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Low Spin Ferrous Iron in the Iron Silicate Deerite

AGRELL[1] recently found a group of new minerals occurring in ferruginous cherts metamorphosed under conditions of the glaucophane–lawsonite schist facies. This metamorphic environment is characterized by a combination of pressures near 10 kbar and temperatures in the range 200°–300° C. The partial pressure of water must be high. High-pressure phases such as jadeite, lawsonite, omphacite and aragonite are found in related rocks. Deerite, one of the new minerals, is essentially a hydrated ferrous–ferric silicate. The mineral is almost opaque to visible radiation and this property suggested that the magnetic behaviour might be of interest.

Deerite was separated from a sample from the type locality, Laytonville, California. The chemical analysis is listed below in Table 1. The formula is appproximately $Fe^{2}_{13}Fe^{3}_{7}Si_{13}O_{44}(OH)_{11}$ with minor substitution of titanium and aluminium for ferric iron and manganese for ferrous iron.

The inverse magnetic susceptibility is shown as a function of temperature in the range 80°–300° K in Fig. 1. The susceptibility is independent of field strength between 2,500 and 6,500 G over the temperature range studied. But the marked deviation from Curie–Weiss law behaviour suggests that ferromagnetism might be observed at lower temperatures. It is hard to suggest an alternative explanation for the temperature dependence of the susceptibility but some field dependence would be expected at lower temperatures if the system is ferromagnetic.

The observed susceptibility per formula unit (that is, 20 iron atoms), corrected for diamagnetic components, is $147,700 \times 10^{-6}$ c.g.s. units at 295° K. Expected susceptibilities for various configurations are summarized in Table 2 and no high spin arrangement will account for the observed susceptibility.

Table 1

Chemical analysis		Formula based on 55 oxygen atoms	
SiO_2	32·95	Si	13·02
TiO_2	0·19	Ti	0·06
Al_2O_3	0·19	Al	0·09
Fe_2O_3	22·81	Fe^3	6·78
FeO	35·24	Fe^2	11·64
MnO	4·00	Mn	1·34
MgO	0·08	Mg	0·05
CaO	0·15	Ca	0·06
Na_2O	—	OH	10·87
K_2O	—	O	44·13
H_2O^+	4·12		
H_2O^-	0·10		
Total	99·79		

Table 2

Configuration	$10^6 \times$ Susceptibility at 295° K Fe^3	Fe^2	Formula unit
High spin, octahedral	14,700	10,200	235,500
Low spin, octahedral	2,250	0	15,750
High spin, tetrahedral	14,700	11,000	245,900

As there is no evidence for antiferromagnetic behaviour, it may be concluded that some of the ions present are in a low spin configuration. It has been suggested[2] that ferrous ions will assume a low spin configuration at lower ligand field strength than ferric ions. This assumption would suggest that of the thirteen ferrous ions about four are in the high spin configuration, perhaps due to a distribution between octahedral and tetrahedral sites. This must be considered an approximation and further refinement would require knowledge of the crystal structure. It seems certain that a range of sites for the M^2 cations will be found in the structure.

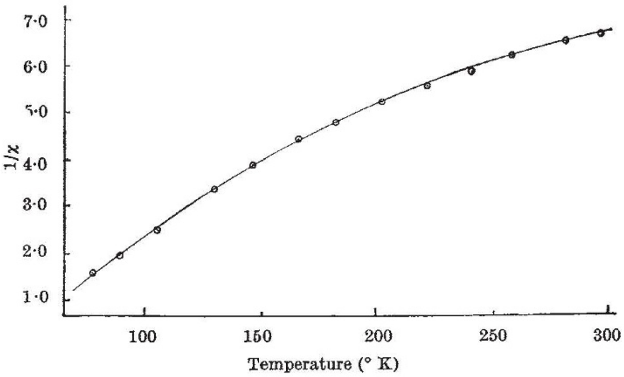

Fig. 1. Temperature dependence of inverse susceptibility of deerite. Units shown correspond to observed pull on the sample.

Deerite is a moderately dense substance in the sense that reactions between silicates and oxides leading to deerite indicate a large negative volume change. This would be expected on the basis of a small low spin ferrous ion. It is also possible that π interaction is enhanced with shorter bond distances and may account partly for the anomalous optical properties. The magnetic properties show that low spin ferrous iron in oxygen co-ordination does not require extreme pressures and leaves little doubt that olivine or spinel phases in the mantle of the Earth will contain iron in this diamagnetic state as previously suggested[3].

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