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PHYSICS OF THE EARTH AND PLANETARY INTERIORS

Physics of the Earth and Planetary Interiors 151 (2005) 107-114

www.elsevier.com/locate/pepi

# Pyrrhotite pTRM acquisition in metamorphic limestones in the light of microscopic observations

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Received 18 February 2004; received in revised form 22 December 2004; accepted 31 January 2005

#### **Abstract**

The potential of pyrrhotite as a recorder of successive, independent pTRMs in graphite bearing metacarbonates depends on the amount and composition of metamorphic fluids. During its formation very low amounts of H<sub>2</sub>S dominated fluids will favour an in situ epitaxy of pyrrhotite at the expense of iron bearing minerals in its surroundings. The resulting particle assemblage of pyrrhotite is within the single domain grain-size range and exhibits a broad blocking temperature spectrum. With increasing fluid content the desulfidation of pyrite controls the pyrrhotite formation and results in large crystals up to several millimeters, where independent pTRMs cannot be expected. As both types are present in regional and contact metamorphic rocks, the existence of pervasive fluids determines whether or not pyrrhotite will be able to record successive pTRMs, i.e. changes of the earth magnetic field (EMF) during metamorphic cooling.

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Keywords: Pyrrhotite; TEM; pTRM acquisition; Metamorphic limestone; Skye; Elba; Manaslu

#### 1. Introduction

During the last two decades the relevance of pyrrhotite as the main carrier of paleomagnetic information in very low-to-low grade metamorphic shales and limestones has been demonstrated (Rochette, 1987; Appel et al., 1991; Ménard and Rochette, 1992; Schill et al., 1998; Crouzet et al., 1999; Gillett, 2003). The formation of pyrrhotite in such rocks is generally connected with the breakdown of pre-metamorphic pyrite and/or magnetite in the presence of low oxygen

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fugacity (f(O<sub>2</sub>)). In its simplest form this transformation can take place as a desulfidation reaction of pyrite as proposed by Carpenter (1974) and Ferry (1981) valid for green schist metamorphic conditions. According to Lambert (1973), this reaction may start already around 200 °C in presence of carbonate and water. After Crerar et al. (1978) pyrrhotite can also be formed by reduction of magnetite due to sulfur or pyrite, if there is either a decrease in oxygen fugacity or an increase in sulfur fugacity (f(H2S)) for the reactions to start. Based on thermodynamic modelling and magnetic data, Gillett (2003) suggests that this reaction also starts at  $\sim$ 200 °C. Alternatively, iron might come from silicates and/or oxides as proposed by Thompson (1972), Ferry (1981) and Tracy and Robinson (1988). Nevertheless, Ferry (1981) assumes that this reaction is negligible as the amount of iron in silicates and oxides is low.

Taking the C–H–S fluid system into account, Ferry (1981) proposed two possible reactions including CH<sub>4</sub> and H<sub>2</sub>O, whereas H<sub>2</sub>O dominates at higher temperatures. Poulson and Ohmoto (1989) favour H<sub>2</sub>S at higher temperatures (>500 °C), low pressure and low f(O<sub>2</sub>). The dominance of H<sub>2</sub>S at higher temperatures leads to a decrease in the H<sub>2</sub>O contingent  $\ll$ 1, followed by the dehydration of hydrous minerals. In a contact metamorphic environment, such H<sub>2</sub>S rich fluids are in direct vicinity to the contact and enable selective transfer of sulfur from pelites into the pluton.

Studying variations and intensities of the EMF requires that the assemblage of ferromagnetic minerals comply with the Thellier laws of additivity and independence of successive pTRMs (Thellier and Thellier, 1959). In other words, the blocking temperature should be the same as the unblocking temperature. As only single domain (SD) – and to a lesser amount smaller pseudo single domain (PSD) - particles fulfill these laws the grain-size of pyrrhotite should not exceed a few micrometer (SD-MD transition 1-2 µm according to Soffel, 1977). It also implies that magnetic interaction between neighbouring SD particles can be neglected. Using magnetic techniques this was already verified for regional metamorphic limestone of the Western Alps (Crouzet et al., 1999; Wehland et al., in press). The aim of our study is to investigate the process of pyrrhotite formation and its appearance in different metamorphic settings under the light of its relevance as a magnetic remanence carrier. Here we focused on the microscopic indications whereas magnetic experiments are described in another paper (Wehland et al., 2005).

#### 2. Samples and methods

Sampling was performed from contact metamorphic limestones from the Isle of Skye (Scotland) and Elba Island (Italy) and on regional metamorphic limestones from the area of Bourg d'Oisans (France, Crouzet et al., 1997) and the Tethyan Himalaya (Manaslu area, Nepal; Schill et al., in press). The latter one has a two-fold nature as near the Manaslu Intrusion the limestone has a clear contact metamorphic overprint.

Pyrrhotite in these samples was detected by the means of rockmagnetic methods like thermal demagnetisation of the Saturation Isothermal Remanent Magnetisation (SIRM) and thermomagnetic measurements of the susceptibility. Magnetite or hematite can exist in low amounts (Wehland et al., 2005).

For the microscopic investigation samples from the contact metamorphic areas were taken with an increasing distance to the respective intrusion and subjected to scanning electron microscopy (SEM), transmission electron microscopy (TEM) and light microscopy (transmission and reflected). For the SEM analysis the surface of the polished samples were treated with diluted (5%) hydrochloric acid for 10 s. The etching was necessary as the high competence contrast between the ore minerals and the calcareous matrix lead to a smeared surface of calcite. The competence contrast was also responsible that the TEM analysis was only successfully performed on samples from Bourg d'Oisans.

#### 3. Results

#### 3.1. Regional metamorphic rocks

Using transmitted and reflected light microscopy, pyrite is identified as the dominant ore mineral and appears in aggregates of up to 0.5 mm. Magnetite and pyrrhotite could not be found by such means. Nevertheless, as the existence of pyrrhotite in these samples is evidenced from rockmagnetic studies, it has to exist in a submicroscopic size of smaller than  $\sim 2 \, \mu m$ .

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