Re-Os isotope and geochemical study of the Vredefort Granophyre: Clues to the origin of the Vredefort structure, South Africa

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ABSTRACT
The origin of granophytic rock dikes (the Vredefort Granophyre) at the 2 Ga Vredefort structure in South Africa has been controversial. They have been interpreted as either impact melt or igneous intrusions. Most Vredefort Granophyre samples have considerably higher Os contents than the country rocks and $^{187}\text{Re}/^{188}\text{Os}$ and $^{187}\text{Os}/^{188}\text{Os}$ ratios that scatter about a 2 Ga isochron. Their initial $^{187}\text{Os}/^{188}\text{Os}$ ratios (at 2 Ga) overlap the meteoritic data range, indicating that all the granophyre samples contain some meteoritic Os. The Re-Os isotopic composition of the granophyre is significantly different from that of local precursor rocks and indicates that up to 0.2% of a chondritic component could be contained in the granophyre. Our results confirm the interpretation that the Vredefort Granophyre represents an impact melt rock.

INTRODUCTION
The Vredefort structure in South Africa, about 120 km southwest of Johannesburg, currently has a diameter of about 100 km, but may initially have been as large as 300 km (e.g., Therriault et al., 1996). The origin of the structure has been debated during most of this century (cf. Reimold, 1993). Recent data show the existence of impact-characteristic shock metamorphic effects in Vredefort rocks, e.g., basal Brazil twins in quartz (Leroux et al., 1994) and planar deformation features in zircon (Kamo et al., 1995), supporting an impact origin as opposed to an internal origin of the structure. The Vredefort event is well dated at 2024 ± 5 Ma from U-Pb ages of zircons from Venture Hall (Walraven et al., 1990), and 1972 ± 24 Ma from muscovite (van Staal et al., 1993). The event had occurred about 30 m.y. after the formation of the Bushveld Complex.

Dikes of granophyric rock with an age of 2 Ga (Walraven et al., 1990) occur in the basement core of the structure and along the boundary between the core and the supracrustal rocks of the so-called collar (Fig. 1A). These rocks have been termed enstatite or bronzite granophyre, but, as the pyroxene in this rock is of hypersthene composition, the name Vredefort Granophyre has been established (Reimold et al., 1990). In the internal model for the origin of Vredefort, an igneous origin of the granophyre was suggested (e.g., Bisschoff, 1972), whereas in the impact model this rock was proposed to be impact melt that was injected into the floor of the impact structure (e.g., French et al., 1989; French and Nielsen, 1990).

To understand the origin of the Vredefort structure and the effect that the Vredefort event had on the ore deposits in the Witwatersrand basin, it is of critical importance to understand the nature and origin of the granophyre (e.g., Therriault, 1992). Several attempts have been made to identify a meteoritic component in this rock, but without success (e.g., Schreyer, 1983; French et al., 1989; Reimold et al., 1990), probably due to limitations of the methods used. Here we apply the sensitive Re-Os isotope technique to some basement rocks and Vredefort Granophyre samples to detect the presence of a meteoritic component.

CHARACTERISTICS
The core of Vredefort consists of two granitic to granodioritic gneiss units that are older than 3 Ga (Hart et al., 1981; Kamo et al., 1995): the central terrane of Inlandsee Leerocynofels (mainly granulitic quartz–K-feldspar gneiss), and the outer annulus of Outer Granite Gneiss, which is a collective name for a suite of heterogeneous, strongly migmatized, granitic to granodioritic gneisses of amphibolite grade. Along a traverse outward from the margin of the core, the collar consists of quartzites and shale formations of the 2.7–2.9 Ga Witwatersrand Supergroup, the mafic extrusives of the Venterdorp Supergroup of about 2.7 Ga age, and the carbonates and quartzites of the 2.25–2.5 Ga Transvaal Supergroup (Walraven et al., 1990).

Vredefort Granophyre dikes are up to tens of metres wide and several kilometres long. All dikes, whether they occur in core gneisses or supracrustal rocks (quartzite, shale) of the collar, have a homogeneous composition and similar clast populations (e.g., Reimold et al., 1990) dominated by granite and quartzite, besides minor shale. Mafic clasts are very rare and only locally important where a granophyre dike crosscuts a mafic intrusion (Reimold et al., 1990; Therriault, 1992). French and Nielsen (1990) concluded from their major element mixing calculations that the granophyre could have formed by impact melting of granite, shale, and 30%–50% basalt, whereas major and trace element mixing models by Reimold et al. (1990) reproduced the granophyre composition by a major granite component with smaller amounts of shale and quartzite. These authors showed that a major mafic contribution is unlikely due to the scarcity of mafic clasts and because the granophyre composition can be perfectly modeled with felsic crustal and supracrustal rocks. All models require that the 2 Ga Vredefort Granophyre be made from significantly older crust (e.g., 2.7–3 Ga).

Texturally, two types of granophyre have been distinguished (Bisschoff, 1972; Reimold et al., 1990; Therriault, 1992): a spherulitic type with up to 5 cm large spherulites of orthopyroxene set in a fine-grained groundmass of feldspar and quartz (Fig. 1A), and a variety of more granular orthopyroxene and feldspar (Fig. 1B). The micropegmatitic matrix occurs in variable amounts in both types. The clasts display...
textures that are indicative of high-temperature alteration and partial melting. Formerly granitic clasts often consist of completely annealed, rounded aggregates of relict quartz set in a fine-grained devitrified matrix of feldspar, quartz, and pyroxene. The typical granophytic texture is found in small areas of such recrystallized clasts, and where more complete melting of the clasts has occurred, the proportion of granophytic component is higher. Some grains remain unannealed, but, in contrast to other Vredefort lithologies, no shock deformation features (e.g., planar deformation features) have been found in clasts in the granophyre.

SAMPLES AND METHODS

Several typical Vredefort Granophyre samples (granular granophyre: BG-4, -7, and -168; spherulitic granophyre: BG-9, -10) were selected for this study (Fig. A). In addition, we analyzed samples of Witwatersrand shale (Parktown Shale Formation, West Rand Group; VG-SNE), a Witwatersrand Supergroup-derived siltstone clast from a core granophyre specimen (BG-S1) (Reimold et al., 1990), an Outer Granite Gneiss sample (OT-1), and two andesitic samples from Ventersdorp Supergroup exposures in the western collar of the structure (UP-61, UP-63). All samples are surface samples. Petrographical descriptions of the above rocks are given in Reimold et al. (1990) and Therriault et al. (1996). Sample selection was made under the assumption that Transvaal Supergroup carbonates would not have been incorporated in the melt due to vaporization, and Witwatersrand quartzites and local granites do not contribute significantly to the Re and Os contents of the resulting melt (see below). Major element compositions (and Y and Nd) were determined by X-ray fluorescence analysis (XRF; in part from Reimold et al., 1990; Reimold, 1991); trace elements, by XRF and neutron activation analysis (e.g., Koeberl et al., 1994a); and Re and Os abundances and Os isotopic compositions, following methods of Shirey and Walker (1995).

### TABLE 1. MAJOR AND TRACE ELEMENT CONTENTS OF VREDEFORT ROCKS AND VENTERSDORP LAVA

<table>
<thead>
<tr>
<th>Sample</th>
<th>SiO₂</th>
<th>TiO₂</th>
<th>Al₂O₃</th>
<th>Fe₂O₃</th>
<th>MgO</th>
<th>CaO</th>
<th>Na₂O</th>
<th>K₂O</th>
<th>P₂O₅</th>
<th>LOI</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>VREDEFORT GRANOPHYRE</td>
<td>70.9</td>
<td>4.4</td>
<td>13.9</td>
<td>12.4</td>
<td>1.1</td>
<td>1.0</td>
<td>0.6</td>
<td>0.2</td>
<td>0.2</td>
<td>0.1</td>
<td>99.9</td>
</tr>
<tr>
<td>Witwatersrand shale (Parktown Shale Formation, West Rand Group; VG-SNE)</td>
<td>74.5</td>
<td>5.1</td>
<td>17.2</td>
<td>8.7</td>
<td>1.8</td>
<td>0.8</td>
<td>0.7</td>
<td>0.3</td>
<td>0.4</td>
<td>0.2</td>
<td>100.0</td>
</tr>
<tr>
<td>Witwatersrand Supergroup-derived siltstone clast from a core granophyre specimen (BG-S1) (Reimold et al., 1990)</td>
<td>72.5</td>
<td>4.8</td>
<td>15.5</td>
<td>9.2</td>
<td>1.5</td>
<td>0.7</td>
<td>0.6</td>
<td>0.2</td>
<td>0.3</td>
<td>0.2</td>
<td>99.8</td>
</tr>
<tr>
<td>Outer Granite Gneiss sample (OT-1)</td>
<td>71.8</td>
<td>5.0</td>
<td>17.2</td>
<td>9.0</td>
<td>1.8</td>
<td>0.8</td>
<td>0.6</td>
<td>0.3</td>
<td>0.4</td>
<td>0.2</td>
<td>100.0</td>
</tr>
<tr>
<td>Two andesitic samples from Ventersdorp Supergroup exposures in the western collar of the structure (UP-61, UP-63)</td>
<td>75.0</td>
<td>4.5</td>
<td>17.6</td>
<td>8.9</td>
<td>1.9</td>
<td>0.8</td>
<td>0.6</td>
<td>0.3</td>
<td>0.4</td>
<td>0.2</td>
<td>100.0</td>
</tr>
</tbody>
</table>

Note: Major element data in wt%, trace element data in ppm, except * in ppb; all Fe as Fe₂O₃.

n.d. = not determined.
tryrocks. In addition, the 187Os/188Os ratios of granophyre samples compared to the country rocks are significantly higher than the average continental crustal rock values. Thus, there is a significant mafic component. Abundances of Os, as well as 187Re/188Os and 187Os/188Os ratios, leading (in old crustal rocks) to high abundances of 187Os from the decay of 187Re. Typical values of 187Os/188Os ratios for the continental crust are about 0.67 to 1.61 (Esser and Turekian, 1988). In general, continental crustal rocks have high Re but low Os concentrations and high Re/Os ratios, leading (in old crustal rocks) to high abundances of 187Os from the decay of 187Re.

The inverse Os concentration in the granophyre range from 0.11 to 1.11 ppb, which is significantly higher than the average of the source rock values. Thus, there is a distinct enrichment of Os in most of the granophyre samples compared to the country rocks. In addition, the 187Os/188Os ratios of the granophyre samples are significantly lower than those of the supracrustal country rocks.

DISCUSSION

The Re-Os isotopic system, which is based on the 2- decay of 187Re to 187Os, is a powerful tool to identify the presence of a meteoritic component in impact-derived rocks (Koeberl and Shirey, 1993; Koeberl et al., 1994a), in distal ejecta layers (e.g., Turekian, 1982; Peucker-Ehrenbrink et al., 1995), and in deep-sea sediments (Esser and Turekian, 1988). In general, continental crustal rocks have high Re but low Os concentrations and high Re/Os ratios, leading (in old crustal rocks) to high abundances of 187Os from the decay of 187Re. Typical values of 187Os/188Os ratios for the continental crust are about 0.67 to 1.61 (Esser and Turekian, 1993). Most meteorites have about five or six orders of magnitude higher absolute abundances of Os, as well as 187Re/188Os and 187Os/188Os ratios that are significantly different compared to those in old crustal rocks. Therefore, small amounts of a meteoritic component can easily be detected in impact breccias or melts derived from continental crustal rocks (see Koeberl and Shirey, 1993, for details). In a 187Re/188Os and 187Os/188Os diagram (see Fig. 2), granophyre data (except BG-168) are scattered close to the 2 Ga reference isochron, whereas all country rock samples are unrelated to this trend.

The large spread in 187Re/188Os ratios in the country rocks is probably due to late-stage Re mobilization during alteration or metamorphic overprint, as Re is mobile under a variety of conditions. Addition of Re must have occurred much more recently than 2 Ga, as 187Os/188Os initials at 2 Ga are negative. Regional thermal metamorphism at Vredefort occurred at times later than 2 Ga (e.g., Reimold, 1993; Spray et al., 1995). The high 187Re/188Os ratios and the presence of a higher proportion of radiogenic 187Os in the shale and in basaltic rocks (UP-61, UP-63) are indicative of old crustal rocks that show open system behavior for Re.

Considering the low Os abundances in the source rocks compared to the granophyre, and their Re-Os isotopic characteristics, the composition of the granophyre can only be explained by admixture of a high-Os, low-Re noncrustal component, such as meteoritic or mantle material. A mantle contribution can be excluded for a variety of reasons. First, Os abundances in mantle rocks are lower than meteoritic abundances by a factor of ≥100. This would require a significant (≥30%) mantle component in the granophyre, which is not evident in the major or trace element composition of the granophyre and contradicts the petrologic observations that show the absence of any mantle-derived clasts in this rock. Second, Rh-Sr and Sm-Nd isotopic analyses were obtained on two granophyre samples (BG-7 and BG-10), yielding initial εNd (2.0 Ga) values of +174 and +187, and εNd (2.0 Ga) of −13.2 and −13.1, respectively (see also Table 2), which are crustal values. Both model ages (TDM and TND) calculated from the Sm-Nd data agree at about 3.2 Ga, in contrast to the 2.0 Ga age of the Vredefort Granophyre. This age is in agreement with the >3 Ga age of granitic basement gneisses (Kamo et al., 1995). Generation of the granophyre from 2.0 Ga granitic crust is also ruled out.

The inverse Os concentration in the granophyre samples shows a linear correlation with the present-day 187Os/188Os ratio, with all granophyre samples following a single trend, similar to the correlation seen in the 187Re/188Os and 187Os/188Os plot (Fig. 2). Note that the 187Re/188Os ratio of the granophyre is controlled largely by the Os content, so that the lowest 187Re/188Os granophyres have the highest Os. This variation alone in suite with such a constant major element composition is enough to suggest admixture of an external, high-Os component. However, to discuss mixing relationships it is necessary to consider the initial Os isotopic compositions at 2 Ga. The granophyre samples have a limited range of initial 187Os/188Os ratios, from 0.13 to 0.22 (except BG-168, which has a negative initial ratio, probably due to recent Re addition). The average initial 187Os/188Os ratio of the granophyre is slightly elevated compared to chondritic values (as also observed by Meisel et al., 1995, for distal impact ejecta), but the range of values overlaps with the
meteoritic data array and indicates that the majority of the Os in the Vredefort Granophyre is derived from a meteoritic component, even though the absolute Os abundance is variable. The Os contribution from the target rocks must have been very limited, as these rocks have low Os concentrations and different isotopic characteristics (Fig. 2). For example, granites and granitic gneisses elsewhere have been found to contain only about 0.003 to 0.007 ppb Os, while having high 187/188 Re and 187/188 Os ratios (e.g., Koeberl et al., 1994a). The same considerations follow from the shale and Venterdorp Lava samples analyzed in this study (Table 1).

A meteoritic component in the Vredefort Granophyre is the only explanation that is in agreement with all observations. This conclusion is supported by some enrichments in Cr, Co, and Ni (Table 1) and Ir (French et al., 1989) in the granophyre compared to the country rocks, although these enrichments are not as unambiguous as the Re-Os data. Assuming chondritic meteorite Os abundances (about 500 ppb), the Vredefort Granophyre contains ≤0.2% of a chondritic component. However, the meteoritic component is not homogeneously distributed, probably due to a nugget effect, as is evident from Figure 2 and the spread in 187/188 Os ratios, in agreement with observations from other impact melt rocks (e.g., Koeberl et al., 1994b). Meteorites contain about ten times less Re than Os, indicating that the Re contribution from the meteoritic material to the granophyre was ≤30%, subordinate to the Os, which is almost exclusively of meteoritic origin. The almost constant Re abundances found in the granophyre samples indicate homogenization during the impact. Thus, the granophyre is a mixture of a large amount of low-Os, high-Re material (crustal rocks) with a small contribution of high-Os, low-Re meteoritic material. All Vredefort Granophyre samples contain meteoritic Os. These results confirm that the Vredefort Granophyre is an impact melt rock.

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