	ePlagioclas	ePlagioclas	eOlivine	
Composition	An94Ab6	An100	An97Ab3	Fo84	

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rich microcrystallites are often present within the Al-rich glass that hampers Al-Mg isotope studies as the Al/Mg ratio changes drastically when the primary beam sputters such phases along with the Al-rich phase. In all the chondrules, the chemical

Table 3.11: Chemical Composition of QUE97008 (L3.05) chondrules.					
	Ch	ondrule#5	(IIA)		
Analysis No.	1	2	3	4	5
Na ₂ O	6.13	5.77	6.97	5.62	0.03
MgO	0.19	0.27	0.25	0.15	39.37
SiO ₂	67.76	70.33	67.13	70.22	37.95
Al_2O_3	13.98	13.29	15.86	13.21	0.02
P_2O_5	0	0	0.02	0.04	0.01
K ₂ O	1.22	1.26	1.2	1.26	0
CaO	0.79	0.64	0.66	0.61	0
TiO ₂	0.48	0.48	0.5	0.45	0
Cr_2O_3	0.04	0	0.02	0.03	0.04
FeO	6.43	5.27	4.85	5.95	20.47
MnO	0.18	0.16	0.17	0.15	0.43
Total	97.18	97.45	97.6	97.66	98.32
Phase	glass	glass	glass	glass	Olivine
Composition					Fo77
Cho	ondrule#16	(IIAB)			
Na ₂ O	1.93	2.36	0.02		
MgO	0.9	0.86	45.66		
SiO ₂	66.49	65.61	40.14		
Al_2O_3	16.02	15.96	0.02		
P_2O_5	0.1	0.08	0.01		
K ₂ O	0.09	0.71	0		
CaO	1.93	2.63	0.04		
TiO ₂	0.58	0.57	0		
Cr_2O_3	0.01	0.02	0.44		
FeO	5.76	6.08	12.29		
MnO	0.21	0.38	0.41		
Total	93.98	95.23	99.02		
Phase	glass	glass	Olivine		
Composition			Fo87		

composition of the major mineral phases (olivine and pyroxene) nearly devoid of Al was routinely measured and these data are also provided in the data tables. They were also analyzed for Mg isotopic composition for obtaining the true initial 26 Mg/²⁴Mg ratio in the chondrules. In nearly all the cases the values are consistent with the reference value of 0.13932.



Figure 3.8: Back-scattered electron images of the analyzed LEW86018 (L3.1) chondrules. The encircled regions are the area of microprobe analysis.



Figure 3.9: Back-scattered electron images of the ALHA77176 (L3.2) chondrules. The encircled regions are the area of microprobe analysis.

	Chondrule#6 (IIA)				
Analysis No.	1	2	3	4	
Na ₂ O	6.84	9.9	5.79	0.01	
MgO	0.36	0.26	0.06	43.24	
SiO ₂	62.52	53.28	55.36	39.44	
Al_2O_3	19.23	21.15	22.98	0.02	
P_2O_5	0.06	0.06	0.04	0.06	
K ₂ O	2.35	1.2	1.65	0.01	
CaO	0.12	0.37	0.56	0.15	
TiO ₂	0.03	0.09	0.09	0.02	
Cr_2O_3	0	0.01	0	0.12	
FeO	3.57	8.91	8.95	18.01	
MnO	0	0.02	0.04	0.38	
Total	95.08	95.21	95.51	101.44	
Phase	Glass	Glass	Glass	Olivine	
composition				Fo81	
(Chondrule#	6A (IIA)			
Na ₂ O	8.1	8.77	0.02		
MgO	0.2	0.31	44.18		
SiO ₂	61.64	62.71	39.58		
Al_2O_3	24.17	19.57	0.02		
P_2O_5	0	0.05	0.05		
K ₂ O	1.58	2.67	0.01		
CaO	0.5	0.12	0.15		
TiO ₂	0.07	0.28	0.02		
Cr_2O_3	0	0.05	0.15		
FeO	2.8	4.03	16.73		
MnO	0.01	0	0.38		
Total	99.04	98.56	101.28		
Phase	Glass	Glass	Olivine		
composition			Fo82		

Table 3.12: Chemical Composition of LEW86018 (L3.1) chondrules.

	Chondrule#24 (IIAB)					
Analysis No.	1	2	3	4	5	6
Na ₂ O	11.45	6.18	5.7	0.02		
MgO	0.43	0.21	0.26	42.77		
SiO ₂	64.69	67.35	61.42	39.19		
Al_2O_3	20.98	21.47	20.9	0.08		
P_2O_5	0.1	0.02	0	0.06		
K ₂ O	0.07	2.12	1.61	0.01		
CaO	0.68	0.05	0.17	0.11		
TiO ₂	0.29	0.06	0.02	0.01		
Cr_2O_3	0.09	0	0.14	0.04		
FeO	1.54	0.79	4.5	18.61		
MnO	0.02	0.03	0.05	0.37		
Total	100.34	98.26	94.77	101.25		
Phase	Glass	Glass	Glass	Olivine		
Composition				Fo80		
		Chond	rule#33 (IIA	AB)		
Na ₂ O	1.31	1.86	1.58	1.96	7.97	0.01
MgO	0.23	0.16	0.62	0.29	0.17	50.02
SiO ₂	45.13	46.33	45.78	46.79	58.56	41.01
Al_2O_3	32.64	32.99	32.71	33.19	29.31	0.11
P_2O_5	0.18	0.21	0.26	0.2	0.06	0.04
K ₂ O	0.03	0.03	0.01	0.03	2.33	0.01
CaO	18.13	17.43	17.4	17.33	0.06	0.31
TiO ₂	0.07	0.03	0.05	0.11	0.07	0.01
Cr_2O_3	0	0.01	0.04	0	0.04	0.2
FeO	0.95	0.76	0.81	0.87	0.87	8.94
MnO	0	0.01	0	0.03	0.01	0.16
Total	98.66	99.8	99.22	100.78	99.45	100.82
Phase	Plagioclas	ePlagioclas	ePlagioclas	ePlagioclas	ePlagioclas	eOlivine
Composition	An88Ab12	2 An84Ab16	6 An86Ab14	4 An83Ab17	7 Ab84Or16	Fo90

Table 3.13: Chemical Composition of LEW86018 (L3.1) chondrules.

Analysis No.	$\frac{1}{1}$	2	3	4	5
	Chond		<u>4</u> B)		
Na ₂ O	5.21	5.16	4.92	4.5	0.01
MgO	0.77	0.3	0.4	0.32	49.56
SiO ₂	69.02	69.95	67.39	69.78	40.94
Al_2O_3	17.23	16.36	15.71	14.2	0.01
P_2O_5	0.16	0.08	0.16	0.13	0.03
K ₂ O	1.26	1.23	1.16	1.2	0
CaO	1.15	0.79	1.77	1.54	0.1
TiO ₂	0.5	0.47	0.63	0.47	0
Cr_2O_3	0.05	0.01	0.05	0.05	0.31
FeO	2.5	3.62	6	6.06	10.04
MnO	0.06	0.15	0.17	0.19	0.32
Total	97.89	98.12	98.36	98.42	101.32
Phase	Glass	Glass	Glass	Glass	Olivine
Composition					Fo90
	Chondr	ule#14B (I	IAB)		
Na ₂ O	5.37	4.35	4.77	5.31	0.01
MgO	0.95	1.09	1.2	0.94	36.77
SiO_2	64.33	68.63	66.12	66.84	38.01
Al_2O_3	17.39	16.57	18.68	18.28	0.01
P_2O_5	0.23	0.28	0.24	0.31	0.06
K ₂ O	1.26	1.26	1.13	1.31	0.01
CaO	1.55	2.02	2.13	1.93	0.18
TiO ₂	0.69	0.69	0.5	0.69	0.01
Cr_2O_3	0.01	0.07	0	0.02	0.22
FeO	3.91	2.69	2.58	3.12	25.09
MnO	0.09	0.08	0.08	0.13	0.57
Total	95.75	97.72	97.43	98.88	100.93
Phase	Glass	Glass	Glass	Glass	Olivine
Composition					Fo72

Table 3.14: Chemical Composition of ALHA77176 (L3.2) chondrules.

3.3 *Cr* distribution in UOC meteorites

The UOCs selected for the present study belong to low petrologic grades (3.0 to 3.3) and are not expected to experience peak temperature exceeding 400 °C on their parent bodies (Huss et al., 2006). Various indices (Thermoluminescence, presence of presolar grains (diamonds), degree of heterogeneity in olivine, amount of primordial ³⁶Ar, degree of matrix recrystallization) have been used to infer the degree of thermal metamorphism experienced by UOCs (Sears et al., 1980; Huss et al., 1995, 2006). In principle, they refer to bulk samples and often different indices indicate different degree of thermal processing for the same meteorite. Recently, Grossman and Brearley (2005) suggested an approach based on chromium (Cr) content in FeO-rich olivine, which they suggested to be very sensitive to metamorphic conditions and changes noticeably even between subtypes 3.0 to 3.2. Since such an analysis can be done in the very chondrule where Al-Mg isotope systematics studies are also conducted, we have obtained data from approximately 20 olivine from each of the analyzed chondrules in all the nine UOCs. Figures 3.10 and 3.11 show the plots of Cr₂O₃ verses FeO in wt% for chondrules in different UOCs analyzed by us for Al-Mg isotope systematics. These data show that Semarkona (LL3.0), the least equilibrated ordinary chondrite, has a mean Cr₂O₃ content of 0.50 wt% in ferroan olivines with a range of 0.4 to 0.6 wt%. Bishunpur (LL3.1) chondrite has a Cr₂O₃ content of 0.089 ± 0.05 wt% in its ferroan olivine which is less than given by Grossman and Brearley (2005). Data for QUE97008 matches those reported by Grossman and Brearley (2005) who reclassified this sample as 3.05. Our data suggest that the petrologic grade of LEW86134 (L3.0) could be L3.05. Figure 3.12 is a plot of mean and standard deviation of Cr₂O₃ content in olivine in UOCs measured in this study (dark circles) and those reported by Grossman and Brearley (2005) (open circles). The results overlap and suggest the samples as well as the chondrules analyzed in this study did not experience high degree of thermal metamorphism and should retain pristine records of Al-Mg isotope records from the time of their formation.



Figure 3.10: Plots of Cr_2O_3 verses FeO in olivine from Semarkona (LL3.0), LEW86134 (L3.0), QUE97008 (L3.05), Bishunpur (LL3.1), Adrar-003 (L/LL3.1) and LEW86018 (L3.1).



Figure 3.11: Plot of Cr_2O_3 in olivine verses FeO in Y-791324 (LL3.1), ALHA77176 (L3.2) and ALHA76004 (LL3.3).



Figure 3.12: Standard deviation versus the mean of Cr_2O_3 content in ferroan olivine in chondrules from UOCs. Dark blue circles are the analysis of present work, open cirles are from Grossman and Brearley (2005).

CHAPTER 4

Al-Mg isotope data for UOC chondrules

Results obtained from ion microprobe study of Al-Mg isotope systematics in Alrich phases of chondrules from nine unequilibrated ordinary chondrites (UOCs) belonging to low petrologic grade are presented in this chapter. These data along with characterization of the analyzed chondrules and chemical composition of the analyzed phases described in Chapter 3 constitute the primary data set of this thesis.

It is already noted in Chapter 2 that one can infer possible presence of excess ²⁶Mg (δ^{26} Mg), normalized to ²⁴Mg, by assuming a linear instrumental mass fractionation effect using the relation (2.2):

$$\delta^{26}Mg = \Delta^{26}Mg - 2 \times \Delta^{25}Mg$$

Further, if it can be shown that δ^{26} Mg is linearly correlated with the abundance of ²⁷Al (normalized to ²⁴Mg), the excess in ²⁶Mg can be attributed to in situ decay of ²⁶Al within the analyzed object. This can be verified from a plot of measured ²⁶Mg/²⁴Mg verses ²⁷Al/²⁴Mg that will define a linear correlation, often referred to as Al-Mg "isochron". The slope of the isochron yields the initial ²⁶Al/²⁷Al ratio in the objects at the time of its formation and the intercept gives initial ²⁶Mg/²⁴Mg:

$$\left[\frac{{}^{26}Mg}{{}^{24}Mg}\right]_{measured} = \left[\frac{{}^{26}Mg}{{}^{24}Mg}\right]_{intial} + \left[\frac{{}^{26}Mg^*}{{}^{24}Mg}\right]$$
(4.1)

$$= \left[\frac{{}^{26}Mg}{{}^{24}Mg}\right]_{intial} + \left[\frac{{}^{26}Al}{{}^{27}Al}\right]_{intial} \times \left[\frac{{}^{27}Al}{{}^{24}Mg}\right]_{measured}$$
(4.2)

where "measured" refers to the measured value corrected for instrumental mass fractionation [²⁶Mg/²⁴Mg] and for yield factor [²⁷Al/²⁴Mg], "initial" refers to the initial or reference value (0.13932) for ²⁶Mg/²⁴Mg and "*" denote the radiogenic component (²⁶Mg from decay of ²⁶Al). If we assume a homogeneous initial ²⁶Al reservoir in the solar nebula, objects with high initial ²⁶Al/²⁷Al will represent early formed objects and those with lower values represent late forming objects. In the following, we present results obtained from this study in terms of measured Mg and Al isotope ratios in Al-rich phases (glass and plagioclase) as well as Al-poor phases such as olivine and pyroxene in the chondrules. The Al-Mg isotope evolution ("isochron") plot as well as the inferred initial ²⁶Al/²⁷Al ratio for each of the twenty nine chondrules analyzed in this study are also presented. Errors quoted for all the data given in the tables and shown in the figures are 2σ . In the case of ²⁶Mg/²⁴Mg values the errors quoted refer to the least significant digits.

4.1 A*l*-M*g* isotope data from LL-chondrites (Observed Falls)

4.1.1 Semarkona (LL3.0)

Our search for glass or plagioclase phases in chondrules from the thin section of Semarkona (LL3.0) meteorite did not reveal any plagioclase (>10 μ m) suitable for isotope studies. However, we could identify feldsphathic glassy mesostasis that are rich in aluminium. We obtained well-defined Al-Mg isotope systematics in all

the four chondrules selected for analysis. The presence of Mg-rich microcrystallites within Al-rich glassy phases created hindrance during analysis of the other chondrules. Olivine and pyroxene present in the chondrules were also analyzed to better constrain the initial ²⁶Mg/²⁴Mg ratio. The results obtained for the four ferromagnesian Semarkona chondrules are presented in Table 4.1 and the data for ²⁶Mg/²⁴Mg verses ²⁷Al/²⁴Mg are plotted in Figures 4.1 and 4.2. The dotted line shown in these figures represents the reference magnesium isotopic composition

Chondrule#	Phase	²⁷ Al/ ²⁴ Mg	²⁶ Mg/ ²⁴ Mg	Initial ²⁶ Al/ ²⁷ Al*
		Semarkona	(LL3.0)	
2	Pyroxene	0.012 ± 0.0051	0.13932 ± 11	
(IIB)	Glass	80.4 ± 8.9	0.13978 ± 40	
	Glass	145.3 ± 11.7	0.14001 ± 37	
	Glass	280.6 ± 47.6	0.14097 ± 41	
	Glass	427.6 ± 15.7	0.14157 ± 65	
	Glass	746.8 ± 95.9	0.14280 ± 87	
	Glass	1052.8 ± 78.4	0.14456 ± 113	
	Glass	1238.0 ± 14.7	0.14670 ± 67	
	Glass	1619.5 ± 181.8	0.14671 ± 138	
	Glass	2384.4 ± 54.7	0.15256 ± 117	$(5.51 \pm 0.33) \times 10^{-6}$
20	Olivine	0.003 ± 0.003	0.13930 ± 13	
(IIAB)	Glass	62.2 ± 4.1	0.14002 ± 31	
	Glass	75.4 ± 3.1	0.14023 ± 33	
	Glass	92.8 ± 4.0	0.14038 ± 43	
	Glass	125.2 ± 9.8	0.14055 ± 35	$(1.10 \pm 0.24) \times 10^{-5}$
21	Olivine	0.0064 ± 0.011	0.13932 ± 10	
(IIAB)	Glass	56.0 ± 3.3	0.14006 ± 73	
	Glass	91.3 ± 11.3	0.14034 ± 50	$(1.15 \pm 0.54) \times 10^{-5}$
36	Olivine	0.012 ± 0.003	0.13931 ± 15	
(IB)	Glass	53.7 ± 7.1	0.14013 ± 46	
	Glass	68.7 ± 1.3	0.14009 ± 38	$(1.24 \pm 0.52) \times 10^{-5}$

Table 4.1: Al-Mg isotope data for chondrules from Semarkona (LL3.0).

*The initial ²⁶Al/²⁷Al ratio for all the chondrules in the present study was inferred from the slope of the isochron obtained by least square fit method (Provost, 1990).

 $(^{26}\text{Mg}/^{24}\text{Mg} = 0.13932; \delta^{26}\text{Mg}=0)$. All the four chondrules show clear excess of ^{26}Mg with $\delta^{26}\text{Mg}$ up to 95‰; the excess is also well correlated with $^{27}\text{Al}/^{24}\text{Mg}$ ratio. In one of the chondrule (CH#2), we found glassy phases having a very wide range in $^{27}\text{Al}/^{24}\text{Mg}$ ratio, from ~80 up to ~2500, and could obtain a very precise isochron with an experimental uncertainty of ~6% in the inferred initial $^{26}\text{Al}/^{27}\text{Al}$ of (5.51 ± 0.33)×10⁻⁶. The initial $^{26}\text{Al}/^{27}\text{Al}$ values for the other three chondrules are (1.10 ± 0.24)×10⁻⁵, (1.15 ± 0.54)×10⁻⁵ and (1.24 ± 0.52)×10⁻⁵, respectively.

4.1.2 BISHUNPUR (LL3.1)

Al-Mg isotope systematics were analyzed in several chondrules from this meteorite. Most of the analyses were hampered by the presence of Mg-rich microcrystallites within the Al-rich phases and only two chondrules yielded data that could be used for estimation of initial 26 Al/ 27 Al. 27 Al/ 24 Mg ratios in the analyzed phases vary from ~60 to 120. The initial 26 Al/ 27 Al ratios obtained for these two Bishunpur chondrules are $(5.8 \pm 3.1) \times 10^{-6}$ and $(5.8 \pm 2.2) \times 10^{-6}$, respectively. The Al-Mg isotope data are given in Table 4.2 and plotted in Figure 4.3.

Chondrule#	Phase	27 Al/ 24 Mg	$^{26}Mg/^{24}Mg$	Initial ²⁶ Al/ ²⁷ Al
		Bishunpur (LL	.3.1)	
45	Olivine	0.005 ± 0.004	0.13934 ± 12	
(IIA)	Glass	61.0 ± 5.0	0.13968 ± 27	
	Glass	100.0 ± 25.3	0.13993 ± 34	$(5.8 \pm 3.1) \times 10^{-6}$
61	Olivine	0.002 ± 0.001	0.13931 ± 11	
(IIA)	Glass	57.7 ± 4.1	0.13975 ± 35	
	Glass	83.0 ± 3.8	0.13983 ± 30	
	Glass	120.8 ± 11.0	0.13994 ± 31	$(5.8 \pm 2.2) \times 10^{-6}$

Table 4.2: Al-Mg isotope data for chondrules from Bishunpur (LL3.1).



Figure 4.1: Al-Mg isotope data for Semarkona chondrules CH#2 (top) and CH#20 (bottom).



Figure 4.2: Al-Mg isotope data for Semarkona chondrules CH#21 (top) and CH#36 (bottom).



Figure 4.3: Al-Mg isotope data for Bishunpur chondrule CH#45 (top) and CH#61 (bottom).

4.2 Al-Mg isotope data from LL-chondrites (Finds)

Apart from the two observed LL falls, Al-Mg isotope systematics have been studied in chondrules from three more UOCs belonging to LL group that are "finds" from African desert (Adrar-003) and Antarctica (Y-791324 and ALHA76004).

4.2.1 Adrar-003 (L/LL3.1)

More than a dozen chondrules from this meteorite were studied; however, only three of these have reasonable ²⁷Al/²⁴Mg ratios for inferring initial ²⁶Al/²⁷Al. The Al/Mg ratio in mesostasis was <15 in the other chondrules that did not allow a meaningful study in these cases. Table 4.3 summarizes the Al-Mg isotope data in Adrar-003. In two of these chondrules plagioclase was analyzed, while in the third it was Al-rich glass. We show in Figures 4.4 and 4.5 the results of Al-Mg isotope systematics for these three chondrules. The data yielded initial ²⁶Al/²⁷Al of (6.3 ± 4.0)×10⁻⁶, (8.4 ± 7.8)×10⁻⁶ and (4.3 ± 4.2)×10⁻⁶, respectively. The initial ²⁶Mg/²⁴Mg in all the cases is consistent with the normal value (0.13932) within the limit of our measurement uncertainties.

4.2.2 Y-791324 (LL3.1)

It was difficult to find high Al/Mg phases in chondrules of this meteorite. We could attempt Al-Mg isotope analysis in only two chondrules where we could find moderately Al-rich glassy phases. The isotope data obtained in them are given in Table 4.4 and the Al-Mg isotope plots are shown in Figures 4.6. The data yielded initial 26 Al/ 27 Al of $(1.22 \pm 0.52) \times 10^{-5}$ and $(1.1 \pm 1.0) \times 10^{-5}$, for these two chondrules. The initial 26 Mg/ 24 Mg inferred from olivine data is indistinguishable from the reference value.

4.2.3 ALHA76004 (LL3.3)

From more than a dozen chondrules identified for analysis, we were successful in obtaining meaningful results in four chondrules from this sample. Presence of Mg-rich microcrystalites within the Al/Mg rich phases makes it difficult to carry out meaningful analysis in the other chondrules. In one of the chondrules (CH#1) we could identify plagioclase and the data obtained matches those for Al-rich glassy phases in the other chondrules. Data for the four chondrules are given in the Table 4.5 and are plotted in Figures 4.7 and 4.8. All the four chondrules have well-defined Al-Mg isotope systematics and yielded initial 26 Al/ 27 Al ratios of $(1.06 \pm 0.33) \times 10^{-5}$, $(1.15 \pm 0.34) \times 10^{-5}$, $(7.1 \pm 2.0) \times 10^{-6}$, and $(1.14 \pm 0.28) \times 10^{-5}$, respectively.

Table 4.3: Al-Mg isotope	e data for	chondrules from	Adrar-003 ((L/LL3.1).

Chondrule#	Phase	²⁷ Al/ ²⁴ Mg	$^{26}Mg/^{24}Mg$	Initial ²⁶ Al/ ²⁷ Al
		Adrar-003 (L/L	L3.1)	
2	Olivine	0.001 ± 0.0002	0.13926 ± 12	
(IIA)	Plagioclase	26.6 ± 1.6	0.13936 ± 19	
	Plagioclase	30.7 ± 0.5	0.13939 ± 16	
	Plagioclase	39.8 ± 1.6	0.13955 ± 32	
	Plagioclase	48.4 ± 1.4	0.13968 ± 47	
	Plagioclase	65.8 ± 1.8	0.13966 ± 51	
	Plagioclase	87.7 ± 2.7	0.13988 ± 57	$(6.3 \pm 4.0) \times 10^{-6}$
3	Olivine	0.007 ± 0.003	0.13934 ± 19	
(IIA)	Glass	24.8 ± 1.3	0.13942 ± 36	
	Glass	31.7 ± 2.7	0.13953 ± 41	
	Glass	37.5 ± 2.6	0.13969 ± 25	$(8.4 \pm 7.8) \times 10^{-6}$
8	Plagioclase	8.8 ± 2.3	0.13903 ± 33	
(IIA)	Plagioclase	11.3 ± 1.5	0.13921 ± 24	
	Plagioclase	17.9 ± 1.8	0.13940 ± 28	
	Plagioclase	21.6 ± 1.8	0.13948 ± 33	
	Plagioclase	25.1 ± 3.0	0.13939 ± 30	
	Plagioclase	28.6 ± 1.0	0.13950 ± 35	
	Plagioclase	31.8 ± 1.7	0.13933 ± 38	
	Plagioclase	33.9 ± 0.6	0.13913 ± 49	
	Plagioclase	39.7 ± 3.2	0.13906 ± 47	
	Plagioclase	44.7 ± 4.4	0.13951 ± 21	
	Plagioclase	51.4 ± 2.2	0.13951 ± 34	
	Plagioclase	60.8 ± 1.8	0.13947 ± 23	
	Plagioclase	64.2 ± 2.0	0.13940 ± 26	$(4.3 \pm 4.2) \times 10^{-6}$



Figure 4.4: Al-Mg isotope data for Adrar-003 chondrules CH#2 (top) and CH#3 (bottom).



Figure 4.5: Al-Mg isotope data for Adrar-003 chondrule CH#8.

	Table 4.4. AF-Wg isotope data for chondrules from 1-791324 (LL3.1).					
Chondrule#	Phase	²⁷ Al/ ²⁴ Mg	$^{26}Mg/^{24}Mg$	Initial ²⁶ Al/ ²⁷ Al		
Y-791324 (LL3.1)						
6	Olivine	0.02 ± 0.01	0.13932 ± 11			
(IIAB)	Glass	39.4 ± 2.5	0.13980 ± 17	$(1.22 \pm 0.52) \times 10^{-5}$		
3	Olivine	0.02 ± 0.01	0.13932 ± 12			
(IIA)	Glass	42.4 ± 3.0	0.13978 ± 42	$(1.1 \pm 1.0) \times 10^{-5}$		

Table 4.4: Al-Mg isotope data for chondrules from Y-791324 (LL3.1).

Table 4.5: Al-Mg isotope data for chondrules from ALHA76004 (LL3.3).

Chondrule#	Phase	²⁷ Al/ ²⁴ Mg	²⁶ Mg/ ²⁴ Mg	Initial ²⁶ Al/ ²⁷ Al
		ALHA76004	(LL3.3)	
1	Olivine	0.006 ± 0.003	0.13930 ± 09	
(IIAB)	Plagioclase	40.1 ± 2.3	0.13981 ± 32	
	Plagioclase	51.6 ± 1.4	0.13990 ± 30	
	Plagioclase	66.3 ± 2.3	0.13993 ± 33	
	Plagioclase	73.9 ± 1.7	0.14005 ± 45	$(1.06 \pm 0.33) \times 10^{-5}$
2	Olivine	0.004 ± 0.002	0.13932 ± 08	
(IIAB)	Glass	28.7 ± 1.3	0.13965 ± 10	$(1.15 \pm 0.34) \times 10^{-5}$
11	Olivine	0.016 ± 0.012	0.13933 ± 09	
(IIAB)	Glass	54.7 ± 9.2	0.13979 ± 42	
	Glass	73.2 ± 2.3	0.13981 ± 33	
	Glass	100.7 ± 6.0	0.14000 ± 31	
	Glass	143.8 ± 19.8	0.14037 ± 38	$(7.1 \pm 2.0) \times 10^{-6}$
31	Olivine	0.016 ± 0.006	0.13932 ± 07	
(IIAB)	Glass	27.1 ± 1.4	0.13965 ± 18	
	Glass	39.1 ± 1.9	0.13983 ± 24	
	Glass	56.0 ± 3.3	0.13997 ± 25	
	Glass	83.8 ± 9.9	0.14020 ± 32	$(1.14 \pm 0.28) \times 10^{-5}$



Figure 4.6: Al-Mg isotope data for Y-791324 chondrule CH#6 (top) and CH#3 (bottom).



Figure 4.7: Al-Mg isotope data for ALHA76004 chondrule CH#1 (top) and CH#2 (bottom).



Figure 4.8: Al-Mg isotope data for ALHA76004 chondrule CH#11 (top) and CH#31 (bottom).

4.3 Al-Mg isotope data from L-chondrites (Finds)

All the four UOCs of L group, LEW86134 (L3.0), QUE97008 (L3.05), LEW86018 (L3.1) and ALHA77176 (L3.2), analyzed in this study are "finds" from Antarctica. Results obtained from studies of Al-Mg isotope systematics in these four UOCs are presented in the following.

4.3.1 LEW86134 (L3.0)

Al-Mg isotope data for four ferromagnesian chondrules selected for isotope studies are presented in Table 4.6 and the Al-Mg isotope plots are shown in Figures 4.9 and 4.10. The range of ²⁷Al/²⁴Mg ratio in the analyzed phases was rather low, between 20 to 60, and it was not possible to obtain very precise initial ²⁶Al/²⁷Al for chondrules in this sample. The inferred initial ²⁶Al/²⁷Al also varies over a wide range (Table 4.6, Figures 4.9 and 4.10). One of the chondrule (CH#36A) is devoid of resolved ²⁶Mg excess, while another chondrule (CH#36) has a value of $(1.63 \pm 0.72) \times 10^{-5}$. The presence of chondrule devoid of detectable ²⁶Al was not expected in this meteorite belonging to the lowest petrologic grade. Average chromium content in olivine in this chondrule (Figure 3.10) is similar to the other chondrules in this meteorites.

4.3.2 QUE97008 (L3.05)

A total of eighteen chondrules were checked for suitability of Al-Mg isotope studies in this meteorite and we finally selected four chondrules having phases with reasonably high Al/Mg ratio. Data obtained for these chondrules are given in Table 4.7 and the Al-Mg isotope data are shown in Figures 4.11 and 4.12. All the four chondrules have ²⁶Mg excess. Even though the inferred initial ²⁶Al/²⁷Al ratios are not precise, one of the chondrules has the highest initial ratio of $(1.95 \pm 0.76) \times 10^{-5}$, amongst the UOC chondrules from both L and LL groups analyzed in this study.

Chondrule#	Phase	²⁷ Al/ ²⁴ Mg	$^{26}Mg/^{24}Mg$	Initial ²⁶ Al/ ²⁷ Al
		LEW86134	(L3.0)	
3	Olivine	0.006 ± 0.008	0.13927 ± 13	
(IIAB)	Glass	42.0 ± 3.4	0.13952 ± 49	
	Glass	56.8 ± 4.1	0.13968 ± 30	$(7.1 \pm 5.4) \times 10^{-6}$
16	Pyroxene	0.014 ± 0.009	0.13931 ± 13	
(IIB)	Glass	36.1 ± 1.3	0.13972 ± 24	
	Glass	40.6 ± 1.0	0.13978 ± 25	
	Glass	45.2 ± 1.1	0.13974 ± 33	
	Glass	54.1 ± 1.8	0.13981 ± 35	$(1.04 \pm 0.42) \times 10^{-5}$
36	Olivine	0.008 ± 0.007	0.13926 ± 17	
(IIA)	Glass	33.2 ± 2.3	0.13981 ± 37	
	Glass	42.2 ± 1.8	0.13990 ± 48	
	Glass	56.1 ± 5.7	0.14021 ± 54	$(1.63 \pm 0.72) \times 10^{-5}$
36A	Olivine	0.006 ± 0.003	0.13931 ± 16	
(IIAB)	Glass	40.5 ± 2.5	0.13934 ± 43	
	Glass	54.3 ± 4.9	0.13909 ± 47	$(-2.2 \pm 7.5) \times 10^{-6}$

Table 4.6: Al-Mg isotope data for chondrules from LEW86134 (L3.0).

4.3.3 LEW86018 (L3.1)

Thirteen chondrules were selected for analyses from this meteorite and four of these have phases with a good spread in Al/Mg ratio (up to 200) suitable for our studies. While in three chondrules Al-rich glassy phases were analyzed, in the fourth chondrule the analyzed phases have plagioclase composition. All the four chondrules yielded data with reasonable precision; these are given in Table 4.8. The Al-Mg isotope plots are shown in Figures 4.13 and 4.14. The data yielded initial 26 Al/ 27 Al values of (7.4 ± 2.0)×10⁻⁶, (6.5 ± 2.6)×10⁻⁶, (6.7 ± 3.6)×10⁻⁶ and (7.6 ± 3.6)×10⁻⁶, respectively, that are indistinguishable within error limits. A weighted mean value of (7.1 ± 1.3)×10⁻⁶ may be obtained for initial 26 Al/ 27 Al values by combining this data set.

Chondrule#	Phase	²⁷ Al/ ²⁴ Mg	²⁶ Mg/ ²⁴ Mg	Initial ²⁶ Al/ ²⁷ Al
QUE97008 (L3.05)				
1A	Olivine	0.1 ± 0.1	0.13934 ± 13	
(IIA)	Plagioclase	41.5 ± 3.5	0.13974 ± 30	
	Plagioclase	59.9 ± 2.7	0.13993 ± 48	$(9.9 \pm 6.0) \times 10^{-6}$
1B	Olivine	0.003 ± 0.0015	0.13933 ± 16	
(IIA)	Plagioclase	36.5 ± 3.0	0.14009 ± 57	
	Plagioclase	43.7 ± 3.1	0.14013 ± 61	
	Plagioclase	48.6 ± 0.6	0.14029 ± 42	$(1.95 \pm 0.76) \times 10^{-5}$
5	Olivine	0.0004 ± 0.00003	0.13935 ± 11	
(IIA)	Glass	50.5 ± 4.2	0.13973 ± 40	
	Glass	74.5 ± 5.6	0.13990 ± 44	
	Glass	91.9 ± 5.5	0.14012 ± 43	$(7.9 \pm 3.6) \times 10^{-6}$
16	Olivine	0.10 ± 0.011	0.13934 ± 21	
(IIAB)	Glass	27.1 ± 0.8	0.13967 ± 38	
	Glass	34.7 ± 6.4	0.13965 ± 52	
	Glass	75.4 ± 3.6	0.14013 ± 88	$(1.1 \pm 0.92) \times 10^{-5}$

Table 4.7: Al-Mg isotope data for chondrules from QUE97008 (L3.05).

4.3.4 ALHA77176 (L3.2)

Most of the chondrules in this meteorite are characterized by phases having low Al/Mg ratio and studies of Al-Mg isotope systematics was carried out only in two chondrules. Even in these cases, the Al/Mg ratio was \leq 50. Although both of them have resolved ²⁶Mg excess, the inferred initial ²⁶Al/²⁷Al values have large errors; (6.5 \pm 5.8)×10⁻⁶ and (9.7 \pm 7.1)×10⁻⁶. The data for these chondrules are given in Table 4.9 and the Al-Mg isotope plot are shown in Figure 4.15.



Figure 4.9: Al-Mg isotope data for LEW86134 chondrules CH#3 (top) and CH#16 (bottom).



Figure 4.10: Al-Mg isotope data for LEW86134 chondrules CH#36 (top) and CH#36A (bottom).

Chondrule#	Phase	²⁷ Al/ ²⁴ Mg	²⁶ Mg/ ²⁴ Mg	Initial ²⁶ Al/ ²⁷ Al
LEW86018 (L3.1)				
6	Olivine	0.01 ± 0.01	0.13929 ± 15	
(IIA)	Glass	48.8 ± 5.9	0.13975 ± 36	
	Glass	68.4 ± 4.9	0.13982 ± 26	
	Glass	98.2 ± 7.4	0.14010 ± 34	
	Glass	201.7 ± 29.8	0.14072 ± 34	$(7.4 \pm 2.0) \times 10^{-6}$
6A	Olivine	0.005 ± 0.001	0.13931 ± 13	
(IIA)	Glass	82.9 ± 7.2	0.13990 ± 26	
	Glass	121.3 ± 12	0.14026 ± 42	
	Glass	216.5 ± 23.8	0.14035 ± 74	$(6.5 \pm 2.3) \times 10^{-6}$
24	Olivine	0.0006 ± 0.0007	0.13930 ± 16	
(IIAB)	Glass	42.1 ± 4.9	0.13973 ± 43	
	Glass	54.9 ± 1.2	0.13977 ± 39	
	Glass	80.7 ± 5.7	0.13984 ± 37	
	Glass	114.6 ± 18.2	0.13996 ± 54	$(6.7 \pm 3.6) \times 10^{-6}$
33	Olivine	0.02 ± 0.01	0.13932 ± 14	
(IIAB)	Plagioclase	53.4 ± 13.1	0.13984 ± 53	
	Plagioclase	124.5 ± 22.1	0.14024 ± 45	$(7.6 \pm 3.6) \times 10^{-6}$

Table 4.8: Al-Mg isotope data for chondrules from LEW86018 (L3.1).

	Table 4.9: Al-Mg	isotope data	for chondrules	from ALHA77176	(L3.2)
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Chondrule#	Phase	²⁷ Al/ ²⁴ Mg	²⁶ Mg/ ²⁴ Mg	Initial ²⁶ Al/ ²⁷ Al	
ALHA77176 (L3.2)					
1	Olivine	0.07 ± 0.06	0.13932 ± 13		
(IIAB)	Glass	29.9 ± 1.5	0.13961 ± 33		
	Glass	37.4 ± 1.6	0.13954 ± 39		
	Glass	52.0 ± 3.9	0.13962 ± 34	$(6.5 \pm 5.8) \times 10^{-6}$	
14B	Olivine	0.04 ± 0.02	0.13924 ± 16		
(IIAB)	Glass	24.7 ± 1.2	0.13933 ± 20		
	Glass	30.3 ± 1.4	0.13956 ± 22		
	Glass	37.8 ± 1.0	0.13973 ± 34	$(9.7 \pm 7.1) \times 10^{-6}$	



Figure 4.11: Al-Mg isotope data for QUE97008 chondrules CH#1A (top) and CH#1B (bottom).



Figure 4.12: Al-Mg isotope data for QUE97008 chondrules CH#5 (top) and CH#16 (bottom).



Figure 4.13: Al-Mg isotope data for LEW86018 chondrules CH#6 (top) and CH#6A (bottom).



Figure 4.14: Al-Mg isotope data for LEW86018 chondrules CH#24 (top) and CH#33 (bottom).


Figure 4.15: Al-Mg isotope data for ALHA77176 chondrules CH#1 (top) and CH#14B (bottom).

CHAPTER 5

Discussion

In this chapter, the implications of the results obtained from studies of Al-Mg isotope systematics in chondrules from UOCs belonging to low petrologic grades (3.0 - 3.3) are discussed in terms of the time of onset and duration of chondrules formation in the early solar system. Combining this information with literature data for CAIs, the life time of the dynamically active solar nebula during which CAIs and chondrules were formed is inferred and compared with previous estimates. The question of possible thermal metamorphism affecting the observed ²⁶Al records in chondrules during their residence in the UOC parent bodies and its implications are also addressed.

The results obtained in this study have revealed the following important features:

- (i) All the twenty nine chondrules except one studied from nine UOCs of low petrologic grade have shown resolved radiogenic ²⁶Mg.
- (ii) The range of initial ²⁶Al/²⁷Al in the analyzed chondrules varies from 1.95×10^{-5} to $<10^{-6}$ and most of them lie in the range of 5×10^{-6} to 1.2×10^{-5} .
- (iii) Absence of resolved ²⁶Mg excess in a chondrule from LEW86134 (L3.0) suggests

possible disturbance in Al-Mg isotope systematics in chondrules from UOCs belonging to low petrologic grades.

(iv) No significant difference in the spread of initial ²⁶Al/²⁷Al is seen in chondrules from UOCs belonging to both L and LL type having petrologic grades ranging from 3.0 to 3.3.

A brief summary of our present understanding of the sources of the short-lived radionuclide, including ²⁶Al, present in the early solar system and distribution of ²⁶Al in the solar nebula is presented before discussing the results obtained in this study.

Isotopic studies of meteorites provide evidence for the presence of several shortlived radionuclides (Table 1.1 of Chapter 1) in the early solar system. Two hypotheses were put forward to explain the presence of these short-lived radionuclides: (i) they are products of solar energetic particle (SEP) interaction with gas and dust in solar nebula and (ii) they are freshly synthesized nuclides from a stellar source injected into the protosolar cloud. Energetic solar particle interaction was proposed initially by Heymann and Dziczkaniec (1976) as a source of ²⁶Al found in CAIs (Lee et al., 1976). Later studies by Wasserburg and Arnold (1987) clearly showed that SEP production cannot concurrently account for the observed abundances of ²⁶Al and ⁵³Mn with the same set of parameters for SEP flux and irradiation duration. It is now well established that SEP interaction cannot co-produce the short-lived radionuclides, ⁴¹Ca, ²⁶Al, ³⁶Cl, and ⁵³Mn to match their initial abundances in the solar system (Goswami et al., 2001; Goswami et al., 2005). Attempt to match the observed initial ²⁶Al/²⁷Al ratio of 5×10⁻⁵ in CAIs will lead to overproduction of the other short-lived radionuclides. The SEP irradiation is also inefficient in producing ⁶⁰Fe and falls short by several orders of magnitude to match its inferred initial abundance in the solar system (Tachibana and Huss, 2003; Mostefaoui et al., 2005; Tachibana et al., 2006). A proposal by Shu et al. (1996) for SEP production of shortlived radionuclides gained importance following the detection of radiogenic ¹⁰B

from the short-lived nuclide ¹⁰Be, which is not a product of stellar nucleosynthesis, in early solar system objects (McKeegan et al., 2000a). However, a combined study of ²⁶Al and ¹⁰Be records in early solar system objects (Marhas et al., 2002) showed that the production of these two nuclides are decoupled. Earlier studies by Sahijpal et al. (1998, 2000) also showed that ²⁶Al and ⁴¹Ca are cogenetic. These facts and the presence of ⁶⁰Fe, a purely stellar nucleosynthesis product, provide convincing argument for a stellar source as the most viable source of short-lived radionuclides (⁴¹Ca, ²⁶Al, ⁶⁰Fe) present in the early solar system. However, a small contribution $(\leq 10\%)$ from SEP interactions to the inventory of some of these nuclides cannot be ruled out (Goswami et al., 2001). A stellar origin for ²⁶Al allow us to use it as a chronometer to infer relative timescales of events in the early solar system. The use of ²⁶Al as a chronometer assumes that it was homogenously distributed in the nebula. This assumption is supported by the facts that (i) the observed ²⁶Al/²⁷Al ratio in most of the petrographically undisturbed CAIs from diverse chondrite classes are tightly clustered around $(4 - 5) \times 10^{-5}$ (MacPherson et al., 1995), (ii) the agreement between the inferred time scales for various events in the early solar system based on both short-lived (e.g., ²⁶Al, ¹⁸²Hf) and long-lived (^{235,238}U) isotopes as chronometers (Halliday and Kliene, 2006) and (iii) the uniform Mg isotope composition in samples of Earth, Moon and Mars (Thrane et al., 2006). Recent analytical modeling also suggests a very short time scale for homogenization of the short-lived nuclides within the solar nebula following their injection from a stellar source (Boss, 2007).

5.1 Onset of chondrule formation

Our data for the twenty nine chondrules from nine UOCs belonging to the L and LL group have nearly doubled the data set for Al-Mg isotope systematics in UOC chondrules. The initial 26 Al/ 27 Al of the UOC chondrules obtained in this study show a range of 1.95×10^{-5} to $<10^{-6}$. The highest initial 26 Al/ 27 Al is clearly below

the canonical value of 5×10^{-5} for CAIs considered to be the first objects to form in the solar nebula. A couple of chondrules have initial ²⁶Al/²⁷Al significantly above ~10⁻⁵: (1.63±0.72)×10⁻⁵ [LEW86134 (L3.0); CH#36] and (1.95±0.76)×10⁻⁵ [QUE97008 (L3.05); CH#1B] (Figures 4.10 and 4.11; errors are 2σ). The highest initial 26 Al/ 27 Al in chondrules from other UOCs are as follows: Semarkona (LL3.0): (1.24 ± 0.52)×10⁻⁵; Bishunpur (LL3.1): (5.8±2.2)×10⁻⁶; Y-791324 (LL3.1): (1.22±0.52)×10⁻⁵; LEW86018 (L3.1): $(7.6 \pm 3.6) \times 10^{-6}$; ALHA77176 (L3.2): $(9.7 \pm 7.1) \times 10^{-6}$; Adrar-003 (L/LL3.1): $(8.4 \pm 7.8) \times 10^{-6}$ and ALHA76004 (LL3.3): $(1.15 \pm 0.34) \times 10^{-5}$, respectively (all errors quoted are 2σ). Most of these values overlap with each other within errors. Data reported earlier for chondrules from UOCs of low petrologic grades (3.0 - 3.4) belonging to the LL group, suggest the following values for the highest observed initial 26 Al/ 27 Al in them: (9±1.6)×10⁻⁶ (Semarkona (LL3.0); Kita et al., 2000), (2.28±0.73)×10⁻⁵ (Bishunpur (LL3.1); Mostefaoui et al., 2002), (1.1±0.7)×10⁻⁵ (Inman (LL3.3); Russell et al., 1996), (9.2±2.4)×10⁻⁶ (Chainpur (LL3.4); Russell et al., 1996). It may be noted that the high value of $(2.28\pm0.73)\times10^{-5}$ Bishunpur chondrules reported by Mostefaoui et al. (2002) was based on analysis of phases with low Al/Mg ratio and needs confirmation (Kita et al., 2005b). Thus, if we consider all the data in totality, a value of 1.2×10^{-5} appears to be the representative upper limit for a majority of chondrules and suggest the presence of a very small fraction (<10%) of

The present study suggest onset of chondrule formation when the initial 26 Al/ 27 Al value in the solar nebula dropped from the canonical value of 5×10^{-5} , seen in CAIs, to 2×10^{-5} . Given the half life of 26 Al (=0.72Ma), this would imply onset of chondrule formation in the solar nebula, at least 1 Ma after CAI formation, a conclusion also reached in earlier studies of chondrules from UOC belonging to LL group (Kita et al., 2000, 2005a; Huss et al., 2001; Mostefaoui et al., 2002). However, our data also suggest a representative upper limit of 1.2×10^{-5} for initial 26 Al/ 27 Al ratio for most of the chondrules from UOC belonging to both L and LL group of

chondrules with initial 26 Al/ 27 Al ratio between 1.2×10⁻⁵ and 2×10⁻⁵ (Figure 5.1)

low petrologic grades (3.0 to 3.3). This would indicate that even though initiation of chondrule formation took place about a Ma after CAI formation, an intense episode of chondrule formation started 0.5 Ma afterwards. The alternative possibility that the representative upper limit of 1.2×10^{-5} for initial ²⁶Al/²⁷Al in a majority of UOC chondrules could be due to a thermal resetting event affecting a majority of the chondrules is a difficult proposition as realistic nebular process that may lead to resetting of Al-Mg isotope records in nearly all the pre-existing chondrules, widely distributed within the inner solar nebula, as individual entities or as constituent of



Figure 5.1: Histogram of initial ²⁶Al/²⁷Al ratio for UOC chondrules analyzed in this study and reported earlier by other groups. [Data source: Semarkona (LL3.0): Kita et al. (2000); McKeegan et. al. (2000b) and Huss et al. (2001), Bishunpur (LL3.1): Mostefaoui et al. (2002) and McKeegan et al. (2000b), Chainpur (LL3.4): Russell et al. (1996) and Huss et al. (2001), Inman (LL3.3), Quinyambie (LL3.4) and Krymka (LL3.1): Huss et al (2001)].

much larger sized objects, is very difficult to perceive. A late formation of chondrules relative to CAIs with a time difference of one to two million years is also supported by Pb-Pb isochron age of chondrules in CR chondrite Acfer 059 (4564.7 \pm 0.6 Ma) and CAIs in the CV chondrite Efremovka (4567.2 \pm 0.6 Ma) indicating a time interval of 2.5 \pm 1.2 Ma between formation of CV CAIs and the CR chondrules (Amelin et al., 2002).

A late formation of chondrules relative to CAIs with a time difference of more than a million years is also supported by Al-Mg isotope data obtained for chondrules from carbonaceous chondrites, using ion microprobe technique (Sheng et al., 1991; Hutcheon and Jones, 1995; Krot et al., 1999; Srinivasan et al., 2000; Marhas et al., 2000; Hutcheon et al., 2000; Kunihiro et al., 2004; Kurahashi et al., 2004). Resolved excess in ²⁶Mg due to ²⁶Al decay is present in CV3 meteorites, Allende, the unique meteorite Acfer 094 (Hutcheon et al., 2000), Efremovka (Marhas et al., 2000; Hutcheon et al., 2000; Srinivasan et al., 2000) and Yamato-81020 (Kunihiro et al., 2004; Kurahashi et al., 2004). The highest initial 26 Al/ 27 Al inferred are: $(6.5 \pm 3.5) \times 10^{-6}$ (Yamato-81020; CO3.0), $(3.4 \pm 1.5) \times 10^{-6}$ (Axtell; CV3), $(9.5 \pm 5.8) \times 10^{-6}$ (Efremovka; CV), $(1.2 \pm 0.4) \times 10^{-5}$ (Acfer094; unique), $(6 \pm 2.6) \times 10^{-6}$ (EET92174; CR), $(6.8 \pm 2.4) \times 10^{-6}$ (Allende; CV). The data for CC chondrules suggest that the highest initial ²⁶Al/²⁷Al ratio in chondrules from UOCs belonging to the LL and L group with petrologic grade 3.0 to 3.3 obtained in this study and by others (Russell et al., 1996; Kita et al., 2000; Huss et al., 2001; Mostefaoui et al., 2002) is nearly similar to that for chondrules from carbonaceous chondrite belonging to CV, CO and CR group within experimental uncertainties. If we exclude the few UOC chondrules with initial ²⁶Al/²⁷Al significantly greater than 10⁻⁵, the onset of an intense chondrule formation epoch ~1.5 Ma after formation of CAIs proposed by us based on data from UOC chondrules is also supported by the data from CC chondrules.

Although a late formation of chondrules, relative to CAIs is supported by Pb-Pb dating of CAI and chondrules from carbonaceous chondrites (Amelin et al., 2002),

a recent study by Bizzarro et al. (2004), who have analyzed Allende CAIs and chondrules using multi-collector inductively coupled plasma mass spectrometry (MC-ICPMS) technique, indicated that the onset of chondrule formation is contemporaneous with that of CAIs and continued for ~1.5 Ma. However, this conclusion needs closer scrutiny as the initial ²⁶Al/²⁷Al inferred from the MC-ICPMS data are for bulk chondrule material, rather than for individual Al-rich constituent phases as in this study. The inferred ratios could therefore represent the time of bulk Al-Mg fractionation, possibly during the formation of chondrule precursor dust grains, and may refer to the time of the formation of chondrule precursors rather than the chondrules themselves (Krot et al., 2005). The overall data for Al-Mg isotope systematics in meteoritic chondrules obtained in this study and those reported previously do support the view that initiation of cAIs and the data for UOC and CC chondrules suggest that an intense chondrule formation process started another half a million years later.

5.2 Duration of chondrule formation

The spread in the distribution of initial 26 Al/ 27 Al in meteoritic chondrules should ideally provide a measure of the duration of chondrule formation, if one can rule out any secondary perturbation affecting the Al-Mg isotope system in these objects. An accurate estimation of the duration of chondrule formation was not successful till now due to the small number of chondrules analyzed, the lack of high precision data and the possibility of thermal metamorphism experienced by some of the analyzed chondrules in a pre-parent/parent body environment. Our data for chondrules from nine UOCs belonging to low petrologic grades (3.0 - 3.3) suggest that the spread in initial 26 Al/ 27 Al values fall within a narrow range of (0.5 - 1.2)×10⁻⁵, excluding a lone chondrule that is devoid of excess 26 Mg, and two chondrules with relatively high

initial ²⁶Al/²⁷Al of $(1.63 \pm 0.72) \times 10^{-5}$ and $(1.95 \pm 0.76) \times 10^{-5}$, respectively (Table 5.1 and Figure 5.2). This is true even if we consider data obtained in previous studies of UOC chondrules (Figures 5.1 and 5.3) belonging to low petrologic grades. This narrow range suggests that large scale chondrule forming processes were operating in the early solar system for less than a Ma.

Earlier studies of Al-Mg isotope systematics in UOC chondrules (Russell et al., 1996; Kita et al., 2000; Huss et al., 2001; Mostefaoui et al., 2002) were done on Al-rich chondrules from meteorites such as Inman (LL3.3), Chainpur (LL3.4), Quinyambie (LL3.4) and ferromagnesian chondrules from the Semarkona (LL3.0) and Bishunpur (LL3.1) yielded durations of chondrule formation varying from 1 Ma to >3 Ma. The initial study of UOC chondrules by Russell et al. (1996) yielded initial ²⁶Al/²⁷Al ratios ranging from $<10^{-6}$ to $\sim10^{-5}$ in Al-rich chondrules from Chainpur (LL3.4). This was interpreted as a signature for an extended duration of >3 Ma for chondrule formation (Figure 5.3). The study of ferromagnesian chondrules from the UOC Semarkona (LL3.0) by Kita et al. (2000) indicated a short duration of <1 Ma for chondrule formation. However, a later study of Bishunpur (LL3.1) chondrules by the same group led them to suggest a relatively longer chondrule formation duration of ≥ 1 Ma (Mostefaoui et al., 2002). The above inferences were based on the expectation that Al-Mg isotope systematics in chondrules from UOCs belonging to petrologic grade 3.0 to 3.4 are not disturbed by thermal metamorphism experienced by them in their parent bodies (LaTourette and Wasserburg, 1998).

A couple of chondrules analyzed in this study have higher initial 26 Al/ 27 Al, but overlap with the other data within experimental uncertainty at 2σ level (Table 5.1). Thus, formation of a small number of UOC chondrules, during a period of ~0.5 Ma, followed by an intense chondrule forming episode lasting ~1 Ma may be inferred from our data. This would imply that the duration of UOC chondrule formation in the early solar system lasted ~1.5 Ma. This conclusion is also consistent with data reported by others for chondrules from UOC belonging to petrologic grades

$CI \Pi D$			
Meteorite	Chondrule#	Initial ²⁶ Al/ ²⁷ Al	age after CAI in Ma*
Semarkona	2	$(5.51 \pm 0.33) \times 10^{-6}$	2.29(-0.06/0.06)
(LL3.0)	20	$(1.10 \pm 0.24) \times 10^{-5}$	1.57(-0.21/0.26)
	21	$(1.15 \pm 0.54) \times 10^{-5}$	1.53(-0.40/0.66)
	36	$(1.24 \pm 0.52) \times 10^{-5}$	1.45(-0.36/0.56)
Bishunpur	45	$(5.8 \pm 3.1) \times 10^{-6}$	2.24(-0.44/0.79)
(LL3.1)	61	$(5.8 \pm 2.2) \times 10^{-6}$	2.24(-0.33/0.50)
Adrar-003	2	$(6.3 \pm 4.0) \times 10^{-6}$	2.15(-0.51/1.05)
(L/LL3.1)	3	$(8.4 \pm 7.8) \times 10^{-6}$	1.85(-0.68/2.74)
	8	$(4.3 \pm 4.2) \times 10^{-6}$	2.55(-0.71/3.91)
Y-791324	6	$(1.22 \pm 0.52) \times 10^{-5}$	1.47(-0.37/0.58)
(LL3.1)	3	$(1.1 \pm 1.0) \times 10^{-5}$	1.56(-0.67/2.40)
ALHA76004	1	$(1.06 \pm 0.33) \times 10^{-5}$	1.61(-0.28/0.39)
(LL3.3)	2	$(1.15 \pm 0.34) \times 10^{-5}$	1.53(-0.27/0.36)
	11	$(7.1 \pm 2.0) \times 10^{-6}$	2.03(-0.26/0.34)
	31	$(1.14 \pm 0.28) \times 10^{-5}$	1.54(-023/0.29)
LEW86134	3	$(7.1 \pm 5.4) \times 10^{-6}$	2.03(-0.59/1.49)
(L3.0)	16	$(1.04 \pm 0.42) \times 10^{-5}$	1.63(-0.35/0.54)
	36	$(1.63 \pm 0.72) \times 10^{-5}$	1.16(-0.38/0.61)
	36A	$(-2.2 \pm 7.5) \times 10^{-6}$	>3
QUE97008	1A	$(9.9 \pm 6.0) \times 10^{-6}$	1.68(-0.49/0.79)
(L3.05)	1B	$(1.95 \pm 0.76) \times 10^{-5}$	0.98(-0.34/0.51)
	5	$(7.9 \pm 3.6) \times 10^{-6}$	1.92(-0.39/0.63)
	16	$(1.1 \pm 0.92) \times 10^{-5}$	1.57(-0.63/1.88)
LEW86018	6	$(7.4 \pm 2.0) \times 10^{-6}$	1.98(-0.25/0.33)
(L3.1)	6A	$(6.5 \pm 2.3) \times 10^{-6}$	2.12(-0.31/0.45)
	24	$(6.7 \pm 3.6) \times 10^{-6}$	2.09(-0.45/0.80)
	33	$(7.6 \pm 3.6) \times 10^{-6}$	1.96(-0.40/0.67)
ALHA77176	1	$(6.5 \pm 5.8) \times 10^{-6}$	2.12(-0.66/2.32)
(L3.2)	14B	$(9.7 \pm 7.1) \times 10^{-6}$	1.70(-0.57/1.37)

Table 5.1: Initial ²⁶Al/²⁷Al and formation age of chondrules analyzed relative to CAIs.

*Initial 26 Al/ 27 Al ratio is taken as 5×10^{-5} for CAIs.



Figure 5.2: Initial ²⁶Al/²⁷Al obtained in this study for UOC chondrules. Data for chondrules from different UOCs are arranged in the increasing order of their petrologic grade. The "zero" of the time line is defined by the canonical CAI initial 26 Al/²⁷Al of 5×10⁻⁵.

3.0 to 3.3. However, as noted earlier, data for Chainpur (LL3.4) chondrules suggest a much longer duration for chondrule formation if we assume that the Al-Mg isotope records in these chondrules are undisturbed. We believe this assumption is not correct and provide our reasons for the same in the next section and exclude data for chondrules with petrologic grade \geq 3.4 in the remaining discussion on duration of chondrule formation.

The question of termination of chondrule formation in the solar nebula remains a



Figure 5.3: Initial ²⁶Al/²⁷Al values of UOC chondrules analyzed in this study (filled circles) and those reported by other groups (open cirlces). The meteorites are arranged in the increasing order of their petrologic grade.

contentious issue primarily due to the low precision in the inferred initial ²⁶Al/²⁷Al ratios that does not allow a very precise estimate. We believe, the precise data obtained in this study for the Semarkona chondrule (CH#2), where we have a large spread in Al/Mg ratio (~80 to ~2500) and a very well-defined Al-Mg isochron that yielded an initial ²⁶Al/²⁷Al value of $(5.51 \pm 0.33) \times 10^{-6}$ (Figure 5.5), provides the best measure for the time of termination of chondrule formation. We note that the highest Al/Mg ratio reported for analyzed phases in UOC chondrules studied earlier are: ~282 (Semarkona (LL3.0); Kita et al., 2000), ~171 (Bishunpur (LL3.1);

Mostefaoui et al., 2002), ~92 (Krymka (LL3.1); Huss et al., 2001), ~77 (Inman (LL3.3); Huss et al., 2001), ~87 and the inferred initial 26 Al/ 27 Al values in these chondrules had typical uncertainties of (\geq 20%) compared to ~6% in the Semarkona chondrule CH#2 obtained in this study. Data obtained for Semarkona chondrules by us and others (Figure 5.6) show that the spread in our data closely match the data of Kita et al. (2000), while data of Huss et al. (2001) and McKeegan et al. (2000b) that have larger experimental errors, overlap with our data within experimental uncertainties. This is also true for the data of Semarkona clast chondrule 10805-5CC1, reported by Hutcheon and Hutchison (1989). In spite of the large errors associated with initial



Figure 5.4: Al-Mg isotope data for all chondrules from UOC belonging to petrologic grade 3.0 to 3.3 analyzed by us [LEW86134 (L3.0), Semarkona (LL3.0), QUE97008 (L3.05), LEW86018 (L3.1), Y-791324 (LL3.1), Bishunpur (LL3.1), Adrar-003 (L/LL3.1), ALHA77176 (L3.2), ALHA764 (LL3.3)]. Data for CH#36A of LEW86134 (L3.0) devoid of ²⁶Al is not included. Also shown are Al-Mg evolution line for two representative values of initial ²⁶Al/²⁷Al that encompass most of the data points.



Figure 5.5: Al-Mg isotope data for Semarkona chondrules CH#2 (same as Figure 4.1).

²⁶Al/²⁷Al ratio in some cases, the overall data do support a short epoch (\leq 1 Ma) of Semarkona chondrule formation (marked by the band shown in Figure 5.6 in term of initial ²⁶Al/²⁷Al values). Based on data obtained in the present study (Figure 5.2 and 5.4), as well as those obtained in previous studies (Figure 5.3 and 5.7) for UOC chondrules, we can draw the following conclusions:

- (a) Initiation of chondrule formation ~1 Ma after formation of CAI, when initial 26 Al/ 27 Al in the nebula dropped to a value of ~2×10⁻⁵.
- (b) Start of an intense epoch of chondrule formation, ~0.5 Ma afterwards (initial ²⁶Al/²⁷Al ~1.2×10⁻⁵) that lasted ≤1 Ma (when initial ²⁶Al/²⁷Al dropped to the value of ~5×10⁻⁶).

The present understanding on the duration of chondrule formation is incomplete if we do not take into consideration data for chondrules from carbonaceous chondrites. The results obtained by various groups from analysis of Al-Mg isotope systematics in chondrules from the carbonaceous chondrites (CC) have shown a range of initial 26 Al/ 27 Al from ${}^{-10^{-5}}$ to ${}^{\leq}10^{-6}$ (Sheng et al., 1991; Srinivasan et al., 2000; Marhas et al., 2000; Hutcheon et al., 2000; Kunihiro et al., 2004; Kurahashi et al., 2004). The lower limit of ${}^{\leq}10^{-6}$ is based on data for chondrules that are devoid of resolved 26 Mg excess. In some of these cases, the analyzed phase was plagio-clase, that are expected to be immune to isotopic disturbance due to low degree of thermal metamorphic history experienced by carbonaceous meteorites such as the CR chondrites. The Al-Mg isotope systematics in the CC chondrules do not provide any clear correlation between their initial 26 Al/ 27 Al and secondary processes affecting their host meteorites. All the chondrites analyzed from CO Yamato-81020 belonging to petrologic grade 3.0, yielded 26 Al excess with initial 26 Al/ 27 Al ratios



Figure 5.6: Initial 26 Al/ 27 Al values of chondrules from the UOC Semarkona analyzed by us and others. Most of the data fall within the band of (5 - 12)×10⁻⁶.



Figure 5.7: Al-Mg isotope data for chondrules from UOCs with petrologic grades 3.0 to 3.4 analyzed by us and others groups. Also shown are Al-Mg evolution lines for two representative values of initial ²⁶Al/²⁷Al. The encircled region includes data for chondrules devoid of resolved ²⁶Mg reported by other groups for UOCs of higher petrologic grades such as Chainpur (LL3.4).

ranging between $(3.7 \pm 1.1) \times 10^{-6}$ and $(6.5 \pm 3.2) \times 10^{-6}$ (Kunihiro et al., 2004), that are lower than typical values in UOCs chondrules. Initial ²⁶Al/²⁷Al in chondrules from CR chondrites that are more pristine than CV chondrite (Krot et al., 2005) and where many chondrules have well defined plagioclase, yielded initial ²⁶Al/²⁷Al ratios much lower than those obtained for CV chondrules that show a spread (Hutcheon et al., 2007) similar to those seen in UOC chondrules. It appears that duration of formation of CC chondrules lasted for a longer period than that for UOC chondrules, although the initiation of chondrule formation took place nearly at the same time in both cases.

5.3 Thermal metamorphism: possible effect on ${}^{26}\text{A}l/{}^{27}\text{A}l$ record in UOC chondrules

Even though most of the UOC chondrules analyzed in this study and those reported previously have initial ²⁶Al/²⁷Al in the range of 0.5×10^{-5} to $\le 2 \times 10^{-5}$ there are notable exceptions, particularly for chondrules from UOCs belonging to petrologic grade 3.4. This was one of the primary considerations that led to our selection of UOCs of petrologic grades between 3.0 to 3.3. UOCs belonging to petrologic grade 3.4 (e.g. Chainpur) are not expected to experience temperature greater than 450 °C in their parent body environment and one does not expect diffusion of Mg in anorthite over scale length of tens of microns even if such temperature persisted over a period of 20 Ma (LaTourette and Wasserburg, 1998). However, as already noted a majority of the Chainpur chondrules analyzed to date are devoid of resolved ²⁶Mg excess (Figure 5.3). Further, the absence of resolved radiogenic ²⁶Mg in one chondrule from the UOC LEW86134 (L3.0) analyzed in this study indicates that Al-Mg isotope system in rare chondrules, even from UOCs belonging to very low petrologic grades, may have been disturbed. Less precise low initial ²⁶Al/²⁷Al values of (0.33 ± 0.43)×10⁻⁵, (0.31 ± 0.68)×10⁻⁵ and (0.48 ± 0.45)×10⁻⁵ were also reported earlier for chondrules from UOCs of low petrologic grades such as Semarkona (LL3.0) and Krymka (LL3.1), respectively (McKeegan et al., 2000b; Huss et al., 2001). The presence of chondrules with relatively low initial ²⁶Al/²⁷Al is clearly evident in Figures 5.3 and 5.7, where we have included data for chondrules from the present study as well as those reported earlier including those for chondrules from UOCs belonging to petrologic grade 3.4. Thus, a reexamination of the possible effect of thermal metamorphism on the ²⁶Al records in UOC chondrules appears to be necessary. Such a possibility was proposed earlier by Huss et al. (2001) to account for the lower initial ²⁶Al/²⁷Al found in chondrules from Krymka (LL3.1), Quinyambie (LL3.4) and Chainpur (LL3.4). They also noted that presence of radiogenic ²⁶Mg in sodium rich

glass but not in anorthite present within Chainpur chondrules (Huss et al., 2001) suggest possible thermal affect on individual chondrules prior to their final incorporation into the host meteorite. However, no viable pre-parent body/parent body setting in which this may happen was proposed by these authors. The processes governing the dynamical evolution of the UOC chondrules during the time interval following their formation and their final incorporation into the parent meteorites, in the context of their ²⁶Al records, is not yet well understood. It is interesting to note that TL sensitivity, the primary indicator of degree of thermal metamorphism, varies by orders of magnitude in individual chondrules from both Semarkona and Bishunpur and they are all lower than that for the bulk meteorite (Sears et al., 1995). The observed variation in TL sensitivity is not related to either chondrule chemistry or the volume of the mesostasis. It is also not clear if these variations reflect imprints of the chondrule formation process itself or post formation thermal history experienced by chondrules in a pre-parent body or parent body environment. Finally, metamorphic temperatures inferred using various indicators such as TL sensitivity, presence of pre-solar grains (diamonds), degree of heterogeneity in olivine, amount of primordial ³⁶Ar, degree of matrix recrystallization or Cr distribution in olivine do not always give concordant results (Huss et al., 2006). A recalibration of metamorphic temperature experienced by UOCs belonging to different petrologic grades, based on studies of multiple indicators, as well as data for Mg diffusion in glass phase representative of chondrule mesostasis is needed for a better understanding of the observed ²⁶Al records in chondrules.

Our results as well as those reported earlier do suggest that Al-Mg isotope systematics in some individual chondrules from UOCs of lower petrologic grade may have been affected during their parent body residence prior to being a part of the host meteorite. In the following, we provide results obtained from modeling of thermal history of UOC parent bodies that sheds some new information on this issue.

Asteroids, the parent bodies of the meteorites, have undergone thermal evolution history during the first few million years of its formation, fueled primarily by the short-lived now-extinct radionuclides ²⁶Al and ⁶⁰Fe. We model this thermal evolution by solving the heat conduction equation numerically using finite difference method and using standard values for the density, decay constant, initial temperature, thermal conductivity, thermal diffusivity and heat capacity (Yomogida and Matsui, 1983). The extent of heating within a meteorite parent body depends on the abundances of ²⁶Al and ⁶⁰Fe at the time of its formation. ²⁶Al with its short half-life will be the dominant heat source in the initial stages. Since the size of the Al-rich phases (glassy mesostasis and plagioclase) in chondrules analyzed for Al-Mg isotope systematics in the present study and also by others are typically less than few tens of microns and laboratory studies suggest that Mg isotope records in plagioclase over such scale length can be thermally reset at a temperature of ~500 °C within a time period of about 1 Ma (LaTourette and Wasserburg, 1998), we tried to investigate the possibility of some individual UOC chondrules experiencing such a thermal history during their residence in meteorite parent bodies. Note that the temperature for resetting Al-Mg isotope records in glassy (mesostasis) phases may be even lower than this value. Calculations were carried out for meteorite parent body of 20 km in radius and of chondritic composition, with initial ²⁶Al/²⁷Al of 1.25×10^{-5} (formation time 1.44 Ma after CAI) and of 6.25×10^{-6} (formation time 2.16 Ma after CAI), and assuming instantaneous accretion. Our calculations show that the time-temperature regime (1 Ma, 500 °C) needed for disturbing Al-Mg isotope system in plagioclase will be experienced by an object residing at ≥ 2 km below the surface in such bodies (Figure 5.8). We therefore, propose that the chondrules with low initial ²⁶Al/²⁷Al ratio present in meteorites of petrologic grades 3.0 to 3.2 and whose number increases significantly in meteorites of petrologic grades 3.3 and 3.4, experienced such a thermal episode as an independent entity, perhaps due to impact related mixing on the parent body, prior to their final incorporation into the host

meteorite. Time-temperature-depth regime for resetting Al-Mg isotope system in glassy phases is expected to be less than that for plagioclase, and this can happen



Figure 5.8: Temperature-time profiles within a object of radius 20 km of chondritic composition at various depths (measured from the surface) due to ²⁶Al heating. The horizontal line marks 500 °C. Results for two initial values of ²⁶Al/²⁷Al ratio at the time of formation of the objects (T = 0) are shown. Al-Mg isotope systematics in plagioclase will be disturbed while residing at a depth of \geq 2 km (case a) and \geq 4 km (case b) below surface (within the shaded region) for periods of \geq 1 Ma.

even in shallower depths within 2 km from the surface. Further, if we consider contribution from the other heat source, ⁶⁰Fe as well, and also consider an extended regolith surface (with poor thermal conductivity) resetting of Al-Mg isotope records can take place in chondrules even further closer to the surface (Sandeep Sahijpal, personal communication). We conclude that parent body thermal processes can explain the presence of rare chondrules with low initial ²⁶Al/²⁷Al ratios in UOC of low petrologic grades (3.0 - 3.3) and their preponderance in UOC of petrologic grade 3.4 and higher.

5.4 LIFE TIME OF THE ACTIVE SOLAR NEBULA

The Ca-Al rich Inclusions (CAIs) and chondrules are generally considered as products of solar nebula processes. Thus the life time of a dynamically active solar nebula may be defined as the time from the start of CAI formation to the cessation of chondrule formation. We consider results obtained in this study and those reported previously on Al-Mg isotope systematics in CAIs and UOC chondrules to estimate the life time of an active solar nebula. Several groups have earlier followed a similar approach and suggested values ranging from ~2 Ma to ~5 Ma (Russell et al., 1996; Kita et al., 2000; Huss et al., 2001; Mostefaoui et al., 2002). Our data for Al-Mg isotope systematics in UOC chondrules coupled with the data for CAIs suggests that formation of CAIs preceded chondrule formation by at least 1 Ma and duration of the chondrule formation was ≤ 1.5 Ma, with the last million years being a period of intense chondrule formation. Thus, the lifetime of the dynamically active solar nebula was at least 2.5 Ma during which high temperature nebular events have taken place in the inner region of the solar nebula leading to formation of CAIs and UOC chondrules. The gap of about 1 Ma between CAIs and chondrules formation would require that the CAI must be incorporated into larger objects soon after their formation to stop them from spiraling into the sun under the influence of gas drag,

with a typical lifetime of a few times 10⁵ years or less (Weidenschilling, 1977), till the initiation of chondrule formation. One may propose a moderately turbulent solar nebula, instead of a quiescent nebula generally considered, to increase the nebular residence time of cm to millimeter sized CAIs and chondrules (Cuzzi et al., 2003). An alternate possibility is continuous formation of kilometer sized bodies during the early evolution of the solar system that will host the CAIs and chondrules and will be immune to such drag forces. Much longer duration of 3 - 5 Ma for an active solar nebula proposed in earlier studies will pose a serious problem for storage and mixing of CAIs and chondrules in the nebula and their final incorporation into meteorite parent bodies.

In contrast to the UOC chondrules, the low initial ²⁶Al/²⁷Al in many chondrules from primitive Carbonaceous chondrites that do not have petrographic evidence for thermal/aqueous alterations (as for e.g. CR chondrites) suggest a much longer duration of CC chondrule formation. Further studies will be needed to explore this difference between the duration of formation of chondrules in the inner (UOCs) and outer regions (CCs) of the asteroidal belt.

CHAPTER 6

Summary and conclusions

In this study ²⁶Al records in Al-rich phases (glass/ plagioclase) in chondrules from nine UOCs belonging to L and LL groups and having petrologic grades ranging from 3.0 to 3.3 have been investigated. The primary objective of the study was to further our understanding of the time of onset and duration of chondrule formation in the early solar system. This work also represents the first detail study of chondrules from UOCs belonging to the L group. A summary of results obtained and the conclusions that can be drawn from this study are outlined below.

- 1. All the twenty nine chondrules, barring a single exception, have resolved radiogenic ²⁶Mg.
- 2. The highest value of initial ²⁶Al/²⁷Al (~2×10⁻⁵) seen in the UOC chondrules analyzed in this study indicates that chondrule formation process started at least 1 Ma after the formation of CAIs, characterized by an initial ²⁶Al/²⁷Al of 5×10⁻⁵. This is consistent with results obtained in earlier studies that suggest that CAI formation preceded chondrule formation by 1 2 Ma.
- 3. Precise Al-Mg isotope data obtained for one Semarkona (LL3.0) chondrule

(CH#2) yielded a very well defined Al-Mg isochron with initial 26 Al/ 27 Al of $(5.51 \pm 0.33) \times 10^{-6}$. This value provides a robust time marker for the effective termination of UOC chondrule forming process.

- 4. ²⁶Al records in chondrules from UOCs belonging to both L and LL groups are similar.
- 5. The above results suggest that the duration of UOC chondrule formation spanned a time scale of ~1.5 Ma. This is significantly lower than the values of >3 Ma proposed in some earlier studies that included data for chondrules from UOCs belonging to petrologic grade 3.4
- 6. A majority of the chondrules (twenty six out of twenty nine) have initial 26 Al/ 27 Al spanning the range of (5 12)×10⁻⁶. This is also true for data reported earlier for UOCs of low petrologic grades (3.0 3.3) and suggests that an intense chondrule forming episode started ~0.5 Ma after the initiation of chondrule formation and lasted for ~1 Ma.
- The life time of an active solar nebula during which CAIs and UOC chondrules formed is ≤2.5 Ma, much shorter than earlier estimates of up to 5 Ma (e.g. Russell et al., 1996).
- 8. The occasional presence of chondrules with low initial ²⁶Al/²⁷Al <5×10⁻⁶, in UOCs belonging to low petrologic grades (3.0 to 3.3) and their preponderance in UOCs of higher petrologic grades most probably represent effect of thermal metamorphism in a meteorite parent body environment when these chondrules were residing at a depth of more than a kilometre prior to their incorporation into the host meteorites.

6.1 Scope for future work

The present study provides a well defined time frame for the time of onset and duration of formation of UOC chondrules. However, some unresolved issues remain and new questions arise that need further attention. Some of these are outlined in the following:

- ²⁶Al data for CAIs and chondrules clearly suggest a hiatus of ~1 Ma between CAI and chondrule formation. It is important to understand why nebular processes were not conducive to formation of either of these objects or if the records from this era are erased in chondrules by thermal processing in a nebular setting.
- 2. The data obtained in this study, combined with existing data for chondrules from carbonaceous chondrites, suggest that the duration of formation of UOC chondrules is significantly shorter than that for the CC chondrules, where low initial ²⁶Al/²⁷Al (<10⁻⁶) is seen in chondrules from primitive meteorites (e.g. CR group) that do not have any petrological signatures for secondary alteration. The reasons for this difference in the duration of chondrule formation in the inner and outer regions of the asteroidal belt where UOCs and CCs have formed is not well understood at present.
- 3. Even though a difficult proposition, ²⁶Al records in chondrules from H chondrites and enstatite chondrites should also be investigated to have a better understanding of both chondrule formation time scales as well as thermal perturbation of ²⁶Al records in chondrules. A focussed study looking for correlation between chondrule formation time and chondrule chemistry, suggested in an earlier study, will also be useful.
- 4. The lower initial ²⁶Al/²⁷Al seen in some chondrules from UOCs of low petrologic grades (<3.3) and more often in higher petrologic grades may be ascribed

to thermal metamorphism experienced by these chondrules in a nebula or parent body environment. However, we do not have adequate understanding of the post-formation evolutionary history of individual chondrules till the time they got incorporated into their host meteorites. Analytical modelling of this evolutionary scenario coupled with thermal history experienced by the chondrules are needed to delineate their post-formation evolution history.

- 5. The inferred duration of ~2.5 Ma for an active solar nebula, and an even longer duration if we consider the data for CC chondrules, poses the well known problem of storage and assimilation of CAIs and chondrules formed during different epochs leading finally to their close proximity within their host meteorites. A viable solution of this problem is yet to be found.
- 6. The well dated UOC chondrules provide an unique set of samples for studying the records of the short-lived nuclide ⁶⁰Fe in early solar system as the CAIs are not compositionally suitable for such a study. This will allow a robust determination of initial ⁶⁰Fe abundance in the early solar system needed to unequivocally identify the stellar source of ⁶⁰Fe and hence the contribution of this source to the inventory of the other short-lived nuclides, such as ²⁶Al, in the early solar system.

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List of publication

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- Rudraswami N. G. and Goswami J. N. (2007) ²⁶Al in chondrules from unequilibrated L chondrites: Onset and duration of chondrule formation in the early solar system, Earth Planet. Sci. Lett., 257, 231-244.
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- 1. **Rudraswami N. G.**, Deomurari M. P., and Goswami J. N. (2004) Al-Mg isotope systematics in ferromagnesian chondrules from the unequilibrated ordinary chondrite Adrar 003: time scale of chondrule formation, Lunar Planet. Sci. XXXV, abs. 1236.
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