AM-82-211

American Mineralogist, Volume 67, pages 1206–1211, 1982

The cation distribution in synthetic Mg–Fe–Ni olivines

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Abstract

Synthetic Mg–Fe–Ni olivines annealed at 1000°C have been studied. The site populations of Fe^{2+}, Mg^{2+} and Ni^{2+} among the M1 and M2 sites have been determined by a combination of Mössbauer spectroscopy and the profile-fitting technique based on X-ray powder diffraction data. The Mg^{2+}–Fe^{2+} cation distribution coefficient, $K_D = [X_{Fe}(M1) \cdot X_{Mg}(M2)]/[X_{Fe}(M2) \cdot X_{Mg}(M1)]$, is close to unity but slightly decreases with increasing nickel content. The Ni^{2+}–(Mg^{2+} + Fe^{2+}) cation distribution is close to that of Ni^{2+}–Mg^{2+} earlier reported for some synthetic Ni–Mg olivines.

Introduction

The cation distribution in natural and synthetic Mg–Fe olivines has been discussed in a great number of publications. The Mg^{2+}–Fe^{2+} distribution seems to be almost random but often with iron slightly ordered into the M1 position. The results, though, are rather confusing and sometimes contradictory. For the cation exchange reaction

$$Mg^{2+}(M1) + Fe^{2+}(M2) \rightleftarrows Mg^{2+}(M2) + Fe^{2+}(M1),$$

the cation distribution coefficient is defined as

$$K_D = [X_{Fe}(M1) \cdot X_{Mg}(M2)]/[X_{Fe}(M2) \cdot X_{Mg}(M1)].$$

K_D-values slightly above 1.0 have been reported for natural olivines of terrestrial and lunar origin (*e.g.*, Rajamani *et al.*, 1975) as well as for synthetic olivines (Warburton, 1978). This tendency of ordering is in agreement with crystal field theory (Walsh *et al.*, 1974). However, the scattering among the reported K_D-values is considerable. It now appears that the Mg^{2+}–Fe^{2+} distribution is variable as a function of temperature, oxygen fugacity, composition, and possibly pressure (Brown, 1980). Using Mössbauer spectroscopy, Virgo and Hafner (1972) observed a decrease in the cation ordering at elevated temperatures. Shinno *et al.* (1974), though, have reported increased ordering at higher temperatures from studies of natural olivines. X-ray diffraction studies of olivines at high temperatures (Brown and Prewitt, 1973) indicated little or no change in the ordering. Will and Nover (1979) have suggested that an increased oxygen partial pressure increases the ordering of Fe^{2+} into the M1 site in olivine, a feature also observed by Shinno (1981). Finally, a correlation between the composition and the Mg–Fe distribution has been suggested by Ghose *et al.* (1976), and there is slight evidence of increasing K_D-values with increasing Fe/(Mg+Fe) ratios in olivines (Finger and Virgo, 1971; Wenk and Raymond, 1973; Brown and Prewitt, 1973). In this respect the presence of other elements is also of great importance. For instance, nickel is a common constituent of natural olivines and the most nickel-rich olivine, liebenbergite, has been found to contain as much as 56.32 wt.% NiO (de Waal and Calk, 1973).

Nickel has been found to strongly prefer the smaller M1 site in Ni–Mg olivines (Rajamani *et al.*, 1975; Bish, 1981) as well as in Ni–Fe olivines (Annersten *et al.*, 1982). Nickel, even in low concentrations, may therefore affect the Mg^{2+}–Fe^{2+} distribution. In this study we have evaluated the cation distribution in one Mg–Fe and two nickel-

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Observed and calculated integrated intensities from the X-ray powder-profile refinements of the Ni-Fe-Mg-olivines H2 and H4 (1982).

			H2		H4	
h	k	l	I_{calc}	I_{obs}	I_{calc}	I_{obs}
0	2	0	3171	3313	2581	1877
1	1	0	2018	1533	2812	2352
0	2	1	6752	5677	4134	4224
1	0	1	2110	3683	1357	2235
1	1	1	7195	8142	5700	6143
1	2	0	3350	2597	1499	1707
1	2	1	3132	2231	1547	1492
0	0	2	2040	2019	439	1419
1	3	0	33128	31062	23920	25419
0	2	2	6928	7297	6523	5834
0	4	0	2570	1503	2681	3153
1	3	1	29832	32051	19677	19789
1	1	2	66723	71446	50456	54536
2	0	0	2868	5200	3068	4046
0	4	1	6590	5786	3318	3688
2	1	0	5360	7875	2555	3667
1	2	2	16462	18243	10672	12481
1	4	0	10177	10247	8165	9129
2	1	1	13611	12996	7170	6522
2	2	0	1592	1608	2180	2612
1	4	1	1004	2683	199	287
1	3	2	11645	11902	8472	9352
2	2	1	1764	2889	501	990
2	3	0	2797	3590	936	2607
0	4	2	4349	4685	4600	4700
1	5	0	8765	8882	6926	8735
2	0	2	154	538	3	257
0	2	3	770	2962	138	438
2	3	1	18	934	10	37
1	0	3	101	1112	270	1486
2	1	2	2093	3582	1691	3768
1	1	3	5461	7052	3368	6702
1	4	2	1904	2031	542	1672
1	5	1	6147	5428	3657	5349
2	2	2	116899	117460	67398	67005
2	4	0	34053	33785	21119	22119
1	2	3	6703	6965	3789	3777
0	6	0	2015	4080	769	1175
2	4	1	15205	15560	10319	10676
2	3	2	2215	2418	1823	3379

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