Sulfide and platinum mineralization in the Merensky Reef: evidence from hydrous silicates and fluid inclusions

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Abstract. The base metal sulfides of the Merensky Reef are associated with hydrous silicates and intense deuteric hydrous alteration of cumulus and postcumulus silicates. Biotite and phlogopite crystallized in the vicinity of sulfides from a volatile-enriched highly fractionated intercumulus melt. Amphibole, chlorite, and talc are later alteration phases of cumulus pyroxene and intercumulus plagioclase. Biotite is often accompanied by zircon, rutile, and quartz. Accessory quartz hosts a complex suite of H$_2$O-NaCl-(CaCl$_2$)-CO$_2$-CH$_4$ fluid inclusions which have thus far not been described from the Merensky Reef. The earliest fluid inclusion compositions are NaCl-(H$_2$O) with less than 10 vol. % water; CO$_2$ coexisting with a halite daughter crystal and brine; and polyphase inclusions with up to six daughter and accidental phases and high contents of divalent cations. The maximum trapping temperature is around 730°C at 4 to 5 kb pressure. Later inclusion generations are H$_2$O-NaCl, CO$_2$, H$_2$O, and pure CO$_2$ and CH$_4$. The presence of Cl-rich fluids during the intercumulus stage of the crystallizing Merensky Reef is directly related to the mode of sulfide precipitation. Prior to sulfide unmixing in a hydrous magma sulfur is likely to be present as H$_2$S. Sulfur saturation causes reaction of H$_2$S with oxides of the silicate melt to form a sulfide melt plus water. During reaction the magma is enriched in water until a separate fluid unmixes. It carries all compounds with high fluid/melt partition coefficients, as well as metals capable of forming OH- and Cl-complexes. Precious metals are assumed to have fractionated into the Cl-rich fluid as Cl-complexes rather than being dissolved in the sulfide melt. During the cooling evolution of the fluid the precious elements precipitate around the periphery of sulfide melt droplets. The model proposed explains the distribution pattern of platinum-group minerals in the Merensky Reef better than any orthomagmatic mineralization concept offered so far.

Introduction

The Merensky Reef of the Bushveld Igneous Complex, South Africa, not only hosts the world's largest workable concentration of platinum metals. It is also one of the most enigmatic horizons in the magmatic stratigraphy of the layered suite. There are a number of unsolved structural and mineralogical problems to which any model should provide an answer. These include the pegmatitic texture of the Reef, its stratiform concentration of base metal sulfides and associated platinum-group elements, the distribution pattern of chromite, and the origin of pothole disturbances affecting the structural setting of the Merensky Reef (Cousins 1969; Lauder 1970; Vermaak 1976; Ballhaus and Stumpf 1985).

This paper will emphasize the following, hitherto unknown or poorly understood, aspects of the Merensky Reef:

1. Base metal sulfides, the main carriers of precious metal minerals, form part of the intercumulus stage of the Merensky Reef and are intergrown with a characteristic association of hydrous silicates and hydrous alteration. Plagioclase in the vicinity of sulfides is enriched in anorthite and altered to chlorite.

2. Postcumulus quartz hosts a complex suite of high-temperature fluid inclusions of the system H$_2$O-NaCl-(CaCl$_2$)-CO$_2$-CH$_4$.

3. The majority of platinum-group minerals and the by far largest grains are concentrated at the periphery of interstitial sulfides towards silicate gangue.

The distribution pattern of precious metal minerals and hydrous silicates associated with sulfides have long been known (Wagner 1929; Liebenberg 1970; Van Zyl 1970; Vermaak and Hendriks 1976; Crocket et al. 1976). Their significance for sulfide and platinum mineralization have, however, largely been ignored in the past. Fluid inclusions have not been described previously.

The paper will therefore illustrate the textural association of base metal sulfides with hydrous silicates. It will, for the first time, describe the fluid inclusions, and will discuss the relevance of these findings to sulfide and platinum mineralization. The data are based on extensive underground work on normal undisturbed Merensky Reef in the Brakspuit shaft area of Rustenburg Platinum Mines, southwestern Bushveld Complex. Investigations are restricted to the mineralized pegmatitic pyroxene orthocumulate (Fig. 1) at the base of the Merensky cyclic unit (Jackson 1970). This horizon will subsequently be referred to as the Merensky Reef. The geology of the Bushveld Complex, the structural setting of the Reef, and its petrography have been documented extensively in the past and need not be repeated here. For a detailed account on these subjects the reader is referred to Wagner (1929), Cousins (1969), Vermaak (1976), Vermaak and Hendriks (1976), and Von Gruenewaldt (1979).
Base metal sulfides and hydrous silicates

The base metal sulfides of the Merensky Reef have received some attention in the past since they host almost all platinum-group minerals (Kingston 1966; Liebenberg 1970; Vermaak and Hendriks 1976; Kinloch 1982). Their mineralogy is relatively simple. The main sulfide minerals, in decreasing abundance, are pyrrhotite, pentlandite, and chalcopyrite, occasionally accompanied by pyrite, cubanite, and rare galena, sphalerite, and sulfarsenides. The majority of sulfide grains are distinctly zoned in accordance with magmatic fractionation trends (Kingston 1966). Pyrrhotite occupies central parts of grains and is surrounded by mosaic aggregates of pentlandite and chalcopyrite. Rare sulfarsenides, as well as platinum-group minerals, closely adhere to contacts against silicate gangue. Sulfide minerals are invariably intercumulus constituents, and the average sulfide content of the Merensky Reef is 2.5 to 2.9 vol.% (Vermaak and Hendriks 1976).

The intergrowth relations of base metal sulfides with coexisting silicates have not received much attention in the past. Almost all sulfides are intergrown with a characteristic suite of low-temperature hydrous silicates and accompanied by intense deuteric alteration (Crocket et al. 1976).

The currently accepted sulfide and platinum mineralization models, however, do not take much cognizance of these findings (Campbell et al. 1983; Naldrett et al. 1985). In spite of the restriction of sulfides to intercumulus positions, they emphasize the importance of very early sulfide unmixing to attain the platinum content of the Reef: Plumes of fresh hot magma enter the magma chamber, spread out somewhere above the stratigraphic level of the Merensky Reef, and mix thoroughly with cooler fractionated melt. The blends are oversaturated with sulfur and minute sulfide melt droplets unmix. When sulfide droplets settle down to the Reef position they come into contact with a large amount of silicate melt and scavenge platinum-group elements.

The textural positions of base metal sulfides on the contrary suggest in-situ sulfide melt unmixing. This must have occurred during the post-cumulus stage, according to textural positions of sulfide minerals. Early sulfide droplets as inclusions in cumulus silicates are lacking completely in the Reef.

The common association of sulfides with hydrous silicates suggests interactions of sulfides with silicate minerals and silicate melt. These might be indicative for the mode of sulfide melt formation in the Merensky Reef. Representative intergrowth patterns are illustrated in Fig. 2 to 5.

Biotite attached to a base metal sulfide grain is euhedral to subhedral against it (Fig. 2). It may occur both as inclusions in and at the periphery of sulfides with or without a direct contact. It is important to stress that biotite may occur anywhere in the vicinity of a sulfide grain. Its location is not controlled by any other silicate, and biotite cannot be related to alteration. Where biotite penetrates a sulfide, the textural relations suggest that biotite crystallization predates sulfide solidification.

Biotite associated with sulfides often contains zircon and rutile inclusions, and is frequently intergrown with quartz (Fig. 3A and B). The association of base metal sulfides with biotite, zircon, rutile, and quartz is very common in the Merensky Reef. It may also be accompanied by rare tourmaline, apatite, and orthoclase.

Other sulfide grains are associated with amphibole (Fig. 4) and, in a similar pattern, with talc. Unlike biotite, amphibole and talc form irregular contacts and rims around sulfide grains, or acicular intergrowth patterns as shown in Fig. 4. Both minerals reveal a distinct affinity to the contacts of sulfides against orthopyroxene or clinopyroxene, or to the alteration products of both. Subhedral crystal shapes such as biotite shows against sulfides are much rarer, implying that amphibole and talc are slightly later than biotite (Wagner 1929).

Plagioclase, the main intercumulus silicate of the Reef, undergoes specific alteration stages in the presence of sulfides and hydrosilicates. Plagioclase is distinctly enriched in anorthite if in contact with any hydrous silicate (see below). It avoids the direct contact with base metal sulfides; sulfide grains are isolated from plagioclase by a fine rim of chlorite. When sulfides become more abundant, plagioclase may be infiltrated by chlorite veinlets (Fig. 5) or may have disintegrated into micro-sized anorthite crystallites embedded in a chlorite matrix ("granulation", Vermaak and Hendriks 1976). Granulation eventually merges with complete chloritization.

Saussuritization of plagioclase is also common. This alteration style determines the general preservation state of the respective Reef portion. Its intensity does not show a small-scale correlation with sulfides or hydrous silicates, but correlates well with the overall sulfide content of the Reef.