GAS-MELT INTERACTION DURING CHONDRULE FORMATION. G. Libourel[1*], A. N. Krot[2] and L. Tissandier[1], [1]CRPG-CNRS 15, Rue Notre-Dame des Pauvres, BP20, 54501 Vandoeuvre les Nancy, France, [*]ENSG-INPL, BP40, 54501 Vandoeuvre les Nancy, France (libou@crpg-cnrs-nancy.fr), [2]Hawai'i Institute of Geophysics and Planetology, School of Ocean and Earth Science and Technology, University of Hawai'i at Manoa, Honolulu, HI 96822, USA.

Introduction: Although the mineralogy, bulk chemistry, isotopic compositions, and textural properties of chondrules are well-documented, the nature of the chondrule-forming events remains enigmatic [1,2]. In most chondrites, chondrules typically have porphyritic textures, with phenocrysts of olivine and low-Ca pyroxene set in glassy or microcrystalline mesostases; the latter typically contain quench crystals of high-Ca pyroxene. These igneous textures are consistent with crystal-growth from a rapidly cooling (10-1000 K hr^{-1}) silicate melt [3]. Chondrules are often surrounded by coarse-grained igneous rims and contain unmelted fragments of earlier generations of chondrules, suggesting that the chondrule-forming process was repetitive. Most chondrules contain relatively high contents of moderately volatile elements (Na, K, Mn, Cr); some contain primary sulfides [2,4]. Chondrules in unmetamorphosed chondrites are surrounded by fine-grained silicate rims which show no evidence for being melted. Based on these observations, it is generally believed that chondrules formed in cold (<1000 K) dusty regions of the solar nebula during localized, brief, repetitive heating events with peak temperatures in the range of ~1800-2100 K resulting in incomplete melting of solid precursors accompanied by limited evaporation-recondensation [2]. However, this nearly closed-system crystallization of chondrule melts is difficult to reconcile with the commonly observed mineralogical and chemical features of porphyritic magnesian (Type I) chondrules – the dominant chondrule type in most carbonaceous chondrites.

Experimental: We focus our survey on Type I chondrules (Fa and Fs <5 mol%) from the LL3.0 ordinary chondrite Semarkona and several CR2 carbonaceous chondrites (EET87730, EET87770, EET92042, EET92147, GRA95229, MAC87320, PCA91082, El Djouf 001), which appear to have been largely unaffected by asteroidal processes such as aqueous alteration and thermal metamorphism.

Results: Type I chondrules (Fig. 1) have large variations in bulk chemistry [5,6], inconsistent with melting of fine-grained precursor materials followed by closed-system crystallization. Evaporation and/or condensation processes of various degrees have been proposed to account for these compositional variations [2].Type I chondrules also show large variations in modal abundances of low-Ca pyroxene and olivine and can be subdivided into porphyritic olivine (PO),

porphyritic olivine-pyroxene (POP), and porphyritic pyroxene (PP) chondrules [5,6]. Low-Ca pyroxene/olivine ratio increases towards chondrule peripheries; olivine phenocrysts are often resorbed and poikilitically enclosed by low-Ca pyroxenes (Fig. 1a,b,c). Low-Ca pyroxene shells in Type I chondrules from CR chondrites are commonly surrounded by silica-rich igneous rims (Fig. 1d). In compositionally zoned chondrules, SiO_2 contents chondrule mesostases increase towards chondrule peripheries. These observations, indicating systematic increase in silica activity towards chondrule peripheries, have been interpreted as an evidence for open-system crystallization of chondrule melts [7].

Sodium contents in quench crystals of high-Ca pyroxenes are positively correlated with those in the surrounding glasses, consistent with igneous partitioning of sodium and inconsistent with its volatilization during chondrule formation [8]. In chondrules with compositionally-zoned mesostases, sodium and potassium contents in mesostases increase towards chondrule peripheries. Because melt inclusions in olivine phenocrysts are highly depleted in these elements, addition of alkalis during crystallization of chondrule melts appears to be required [8,9].

Discussion: According to a closed-system model, the observed range in chondrule bulk chemistry and modal mineralogy was inherited from solid precursor materials which subsequently experienced melting to various degrees followed by igneous crystallization. As a result, chondrule melt evolution must follow both phase relationship rules and mass balance equations. For example, in the case of magnesian PO and POP chondrules, crystallization of olivine and low-Ca pyroxene should result in enrichment of the residual melt in SiO_2 and incompatible elements such as CaO, Al_2O_3, and TiO_2; melt composition should evolve in the direction away from olivine and low-Ca pyroxene on an appropriate phase diagram. However, glassy mesostases in Type I chondrules appear to have not recorded crystallization of olivine or low-Ca pyroxene. Contrary to theoretical calculations and experiments on chondrule-like compositions, SiO_2 contents in these glasses are negatively correlated with Al_2O_3, CaO, and TiO_2 contents [4-6]. This trend covers large range of glass compositions (from 40 to 85 wt% SiO_2) from the Semarkona and CR chondrite chondrules; the similar trend is also observed from core to

rim in individual chondrules [4]. Although melt inclusions in olivine phenocrysts are considered to be more pristine than chondrule mesostases, their compositions follow the same trend [4]. When plotted on a CMAS (CaO-MgO-Al$_2$O$_3$-SiO$_2$) phase diagram, chondrule glasses form a well-defined linear trend that is inconsistent with liquid lines of descent controlled by olivine or low-Ca pyroxene crystallization. Although alkalis, notably sodium, are known to widen considerably the stability fields of low-Ca pyroxene and olivine [10], this will not affect our conclusion that olivine and low-Ca pyroxene do not control the composition of Type I chondrule mesostases. Therefore, a closed-system crystallization of chondrule melts is not a viable model for chondrule formation.

We suggest that melt compositions of Type I chondrules have been buffered by the surrounding nebular gas and that evolution of chondrule melt compositions resulted mostly from high-temperature gas-melt interaction.

Depending on the temperature at which such an interaction takes place and its duration, changes in the melt composition may lead to crystallization of new phases, when saturation is reached (preferentially low-Ca pyroxene and silica), and/or to the dissolution of pre-existing phases, which are no longer in equilibrium (mainly olivine). Since transport of elements from the gaseous interface into the melt, crystal nucleation/growth and phase dissolution are all diffusion limited and kinetically controlled, the present model may account for chemical and mineralogical zoning and variations of Type I chondrules discussed above.

References: [1] R.H. Hewins (1997) *Rev. Earth Planet. Sci.* 25, 61. [2] B. Zanda (2004) *Earth Planet. Sci. Lett.* 224, 1. [3] S.J. Desch, H. C. Connolly, Jr. (2002) *Meteorit. Planet. Sci.* 37, 183. [4] A.N. Krot. et al. (2005) *Meteorit. Planet. Sci.* (in press). [5] R.H. Jones (1994) *Geochim. Cosmochim. Acta* 58, 5325. [6] R.H. Jones, R. D. Scott, (1989) *Proc. Lunar Planet. Sci. Conf. 19th*, 523. [7] L. Tissandier et al., (2002) *Meteorit. Planet. Sci.* 37, 1377. [8] G. Libourel et al, (2003) *Lunar Planet. Sci.* 34, 1558. [9] J.N. Grossman, C.M.O'D. Alexander. (2004) *Meteorit. Planet. Sci.* 39 suppl. A45. [10] I. Kushiro, *Amer. J. Sci.* 275, 411 (1975).

Figure 1. *Representative BSE images (a-c) and X-ray elemental map in Si Kα (d) of (a) porphyritic olivine (PO), (b,d) porphyritic olivine-pyroxene (POP), and (c) porphyritic pyroxene (PP) magnesian (Type I) chondrules from the LL3.0 ordinary chondrite Semarkona (a-c) and CR carbonaceous chondrite PCA91082 (d).* **a**) *PO chondrule consists of forsteritic olivine and low-Ca pyroxene surrounded by glassy mesostasis; high-Ca pyroxene and Fe,Ni-metal nodules are minor. Olivine grains contain rare melt inclusions and are corroded by low-Ca pyroxene in chondrule periphery; the latter occur as elongated grains parallel to the chondrule edge.* **b**) *POP chondrule contains a large, heavily-corroded grain adjacent to several small euhedral crystals of forsteritic olivine surrounded by glassy mesostasis with high-Ca pyroxene quench crystals. It is surrounded by a continuous shell of low-Ca pyroxene.* **c**) *PP chondrule consists of rare heavily-corroded grains of forsteritic olivine poikilitically enclosed in low-Ca pyroxene and glassy mesostasis with high-Ca pyroxenes. The low-Ca pyroxene grains near the chondrule periphery occur as elongated grains parallel to the chondrule edge.* **d**) *POP chondrule contains heavily-corroded olivine grains poikilitically enclosed by low-Ca pyroxene. The chondrule is surrounded by a continuous silica-rich igneous rim; no silica occurs in the chondrule core. cpx = high-Ca pyroxene; gl = glassy mesostasis; incl = melt inclusions; met = Fe,Ni-metal; ol = forsteritic olivine; px = low-Ca pyroxene; sil = silica.*