Fe-Ni SYSTEMATICS IN Fe-RICH OLIVINE AND ENSTATITE CHONDRULES IN SEMARKONA CHONDRITE UTILIZING A NANOSIMS 50L ION MICROPROBE. M. Ito[1,2] and S. Messenger[1]. [1]Robert M. Walker Laboratory for Space Science, ARES, NASA JSC, 2101 NASA Parkway, Houston TX 77058, USA. [2]LPI-USRA, Houston TX 77058. (motoo.ito-1@nasa.gov, scott.r.messenger@nasa.gov).

Introduction: Now-extinct ^{60}Fe identified as ^{60}Ni excesses in meteorites and their components provides constraints on the formation time scales, nearby late-nucleosynthesis injection events, and the evolution of the early Solar System [e.g., 1,2]. The first clear evidence of excesses in ^{60}Ni from the decay of ^{60}Fe ($t_{1/2}$ = 2.62 Ma [3, was 1.5 Ma]) was reported in a eucrite meteorite that formed in a differentiated parent body [1]. Recently, ^{60}Ni excesses were found in Fe-rich silicates (pyroxene and olivine) [4-10], sulfides [11-17], and magnetites [16,17] in unequilibrated ordinary and enstatite chondrites, and a eucrite.

Sulfides are promising target minerals to measure excesses in ^{60}Ni because of their high Fe/Ni ratios. However, previous studies of Fe-Ni systematic in sulfides [11-17], and Fe diffusivity in FeS [18] and Ni in NiS [19] imply that Fe-Ni system in sulfides might be disturbed by thermal metamorphism (300~500ºC, 12,13) and/or aqueous alteration during parent body process [20]. Ni diffusion kinetics have been well studied in olivine [21,22] but not in pyroxene. The diffusion kinetics of Ni in olivine D_{Ni}(olivine) is slower than in sulfide $D_{Ni\ or\ Fe}$(sulfide) by 5 to 10 orders of magnitude at 500ºC. Tachibana and coworkers, thus, pointed out that silicates should be the least susceptible to isotopic disturbance by thermal alteration in the parent body of unequilibrated chondrites [5,12]. More recently, ferromagnesian pyroxene chondrules in UOCs were found to have disturbed Fe-Ni isochrons, suggesting partial resetting by alteration processes or different timing of metamorphic processes [10]. The diffusion kinetics of the Fe-Ni systematics in olivine, enstatite and sulfide plays an important role in the effectiveness of the Fe-Ni systematics in these minerals for a proper chronological interpretation.

In this study, we established a high-precision and high-spatial resolution technique for Fe-Ni systematics, and report observations of ^{60}Ni isotopic anomalies in Fe-rich olivine and Fe-rich enstatite chondrules in Semarkona unequilibrated ordinary chondrite (UOC) utilizing the JSC NanoSIMS 50L ion microprobe.

Experimental: A study of Fe-Ni systematics was performed in Fe-rich chondrules (olivines and enstatites) from UOC Semarkona (LL3.0). In this section, we found that three olivines were type II chondrule with mg#81-88, and enstatites showed some variations of mg# which are Type I (mg#93) and II (mg#80 and 85) chondrules (Fig. 1).

Fe and Ni isotopic measurements were carried out using the JSC NanoSIMS 50L ion probe. A focused primary O$^-$ beam of 1.5-2.0 nA was rastered over 15×15 to 20×20 μm areas on samples and standards. We evaluate the measurement conditions, the instrumental mass fractionation, relative sensitivity factor, and the precision and accuracy for Ni isotopic measurement through analyses of terrestrial standards of olivine and pyroxenes. Positive secondary ions of ^{57}Fe$^+$, ^{60}Ni$^+$, ^{61}Ni$^+$, and ^{62}Ni$^+$ were measured using four electron multipliers in multidetection mode at a high mass resolution of ~6,000 that is sufficient to separate all relevant isobaric interferences. We used beam blanking to minimize crater edge effects. In this mode, secondary ions from the edge (accounting for ~18% of the scanned area) are excluded from the dataset. Each measurement consisted of 100 cycles of with 30-120 sec/cycle (8,000-28,000 μs/pixel) counting time depending on Fe/Ni ratio, lasting 1-3 hours. Each run was started after stabilization of the secondary ion beam following 5 min of pre-sputtering. The sample was coated with a 12 nm C film to mitigate electrostatic charge on the sample surface. During the analysis, the mass peaks were centered automatically after every 10 cycles. Data were corrected for EM dead time (44 ns) and QSA effect. Excesses of ^{60}Ni are calculated a equation [11]: δ^{60}Ni (‰) = Δ^{60}Ni − (−Δ^{62}Ni), where $\Delta^{60\ or\ 62}$Ni (‰) = ($^{60\ or\ 62}$Ni/^{61}Ni)$_{sample}$/($^{60\ or\ 62}$Ni/^{61}Ni)$_{standard}$ − 1) x 1000. (^{m}Ni/^{61}Ni)$_{standard}$ values were 23.100 for m = 60, and 3.1760 for m = 62 [24].

Terrestrial standards of San Carlos olivine and enstatite were used to correct instrumental mass fractionation for Ni isotopes and a relative sensitivity factor
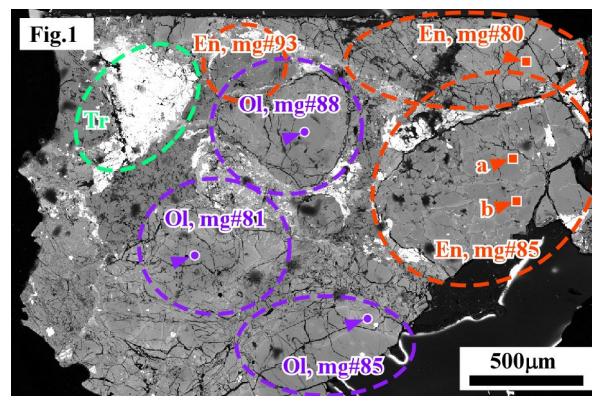

Fig. 1. BSE image of a section of Semarkona chondrite. En: enstatite, Tr: troilite, Ol: olivine

$[(Fe/Ni)_{true}/(Fe/Ni)_{NanoSIMS} = 1.06 \pm 0.01]$ for $^{56}Fe/^{61}Ni$ ratio for analyzed mineral phases in Semarkona chondrules.

Results and Discussions: We analyzed ^{60}Fe-^{60}Ni systematics in two pyroxene (mg#80 and 85) and three olivine (mg#81, 85 and 88) chondrules from Semarkona (Fig. 1). We found clear evidence for extinct ^{60}Fe in olivine and enstatite chondrules in Semarkona (Fig. 2). Data points from different chondrules lie within analytical errors on a straight line with an inferred $(^{60}Fe/^{56}Fe)_0$ ratio of $(6.2 \pm 2.2) \times 10^{-7}$ (2σ), and an initial $\delta^{60}Ni$ of 5.6 ± 7.5 (2σ). In terms of chronological interpretation, these chondrules formed within +- 1.5 Ma, based on the 2σ analytical error in the $(^{60}Fe/^{56}Fe)_0$ ratio. One measured point (mg#85a) from enstatite shows lower $\delta^{60}Ni$ value than others, possibly due to a later isotopic disturbance in enstatite during parent body metamorphism.

Table 1: $(^{60}Fe/^{56}Fe)_0$ of olivine and enstatite chondrules from Semarkona

	$(^{60}Fe/^{56}Fe)_0$	Instrument	REF
	3.1×10^{-7}	ims-6F	4
	$1.9 - 3.4 \times 10^{-7}$	ims-1270	5
Olivine or	$2.2 - 3.7 \times 10^{-7}$	ims-1270	6
Enstatite	2.9×10^{-7}	ims-4F	7, 9
Chondrules	$0.8 - 3.2 \times 10^{-7}$	ims-1280	8
	$1.7 - 4.8 \times 10^{-7}$	ims-4F & 1280	9

We summarize published initial $^{60}Fe/^{56}Fe$ ratios for olivine and enstatite chondrules from Semarkona in Table 1. Chondrules show $^{60}Fe/^{56}Fe$ ratios of $(0.8-4.8) \times 10^{-7}$ while $^{60}Fe/^{56}Fe$ ratios in troilites are, $(7.5-16.8) \times 10^{-7}$ [15-17], higher by a factor of 2 to 3 compared to chondrules. Our inferred initial $^{60}Fe/^{56}Fe$ ratio of 6.2×10^{-7} is slightly higher than the ratio previously observed in chondrules. This variation in the $^{60}Fe/^{56}Fe$ ratios among chondrules in Semarkona implies a long time scale for chondrule formation of 1 to 8 Ma. This is consistent with current chronological interpretation of chondrule formation that began ~1 Ma after CAI formation and persisted for several Ma [25,26].

There is some uncertainty in the initial $^{60}Fe/^{56}Fe$ ratio for the Solar System. Birck and Lugmair [23] proposed an initial $^{60}Fe/^{56}Fe$ ratio of $(1.6 \pm 0.5) \times 10^{-6}$, whereas and Quitté and coworkers [24] report a value of $(4.7 \pm 2.9) \times 10^{-6}$, both from the analysis of CAIs. The value we derive from our study of Semarkona chondrules, $(0.6-2.1) \times 10^{-6}$, is consistent with the lower value proposed by [23], and is based on the standard ^{60}Fe half life of 1.5 Ma. A recent proposed revision of the ^{60}Fe half life to 2.62 Ma yields an initial $^{60}Fe/^{56}Fe$ ratio of $(0.5-1.4) \times 10^{-6}$. Wasserburg and colleagues [27] proposed an initial $^{60}Fe/^{56}Fe$ ratio for the Solar System of $(\sim 10^{-7}$ to $2 \times 10^{-6})$ based on theoretical modeling of short-lived nuclei. Our estimated $^{60}Fe/^{56}Fe$ ratio is in

Fig.2 Semarkona
$^{60}Fe/^{56}Fe = (6.2 +- 2.2) \times 10^{-7}$
$\delta^{60}Ni = 5.6 +- 7.5$ per mil
Errors: 2σ
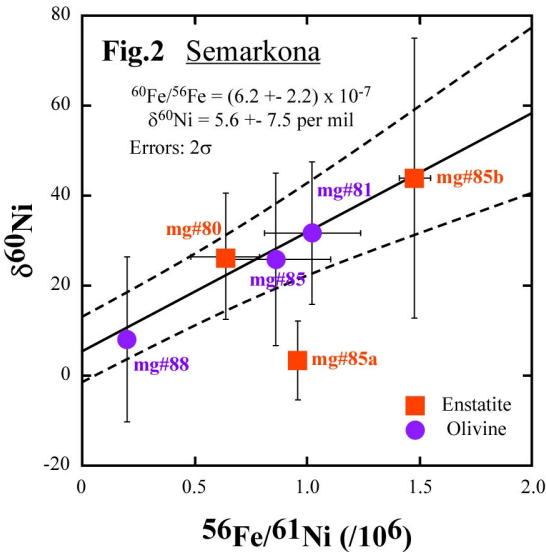
agreement with both the Wasserburg model and the ratio derived from CAI measurements.

References: [1] Shukolyukov A. and Lugmair G.W. (1993) *Science*, 259, 1138-1142. [2] Bizzarro M. et al. (2007) *Science* 316, 1178-1181. [3] Rugel G. et al. (2009) *Phys. Rev. Lett.*, 103, 072502. [4] Huss et al. (2004) *LPS* 35, 1811. [5] Tachibana S. et al. (2005) *LPS* 36, 1529. [6] Tachibana S. et al. (2006) *Astrophys. J.*, 639, L87-L90. [7] Goswami J.N. et al. (2007) *LPS* 38, 1943. [8] Tachibana S. et al. (2007) *LPS* 38, 1709. [9] Mishra R.K. et al. (2009) *LPS* 40, 1689. [10] Tachibana S. et al. (2009) *LPS* 40, 1808. [11] Tachibana S. and Huss G.R. (2003) *LPS* 34, 1737. [12] Tachibana S. and Huss G.R. (2003) *Astrophys. J.*, 588, L41-L44. [13] Guan Y. et al. (2003) *Meteoritics & Planet. Sci.,* 38, A138. [14] Guan Y. et al. (2004) *LPS* 35, 2003. [15] Mostefaoui S. et al. (2003) *LPS* 34, 1585. [16] Mostefaoui S. et al. (2004) *LPS* 35, 1271. [17] Mostefaoui S. et al. (2005) *Astrophys. J.*, 211, 271-277. [18] Condit R.H. et al (1974) *Oxid. Metals*, 8, 409-455. [19] Kotani K. et al. (1979) *Bull. Chem. Soc. Jpn.*, 52, 1531-1532. [20] Krot A.N. et al. (1997) *Geochim. Cosmochim. Acta*, 61, 219-237. [21] Petry C. et al. (2004) *Geochim. Cosmochim. Acta*, 68, 4179-4188. [22] Ito M. et al. (1999) *Phys. Chem. Mineral.*, 26, 425–431. [23] Birck J.L. and Lugmair G.W. (1988) *Earth Planet. Sci. Lett.*, 90, 131. [24] Quitté G. et al. (2007) *Astrophys. J.*, 655, 678-684. [25] Kita N.T. et al. (2000) *Geochim. Cosmochim. Acta*, 64, 3913-3922. [26] Russell et al. (1996) *Science* 273, 757–762. [27] Wasserburg G.J., et al. (2006) *Nucl. Phys.* A 777, 5-69.