UVAROVITE GARNET AND SOUTH AFRICAN JADE (HYDROGROSSULAR) FROM THE BUSHVELD COMPLEX, TRANSVAAL

J. J. Frankel, University of Natal, Durban, South Africa.

Abstract

Uvarovite garnet is an uncommon silicate matrix in chromitite seams in the Norite body of the Bushveld Igneous Complex, Transvaal, Union of South Africa. Separated uvarovite samples were analyzed chemically, and unit cell, specific gravity and refractive index values determined. Uvarovite, grossularite and andradite are the dominant "end members." A linear relationship between chromic oxide content and physical properties is modified by titanium and iron. The deviations are discussed and estimated. The green color is tinged with brown where titanium and iron are high. The associated chromite grains have a chemical composition similar to that of nearby chromitite seams. Uvarovite-diopside layers between uvarovite-bearing chromitite seams in the Eastern belt of the Norite are described. It is considered that the uvarovite was formed in pyroxenite and chromitite where adequate chromium was available, by metamorphic metasomatic processes after the consolidation of the Norite body itself.

Introduction

Uvarovite, the bright green chromian garnet, was first reported associated with chromitite seams, from the eastern part of the Bushveld Complex (Hall, 1908, p. 60; Hall and Humphrey, 1909, p. 74). Grossularite, calcium aluminium garnet, in a massive form associated with anorthosites and pyroxenites, was described from the Brits area in the western part of the Complex (Hall, 1925). Several occurrences of green garnet, probably grossularite, and other varieties in metamorphosed dolomitic rocks at the contacts with the base of the Complex have also been recorded (Kynaston, 1909, p. 20; Wagner, 1929, p. 175). A few further details of garnet in the Complex were given by Hall (1932) and Kupferbürger (1937).

My interest in these garnets arose during an examination of chromite deposits in the Bushveld Complex with the late Dr. J. Galletich. Professor E. Mendelssohn, Witwatersrand University, later gave me a uvarovite-bearing chromitite of remarkable grain size variation; I also received specimens from Professor B. V. Lombaard, Mrs. A. Blignaut of the Johannesburg Public Library Museum and Dr. J. D. Steyn of the Union Geological Survey Museum, Pretoria. A detailed investigation of uvaro-
vite and of South African Jade which could add to known mineralogical data was then possible.

The localities of the specimens investigated are shown on the map (Fig. 1). In addition to these localities, several others are known.

The chromium-bearing garnet is less abundant and more restricted in its associations than other members of the garnet family. Uvarovite is usually found as a matrix to closely-packed chromite grains, in thin veins or scattered in metamorphosed sediments. The crystals are generally small and intimately associated with other minerals. This makes a clean separation of uvarovite difficult and may account for the paucity of good chemical analyses coupled with adequate physical data. Thus correlation of chemical composition and physical properties in the chrome-bearing varieties of the ugrandite series has been based upon sparse and incomplete information.

The distribution and mineral associations of uvarovite and grossularite in the Bushveld Complex are dissimilar in many respects, although they do occur more or less in the same zone above the base of the Norite body. The uvarovite is rarely associated with grossularite and vice-versa. For this there must be some fundamental reason. Aims of the present study, therefore, were to clarify the relationships between these garnets and to contribute towards an understanding of their origins in the Bushveld Complex.

Fig. 1. Geological sketch map of the Central Transvaal, showing the extent of the Norite body of the Bushveld Complex and the localities of specimens here described. (Modified from Union Geological Survey Maps.)
THE FIELD SETTING OF THE UVAROVITE AND SOUTH AFRICAN JADE

The field occurrence of the garnetiferous rocks will be made clear from the following brief account of the Bushveld Igneous Complex. This basin-like structure occupies an area, elongated along an east-west axis, in the central part of the Transvaal Province of the Union of South Africa. The surrounding, underlying sediments dip towards the centre of the Complex of which much is covered by later formations. The main continuous exposures are preserved in two areas, known as the Eastern and Western Belts (Fig. 1).

The Complex comprises two major rock groups; an upper Red Granite overlying and bordered by a lower Norite-gabbro body. The Norite body exhibits “pseudo-stratification” or rifting which agrees in a general way with the low to moderate dip of the floor strata. It has been sub-divided into several zones, of which the Critical zone rather low down in the body, is composed of a well pseudo-stratified, diversified, layered succession of peridotite, anorthosite, pyroxenite and chromitite “seams.”

The chromitite seams consist of chromite crystals with little interstitial silicate mineral. They range from thin partings to robust seams several feet thick that persist for long distances along strike and parallel to the pseudo-stratification of the enclosing rocks. Disseminations of chromite crystals are common in the pyroxenites.

There are two well-defined groups of seams in the Eastern Belt, designated Lower and Middle groups. They are well exposed in the hilly and mountainous country. There is also an Upper group in the Western Belt where the country is flat. Faulting or black turf overburden often make outcrop correlation uncertain.

The Bushveld uvarovite occurrences are all associated with chromitite seams, at the contacts with surrounding rock, as cross-cutting veins, as lenses and bands within the seams, or in lenticular bodies with disseminated chromite not far from a true chromitite seam.

In the field the uvarovite appears to take the place of pyroxene and feldspar within the chromitite seams or at the contacts with anorthositic or pyroxenitic rocks. Sometimes, masses of almost pure uvarovite associated with crystals of diopside and zoisite and only very small amounts of disseminated chromite grains, are marginal to chromitite seams. Most of the uvarovite in the Eastern Belt is associated with the Lower group chromitite seams; the examples from the Western Belt may come from the Middle or Upper groups.

The rock prized as an ornamental stone and known as South African Jade occurs in the almost featureless flat country about 40 miles due west of Pretoria. Although poorly exposed through a black turf overburden, the field relationships have been clearly revealed in trenches and pits.
Two or three bands of massive pale green rock, from a few inches to two feet thick are separated by bands of light-colored massive anorthosite. An anorthosite-norite with jade, rests on a chromitite seam below the main jade seams. Scattered thin bands or streaks of chromite crystals also lie within the jade. The jade layers and the associated rocks conform to the characteristic pseudo-stratification of the Norite body and dip at 15 to 20 degrees towards the north.

According to Hall (1932, p. 415), the jade horizon can be traced at intervals along many miles of strike, and where it leaves the chromitite horizon it is a white lime silicate hornfels. Occurrences of similar rock have also been reported from the Potgietersrust area in the northern part of the Complex and from the eastern part as well.

**Uvarovite**

*Experimental Procedure*

The uvarovite is generally the matrix around fine chromite crystals so that separation cannot be achieved by simple hand-picking; similarity in grain size of garnet and chromite does not allow of separation by sieving. In most of the specimens garnet was only liberated from the associated minerals after crushing to minus 200 mesh Tyler. The following procedure for separating the garnet from other minerals was adopted.

Gentle crushing in a diamond mortar released larger chromite grains, so that a fair percentage could be eliminated by hand-picking and coarse sieving. The minus 200 mesh powder was gently deslimed, dried, and repeatedly centrifuged in Clerici solution S.G. 3.8 to 3.9. In this way free particles of garnet and other lighter minerals were separated from the chromite and aggregate chromite-garnet grains. The lighter minerals were removed from the garnet by centrifuge treatment in methylene iodide or suitably adjusted Clerici solution. All samples were carefully checked microscopically for purity. The minute amount of chromite in scattered aggregate grains still present after purification cannot have a significant effect on the chemical analyses of the garnet. The garnet powders were thoroughly washed to remove adhering heavy liquid and were finally oven-dried.

The determination of specific gravity on the fine powders proved difficult, but repeated measurements using Ellsworth's vitreosil pyknometers gave reasonably consistent results and all values were then calculated to 20° C. A few values were checked by suspension of grains in Clerici solution suitably diluted. The refractive index measurements were made in West's high refractive index liquids in sodium light.

The color of the garnet was measured on a Donaldson Trichromatic Colorimeter which I had previously used with some success (Frankel.
UVAROVITE GARNET AND SOUTH AFRICAN JADE

1953). Colors are also described by comparison with a standard color chart.

Full chemical analyses were not made on all specimens, mainly owing to lack of sufficient material. The accuracy of ferrous oxide determinations in garnets is often questioned and considerable attention was paid to this matter by the analysts, who used several methods, all of which gave reasonably consistent results. Mr. Schutte used Hey’s method (1941), while Mr. Herdsman carried out the determination by simple solution in phosphoric acid in constant flow of carbon dioxide and checked against a blank and a standard powder. Particular attention was also paid to the determination of combined water in the uvarovite.

The National Physical Laboratory reported that the Debye-Scherrer photographs were taken in a camera of 114.59 cm. diameter according to the Straumanis principle, using either Cu Kα or iron-filtered Co radiation. Exposure times were approximately 7 and 16 hours respectively.

Chemical and Physical Data on Uvarovite

The chemical analyses (Table I) of the garnet can be recast as isomorphous mixtures of “end members”; mainly uvarovite, grossularite and andradite which, except for Specimens Nos. 1 and 2, range in approximate proportions from 5:5:1 through 4:2:1 to 2:2:1, respectively. Chromium is the dominant trivalent ion in most of the specimens which are, therefore, called “uvarovite.” There are also small amounts of pyrope and almandine. In Specimen No. 1 only, is grossularite the dominant “end member.”

The molecular ratios show considerable divergence from the ideal formula (3:1:3).

<table>
<thead>
<tr>
<th>Analysis</th>
<th>RO₂</th>
<th>R₂O₃</th>
<th>RO</th>
</tr>
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<tr>
<td>4</td>
<td>2.91</td>
<td>1</td>
<td>3.06</td>
</tr>
<tr>
<td>5</td>
<td>2.89</td>
<td>1</td>
<td>3.33</td>
</tr>
<tr>
<td>6</td>
<td>3.48</td>
<td>1</td>
<td>3.91</td>
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<tr>
<td>7</td>
<td>3.09</td>
<td>1</td>
<td>3.65</td>
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<tr>
<td>10</td>
<td>2.97</td>
<td>1</td>
<td>3.33</td>
</tr>
</tbody>
</table>

The unit cell contains an “excess” of Ca ions (Table II), and recasting the composition into “end members” also gives excess CaO that makes the true proportions of “end members” difficult to estimate. Betekhtin (1946, p. 69) also found calculation of uvarovite analyses unsatisfactory.

Vermaas (1952) experienced similar difficulties with manganese-iron garnet. He suggests that near agreement between determined and calculated specific gravity values in such garnets, indicates that excess atoms occupy positions within the unit cell. Since the measured and calculated specific gravity values of the uvarovite specimens are in good agreement,
### Table I. Chemical Analyses of Uvarovite

<table>
<thead>
<tr>
<th></th>
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<th>9</th>
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<tbody>
<tr>
<td>SiO₂</td>
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<td>—</td>
<td>33.50</td>
<td>34.64</td>
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<td>35.69</td>
<td>34.52</td>
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<td>TiO₂</td>
<td>0.38</td>
<td>n.d.</td>
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<td>1.97</td>
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<td>1.31</td>
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<tr>
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<td>—</td>
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<td>7.50</td>
<td>n.d.</td>
<td>n.d.</td>
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<td>6.30</td>
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<td>3.96</td>
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<td>MnO</td>
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<td>—</td>
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<td>MgO</td>
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<tr>
<td>CaO</td>
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<td>—</td>
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<td>—</td>
<td>100.01</td>
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<td>99.47</td>
<td>100.22</td>
<td>—</td>
<td>100.22</td>
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</table>

8. Kafferskraal, 915, Rustenburg district, Western Belt. Partial Analysis. Analysts W. H. Herdsman (Fe₂O₃, Cr₂O₃, FeO). J. J. Frankel (SiO₂, TiO₂, CaO). McLachlan & Lazar (H₂O+0.48%).
10. Doornbosch, 423, Lydenburg district, Eastern Belt. Total includes SrO 0.11%. Analyst W. H. Herdsman. FeO by F. J. de Wet.

The excess of RO group may also be accommodated in cavities in the garnet. Packing index is close to that given for garnets by Fairbairn (1933).

Alderman (1935) suggested that Si and Fe²⁺ ions are replaced by Al with Fe³⁺ entering the RO group. This replacement would account for excess R₂O₃ in almandine. Fleischer (1937) who showed that most garnets conform closely to the ideal ratio, thought that Alderman’s observed discrepancies might be due to under-estimation of ferrous oxide, so that more iron would be reported in the R₂O₃ group. Fleischer did not, however, deal specifically with uvarovite nor with garnets containing more than 0.5 per cent TiO₂ and it is in such garnets containing much TiO₂ or FeO that deviations from the ideal ratio are found.

Although, according to Kunitz (1936), Ti replaces Si, the exact behavior of Ti in garnet is not known with certainty and Zedlitz (1933) has
suggested that Ti may possibly be in the trivalent state and so cause a corresponding amount of FeO to be reported as FeO. In calculations of the molecular ratios of Bushveld uvarovite given above, all Ti has been added to Si, although all or part can occur as TiO$_2$ and Al can replace Si. Further calculations were made in which all the Ti was assigned to the FeO group with an equivalent amount of ferric iron subtracted from the iron of the RO group, and excess Al was added to Si. These results approached the ideal formula more closely, but the RO group still showed excess Ca.

Attempts to correlate chemical composition of garnet with physical

### Table II. Number of Atoms (0=96) in Unit Cell of Uvarovite

<table>
<thead>
<tr>
<th></th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
<th>10</th>
<th>Theoretical</th>
</tr>
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<tr>
<td>O (O)</td>
<td>96</td>
<td>96</td>
<td>96</td>
<td>96</td>
<td>96</td>
<td>96</td>
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<tr>
<td>Si</td>
<td>21.86</td>
<td>22.43</td>
<td>23.87</td>
<td>21.88</td>
<td>22.43</td>
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<td>Ti</td>
<td>0.97</td>
<td>1.09</td>
<td>0.20</td>
<td>1.23</td>
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<td></td>
<td>22.83</td>
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<td>24.07</td>
<td>23.11</td>
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<tr>
<td>Al</td>
<td>7.02</td>
<td>7.20</td>
<td>6.85</td>
<td>5.80</td>
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<tr>
<td>Fe$^{+}$</td>
<td>3.01</td>
<td>3.07</td>
<td>1.06</td>
<td>3.09</td>
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<td>—</td>
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<td>Cr</td>
<td>5.75</td>
<td>5.88</td>
<td>5.90</td>
<td>6.07</td>
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<td></td>
<td>15.78</td>
<td>16.15</td>
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<td>Fe$^{+}$</td>
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<tr>
<td>Mg</td>
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<tr>
<td>Ca</td>
<td>23.55</td>
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<td>24.74</td>
<td>27.10</td>
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### Table III. Specific Gravity, Optical and X-Ray Data for Uvarovite

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<tbody>
<tr>
<td>(± .005)</td>
<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>Calculated S.G.</td>
<td>3.767</td>
<td>3.758</td>
<td>3.767</td>
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<td></td>
<td></td>
<td></td>
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<td></td>
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<tr>
<td>R.J. (Na light)</td>
<td>1.783-</td>
<td>1.817-</td>
<td>1.824</td>
<td>1.833</td>
<td>1.830</td>
<td>1.801</td>
<td>1.832</td>
<td>1.828-</td>
<td>1.839-</td>
<td>1.837</td>
</tr>
<tr>
<td>(± .002)</td>
<td>1.786</td>
<td>1.820</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
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<td></td>
</tr>
<tr>
<td>α$_{0}$ (± .001 Å)</td>
<td>11.915</td>
<td>11.946</td>
<td>11.955</td>
<td>11.973</td>
<td>11.961</td>
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<td>11.989</td>
<td>11.955</td>
<td>11.969</td>
<td>11.975</td>
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<tr>
<td>Mol. wt.</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>480.0</td>
<td>485.4</td>
<td>482.2</td>
<td>493.4</td>
<td>—</td>
<td>—</td>
<td>488.8</td>
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<tr>
<td>Packing Index</td>
<td>6.2</td>
<td>6.3</td>
<td>6.2</td>
<td></td>
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</tbody>
</table>
J. J. FRANKEL
Taslr lV. X-R,rv PowrR SpAcrNG D.rra lon Uvenovtr
dAdAdA

2.9694   m-s  2.9751   m  4.2187  ?
2.6541   s   2.6594   s  2.9683   m-s
2.5334   w   2.5351   w  2.6535   s
2.4268   m-s  2.4280   m  2.5300   w
2.3293   m   2.3012   m-w  2.4268   m-s
2.1708   m   2.1728   m-w  2.3222   m-w
1.9317   m-s  1.9311   m  2.3138   m
1.8768   w   1.8851   m-w  1.9841   w
1.7175   m   1.7191   m-w  1.7223   m-w
1.6515   m-s  1.6536   m  1.6529   m-s
1.5928   s   1.5943   s  1.5910   s
1.4903   m   1.4991   m-w  1.4918   m-w
1.3343   m   1.3352   m  1.3343   m-w
1.3021   m   1.3035   m  1.3023   m
1.2726   m   1.2738   m  1.2720   m-w
1.2062   w   1.2058   w  1.2063   w
1.1095   m   1.1140   m  1.1083   m
1.0905   m   1.0922   m  1.0902   m
1.0559   m   1.0575   m  1.0560   m-w
0.99601  w   0.99839   m-w  0.99564   w
0.98237  w   0.98380   w  0.98236   w
0.96985  m-s  0.97108   m  0.97010   m
0.90188  vw   —   0.9 ±   ?
0.89156  m   0.89253   m  0.89148   m
0.88179  m-w  0.88280   m-w  0.88173   m-w
0.88137  m-w   —   —   —   —   —
0.86346  m   0.86460   m-w  0.86363   m-w
0.82971  m   0.83087   m-w  0.82995   w
0.82208  m-s  0.92324   m  0.82197   m
0.81436  s   0.81582   s  0.81453   s

w=weak m=medium s=strong.

properties have been made for three "end members" (Ford, 1915), and
correlation involving four "end members" has been reasonably success-
ful. Levin (1949, 1950) has shown that five components can account for
99 per cent or more of the composition of a garnet. By means of partial
chemical analysis and the three physical properties, refractive index
(R.I.), specific gravity (S.G.) and unit cell dimension (a0), the molecular
proportions of the five compositional molecules can be obtained by set-
ing up and solving five simultaneous equations. His investigation did
not include garnets containing the uvarovite "end member."
von Knorring (1951) showed that a direct relationship exists between the above three physical properties and the chromic oxide content in uvarovite. This is to be expected, because the replacement of Al$^{3+}$ (ionic radius 0.57 Å) by Cr$^{3+}$ (0.64 Å) or Fe$^{3+}$ (0.67 Å) must expand the lattice of grossularite as the composition approaches that of uvarovite or andradite. Although von Knorring made the first study of uvarovite in which full chemical analyses were correlated with the physical properties, he investigated three specimens only. It was hoped that the larger number of

![Fig. 2. Variation of unit cell with chromium content. Bushveld uvarovite specimens—Arabic numerals. von Knorring’s specimens—Roman numerals.](image)

samples available in the present investigation would confirm and extend his findings.

However, when chromic oxide content was plotted against $a_0$, S.G., and R.I. in the manner adopted by von Knorring, the results did not show the perfect linear relationships he obtained (Figs. 2, 3, and 4). Specimens which have Cr$_2$O$_3$ contents similar to those examined by him, have larger unit cell dimensions and higher R.I. values. Only Specimen No. 6 with a Cr$_2$O$_3$ content near that of von Knorring’s Sample No. 1 has similar physical properties; it is, however, low in titanium and iron. Titanium was not reported in von Knorring’s analyses, so that there is, presumably, little or none.

No linear relationship exists between unit cell dimensions and Cr$_2$O$_3$ contents in Bushveld uvarovite (Fig. 2). Zedlitz (1933) found a linear increase in the lattice constant of melanite with increase in TiO$_2$ and he suggested (1935) that one per cent of TiO$_2$ increases the unit cell by 0.01 Å. This expansion is thought to be due to replacement of Si by Ti which.
has a larger ionic radius. When the unit cell values of the Bushveld uvarovite are adjusted using this "correction," the amount of scatter of the plotted points of Fig. 2 is not appreciably reduced. The deviations from the linear relationship between unit cell dimension and \( \text{Cr}_2\text{O}_3 \) content are not due in any degree to titanium but rather to iron.

The effect of iron is shown by the difference between unit cell dimensions of von Knorring's low iron specimens and those of Bushveld uvarovite of similar \( \text{Cr}_2\text{O}_3 \) content. By calculation (total iron regarded as ferric oxide) one per cent of ferric oxide increases the unit cell dimension by 0.09 Å approximately.

Figure 3 shows a general increase in S.G. as \( \text{Cr}_2\text{O}_3 \) content increases. Where \( \text{Cr}_2\text{O}_3 \) contents are similar, S.G. differences are obviously due to variations in the amount of iron. In Specimen No. 9, for example, which contains only a small amount of titanium, the higher iron content offsets possible lower values of physical properties. High refractivity of iron raises the R.I., but titanium which has a low refractivity appears also to have some elevating effect on R.I. in many minerals for reasons not clearly known; as Fairbairn has remarked, other factors must be operative.

Figure 4 shows an almost linear relationship between R.I. and \( \text{Cr}_2\text{O}_3 \) content, roughly parallel to the straight line drawn by von Knorring, or
the line linking pure grossularite and uvarovite. It is clear that while the TiO₂ content has modified the R.I.—Cr₂O₃ relationship, the appreciable iron content has also raised the R.I. value.

The refractivities of Fe³⁺ and Cr³⁺ ions are probably similar, so that ferric oxide can be considered equivalent to chromic oxide. Accordingly when the amounts of ferric oxide and chromic oxide are combined for each analysis, and the values of “chromic oxide” plotted against R.I., the curve so produced is nearly parallel and closer to the curve obtained by von Knorring and also to that of grossularite-uvarovite.

The replacement of small by larger radius ions expands the unit cell while large ions like Ca in grossularite are surrounded by distorted polyhedra (Menzer, 1929). The present study supports the theory that the garnet structure is probably “elastic” like that of micas (Jaffe, 1951), and capable of taking up a wide range of ions.

Statistical methods of correlation of chemical composition and physical properties could not be applied to the small number of uvarovite occurrences for which both these factors are accurately known. It was not possible to use Levin’s algebraic method for the calculation of chemical composition from partial analysis and physical properties.

Quantitative color measurements were made on three specimens of minus 200 mesh uvarovite powder using a Donaldson Trichromatic Colorimeter. There is a slight increase in the red component for uvarovite of higher titanium and iron contents.

![Graph showing variation of refractive index with chromium content.](image-url)
The colors of minus 200 mesh powder of a representative number of specimens were compared with the matt surface of the British Trade Council Color Chart. The following are some of the closest color matchings.*

<table>
<thead>
<tr>
<th>Specimen No.</th>
<th>Reference Color</th>
<th>Limpet green</th>
<th>Georgian green</th>
<th>Bronze green</th>
<th>Leaf green</th>
<th>Woodpecker green</th>
<th>Moss green</th>
<th>Rockingham green</th>
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<tr>
<td>No. 1</td>
<td>Reference CC 97</td>
<td>0.382</td>
<td>0.439</td>
<td>0.017</td>
<td>0.382</td>
<td>0.439</td>
<td>0.017</td>
<td>0.382</td>
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<tr>
<td>No. 3</td>
<td>Reference CC 49</td>
<td>0.397</td>
<td>0.437</td>
<td>0.166</td>
<td>0.397</td>
<td>0.437</td>
<td>0.166</td>
<td>0.397</td>
</tr>
<tr>
<td>No. 5</td>
<td>Reference CC 84</td>
<td>0.375</td>
<td>0.452</td>
<td>0.173</td>
<td>0.375</td>
<td>0.452</td>
<td>0.173</td>
<td>0.375</td>
</tr>
</tbody>
</table>

Both the qualitative and quantitative measurements show that with increase in iron content the green color becomes deeper, while the uvarovite of higher titanium content has a “rusty” green color due to an increase in the amount of red or purple. The purple color of titan-augite is considered to be due to a similar effect.

The D.T.A. curve (Fig. 5, No. 1) shows that the uvarovite undergoes no change on heating to 1100°C.

![Graph showing D.T.A. curves of uvarovite and South African Jade](image)

**Fig. 5.** Differential thermal analysis curves of (1) Uvarovite, de Kafferskraal, No. 395. Table I, Analysis No. 7). (2) South African Jade, Buffelsfontein, No. 205. (Table VI, Analysis No. 1)

**The Petrography of the Uvarovite-Bearing Rocks**

Thin section examination showed that the relationship between uvarovite and chromite is always very much the same. The following account describes the features of those rocks from which the garnet had been separated and analyzed.

*All the color measurements were kindly made by Mrs. P. Angus-Leppan, Paint Industries Research Institute, University of Natal.*
No. 1. Lydenburg district. There is no chromite in the section which is made up of large plates (up to 1.8 mm. across) of slightly pleochroic, pale green orthopyroxene holding exsolution lamellae, and diopside (2Vγ 58°, cZ 40°, γ 1.706). The diopside may have replaced an earlier pyroxene. Very pale green garnet grains fully isotropic, 0.4 to 0.6 mm. diameter, replace both pyroxenes which are cloudy near the garnet.

No. 2. Klipfontein, No. 119. Large plates of colorless to pale green diopside 0.9 to 1.8 mm. across (2Vγ 58°, cZ 36°, γ 1.706), and markedly zoned orthopyroxene 2V, 68°, with a pale pink to green, pleochroic core rimmed by diopside, are partly replaced by uvarovite.

Fig. 6. A and B. Highly corroded chromite remnants (black) in uvarovite (stippled). Clear areas are mainly zoisite. Vygenhoek, No. 209. C. Chromite grains linked in short chains in uvarovite. A few small serpentinized olivine grains are present. de Kafierskraal, No. 359. D. Corroded chromite grains in uvarovite. de Kafierskraal, No. 359.

Highly corroded chromite grains 0.07 to 0.45 mm. across, lie in the irregular patches of garnet which are also peppered by chromite granules 0.01 mm. or less. The garnet is deeper green where it surrounds chromite grains. There is also a little interstitial zoisite.

No. 4. Vygenhoek, No. 209. The chromite grains 0.15 mm. average diameter, are isolated or lie in short, straight to curved chains in the deep green, granular uvarovite. The chromite is rounded and embayed and almost completely absorbed by the garnet (Fig. 6, A and B). Patches of zoisite (0.12 mm. across) partly replace the little diopside present. Small, brown-stained inclusions 0.02 mm.² in the garnet may be serpentinized olivine.

No. 5. Uitvalgrond, No. 71. The chromite is scattered as individual slightly rounded dodecahedral or octahedral grains 0.04 to 0.3 mm. in diameter, or as small clusters in a translucent, pale olive green garnet. The subhedral uvarovite grains range from 0.08 to 0.5 mm., with an average diameter of 0.15 mm. Small irregular patches of diopside and serpentine are interstitial to, or lie within the garnet or surround the chromite grains.
No. 6. Derdegelid, No. 141. The chromite grains (0.08 to 0.45 mm. diameter) are slightly rounded and show few corrosion effects even under high power. They lie in subhedral to euhedral grains of uvarovite. Patches of a lath-like mineral, probably zoisite, and a little diopside are interstitial to the garnet. Small, serpentinized granules, probably forsterite originally, are imbedded in the garnet.

No. 7. de Kafershraal, No. 359. In addition to isolated or small clusters of grains, the chromite crystals form short, roughly parallel chains (Fig. 6, C). Although the crystals are frequently euhedral decahedrons, 0.05 to 0.4 mm. in diameter, ragged grain margins demonstrate corrosion (Fig. 6, D). The dull green uvarovite of the same grain size as the chromite, contains a noticeable amount of small, elongate grains from 0.004×0.012 mm. to 0.027×0.054 mm. in size. The colorless grains generally have a brown surface stain and show low interference colors. They are considered to be serpentinized olivine. A small amount recovered during the uvarovite cleaning process gave a faint x-ray pattern resembling that of chrysotile and a spectrographic analysis showed magnesium and silicon as main constituents.* A few serpentine patches interstitial to garnet are present.

No. 8. Kafierskraal, No. 915. Euhedral chromite grains 0.07 to 0.25 mm. in diameter lie in curved or short, parallel chains in the green uvarovite matrix of similar grain size. There is little corrosion of the chromite grains which are sometimes surrounded by granules of diopside (?) 0.01 mm. or less in size. There are small inclusions resembling augite in the garnet and some interstitial serpentine.

No. 9. Hendriksplaats, No. 357. In the field this occurrence is vein-like and cuts through a chromitite seam (Hall, 1908, p. 60). The hand specimen is made up of patches of chromite crystals in the bright green uvarovite, and irregular, grey veins up to 5 mm. wide merge with larger aggregates of diopside crystals several cms. across.

Specimen No. 3 Winterveld, No. 424 is so similar to this material that the following account serves to describe both.

The pale green uvarovite grains average 0.1 mm. diameter and larger grains up to 0.3 mm. diameter are linked in distinct chains. Some grains which have color zoning show slight anomalous birefringence. The large garnet grains are intimately associated with colorless diopside (2V, 58°, 1 704) in plates several mm. across. Fragments of partly replaced diopside in the garnet are in optical continuity.

Chromite is scattered throughout the garnet as residual granules of a few microns and as larger, angular and embayed grains 0.25 mm. in diameter. The margins of the larger chromite grains are bordered by brown, translucent zones of replacing garnet which still contains some of the chromite substance (Plate 1 (1)). Chromite grains within pyroxene plates are bordered by granules of uvarovite (Plate 1 (2)). Small circular areas of augite 0.01 mm. diameter lie in the chromite grains. There are also interstitial patches of serpentine.

No. 10. Doornbosch, No. 423. Within the bright green garnet matrix, large, anhedral chromite crystals from 2 to 7 mm. across are linked in chains (Fig. 7), while most of the subhedral chromite grains 0.2 to 0.4 mm. in diameter lie between the chains in haphazard fashion. A colorless mineral of low R.I. (1.450) and birefringence about 0.002, fills some interstitial areas between, or forms extremely thin borders to chromite grains. It was determined as gmelinite from an x-ray photograph.† A little calcite interstitial to chromite is also present. Polished section shows a few chalcopyrite particles and silvery grains of 2 microns diameter may be hematite or platinoids.

* Kindly determined by Dr. S. A. Hiemstra, Union Geological Survey.
† Kindly determined by Dr. H. Heystek, National Chemical Research Laboratory.
Other uvarovite-bearing rocks. In the Winterveld-Hendriksplaats area of the Eastern Belt, thin, fine-grained chromitite seams of the "hard lumpy" variety of ore are separated by irregular patches and bands of green compact rock 0.3 to 5.0 cms. thick. They are similar to South African Jade in appearance but are duller and the hardness is frequently less than 6.5.

Under the microscope the green bands and irregular patches are translucent, colorless to grey, and composed mainly of laths of stocky crystals either in stellate or haphazard arrangement. There is a faint, ghost outline suggestive of original pyroxene plates. The thin, lath-like crystals, 0.1X0.02 mm., identified as diopside,* are associated with granular zoisite. Small, almost equant, colorless patches with radiating cracks and sheaf-like aggregates may be prehnite.

Small, pale green or yellow-green, subhedral uvarovite crystals (n=1.830) are scattered throughout the rock. The amount of garnet increases at the contact with the chromitite where replacement of the chromite is clearly demonstrated (Plate 1, (3A)). There is also a blue-green, anisotropic mineral of low birefringence which resembles the green uvarovite. It is probably a chromium-bearing hydrogrossular. It replaces chromite but is itself replaced by colorless chlorite and uvarovite which surround it (Fig. 8).

Chromite in individual grains or small clusters may be partly or completely replaced by the hydrogrossular, chlorite and uvarovite (Plate 1, (3B)). Occasional grey or pale cream, translucent patches of low interference color that have a ghost outline of original pyroxene are also thought to be hydrogrossular. Thin veins of later calcite and possible magnesite have partly replaced chains of uvarovite crystals and diopside matrix.

Two specimens of the green bands were partially analyzed. The material for Analysis No. 2 was treated in hot concentrated hydrochloric acid and then after water washing, strongly heated.

* Confirmed by an x-ray powder photograph taken by Dr. Heystek.
Fig. 8. Contact between diopside-rich band (di) and chromitite, showing corroded and replaced chromite (black) in green uvarovite (u), dark blue-green birefringent garnet (heavy stipple) and colorless chlorite (c).

<table>
<thead>
<tr>
<th></th>
<th>(1)</th>
<th>(2)</th>
</tr>
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<tbody>
<tr>
<td>SiO₂</td>
<td>39.8</td>
<td>49.2</td>
</tr>
<tr>
<td>R₂O₃</td>
<td>12.0</td>
<td>12.4</td>
</tr>
<tr>
<td>CaO</td>
<td>24.0</td>
<td>25.3</td>
</tr>
<tr>
<td>MgO</td>
<td>17.2</td>
<td>13.4</td>
</tr>
<tr>
<td>Ignition loss</td>
<td>7.2</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>100.2</td>
<td>100.3</td>
</tr>
</tbody>
</table>

Analyst J. J. Frankel.

1. Pale green band between chromitite seams, Winterveld, No. 424.
2. Acid and heat treated material from green band, Winterveld, No. 424.
Both analyses cast into “norms” give dominant diopside with zoisite, and for Analysis No. 1, some magnesium carbonate as well.

**CHROMITE ASSOCIATED WITH UVAROVITE**

The chromite crystals of the Doornbosch specimen (No. 10), have a wide grain size range known only in a few of the many chromitite outcrops. Because the crystals are not interlocked, this rock is very friable, and it disintegrates when lightly tapped with a hammer. This treatment, preliminary to the concentration of the uvarovite, fractured few of the larger crystals; any stray fragments were easily hand-picked from the finer crystals. After separation of uvarovite, the chromite was cleaned in hydrofluoric acid. The sample was sifted over Tyler sieves and four of the grades so produced were analyzed (Table V).
There is no well-defined grading in crystal size which would be expected had the chromitite been formed by crystal settling. The large crystals lie in scattered clusters or chain structures in the rock. It has been suggested on geochemical grounds that early formed chromite crystals are richer in Cr, Al and Mg, while later crystals have more Fe²⁺, Fe³⁺ and Ti (Malhotra and Prasada, 1956). This appears to be true for some chromitite seams where crystals at the base have more Cr₂O₃ than those at the top (Frankel, 1949). Except for the very slightly higher percentage of Cr₂O₃ in the coarse crystals, there are no distinct trends in the four graded fractions. It is possible, therefore, that the chromitite crystallized slowly from a re-fused chromitite (Sampson, 1932, p. 143); the larger crystals having formed first.

The analyses Nos. 1 to 4 resemble those quoted for the Main Seam of the Lower group on adjacent farms (de Wet, 1952, p. 150), but they differ from an analysis from Doornbosch farm itself (Kupferbürger et al., 1937, p. 25).

Chromite from the de Kafferskraal uvarovite-bearing chromitite has a lower Cr₂O₃ content than the majority of seams in the two groups identified in the Eastern Belt. The field relationships of this rock are obscure—it might belong to the Middle group in which chromite generally has less Cr₂O₃ than the underlying group of seams.


(3A) Uvarovite crystals in fine-grained diopside rock. Slightly corroded chromite grains are bordered by uvarovite and chlorite. Winterveld, No. 424. Plane polarized light.

(3B) Chromite grains in diopside rock replaced by colorless chlorite, blue-green hydrogrossular (?) and uvarovite. Winterveld, No. 424, Plane polarized light.

(4) South African Jade. Gabbro texture as seen in plane polarized light. Between crossed nicols this slide is fully isotropic.

**THE ORIGIN OF THE UVAROVITE**

Although some chromitites hold significant amounts of plagioclase, it was not the dominant constituent in any of the uvarovite-bearing chromitite specimens. Uvarovite formation, therefore, required the addition of
Interstitial serpentine could be derived from orthopyroxene, whereas the isolated small grains of serpentinized olivine, are similar to those found in impure magnesium limestones that have undergone low grade thermal metamorphism. This suggests that some magnesium carbonate was present long before uvarovite was formed. It could have been produced by the action of carbonated waters, perhaps of deuteric origin,
on enstatite. Forsterite would develop early with increase in temperature, and the introduction of calcium in heated waters and an increased metamorphic grade, would assist uvarovite formation. The moderate amount of aluminium required would be available from feldspar dissociation.

Thin bands of orthopyroxene between chromitite seams are also considered to have been partly converted to carbonate. Lime metasomatism and heat increase, assisted the formation of diopside and “grossularite,” and uvarovite was a final product.

Uvarovite is also associated with diopside and zoisite in veins cutting across chromitite seams. Betekhtin (1946, p. 70) reported a similar association which he attributed to pneumatolytic-hydrothermal agencies. Fortier (1946) has also described the formation of chlorite and uvarovite from chromite, and many other examples are on record.

Synthetic uvarovite is readily produced in an anhydrous environment at 855°C (Hummel, 1950). This temperature is obviously too high for the metamorphic/metasomatic processes outlined above for Bushveld uvarovite and associated minerals. Perhaps high pressure and the presence of water, lower the formation temperature. Nevertheless, it is clear that when chromium is available, the ugrandite garnet, uvarovite, forms relatively easily in nature in either hydrous or anhydrous environment.

It is suggested that in a magmatic body like the Bushveld Norite there would be local surges of heat coupled with slight dynamic effects, due to sagging in the body. These surges would give rise to simple metamorphic effects, contemporaneous or slightly later than the introduction of heated lime-bearing solutions.

There is also the possibility that the calcium came from small amounts of carbonate xenoliths derived from the floor of the Complex and which were completely digested in the Norite body in the vicinity of the Lower group of chromitite seams. This would account for the erratic and irregular nature of the uvarovite-bearing chromitites.

South African Jade (Hydrogrossular)

Uvarovite holding a large amount of the grossularite “end member” occurs with diopside and zoisite and a little disseminated chromite (Specimens Nos. 1 and 2 described above). Garnet, essentially grossularite, has been reported as a metamorphic mineral at the contacts of the Norite with underlying carbonate rocks from many localities in the Eastern and Western Belts (Kynaston, 1909, p. 20; Wagner, 1929, p. 175).

Hall (1925) described the rock prized as an ornamental stone, and known as South African Jade from the farms Turffontein No. 356 and Buffelsfontein No. 205 in the Western Belt, as a massive fine-grained
variety of grossularite. Although it contains no nephrite or jadeite, this material will be referred to as Jade, using the term in the general sense for compact tough minerals suitable for the making of ornaments. In hand specimen the rock is pale green to grey, massive and translucent with scattered chromite grains. It is quite distinct from the sparkling crystalline granular uvarovite in chromitite.

Thin sections are mainly colorless or very pale cream-brown and faintly cloudy. Except for faint straight "cleavage" traces or crystal faces, the garnet shows no crystal outlines, and between crossed nicols is either isotropic or gives grey interference colors of anomalous birefringence. Zoning and the remarkable twinning effects described by Hall, simulate twinning of albite.

In several slides, however, outlines of stout laths arranged with somewhat cloudy interstitial patches are seen in plane polarized light; between crossed nicols the whole rock may be fully isotropic (Plate 1 (4); Plate 2 (1, 2)). This ghost texture is identical to that of the feldspar-pyroxene associations commonly seen where the noritic rocks grade into anorthosite. A particularly good comparison can be made with the anorthositic layer associated with the Merensky Reef chrome band.

Partial transformation of pyroxenite to jade at the contact of a chromitite seam was observed. The small amount of feldspar and larger areas probably of clinopyroxene, were completely replaced by isotropic jade; the remaining pyroxene, dominantly enstenitic, is fairly fresh (Plate 2 (3, 4)). Only a few of the chromite grains show green chlorite-like margins. No zoisite was seen. However, a little zoisite is common as a well distributed mineral in many specimens. It is dominant in saussuritized anorthosite where feldspar (An_{62}) shows partial or complete alteration to saussurite which grades into a more clearly defined aggregate of small garnet crystals in the zoisite. This in turn grades into clear isotropic jade, in which no trace of original minerals is preserved.

Green isotropic rims around chromite are probably a true uvarovite. Many chromite grains, however, are either slightly corroded or completely replaced by jade.

Slightly wavy thin transparent bands alternate with somewhat less cloudy zones in many specimens. In thin section the white bands are slightly darker than the rest of the isotropic jade and they lie close to the clear-cut margins of saussuritized zones. Rhythmic replacement as jade developed from saussurite may account for the banding.

Hall's Analysis No. 5, of his specimen No. 616-c5 (Slide 1128) in spite of its high total, approaches the theoretical analysis of grossularite more closely than any of his other analyses. In thin section, this rock is colorless, clear and fully isotropic with no relict structures. Variations in the
other analyses may be attributed to small and variable amounts of zoisite and residual pyroxene and feldspar. The trace of chromium reported is due, either to the chromium in the garnet, or to scattered chromite grains.

Hall gave some physical data but made no comment thereon. In 1928 van der Lingen found that the transmission spectrum of the South African Jade showed the presence of OH groups in the molecule itself, and not in water of crystallization. He concluded that the material is not grossularite. Herbert Smith (1949, p. 312) stated that the specific gravity and refractive index of the jade have slightly lower values than those usually recorded for grossularite.

There is more combined water in all Hall's analyses than is required for the small amounts of zoisite. In the present investigation combined water and physical properties were determined on several different specimens. A complete chemical analysis was also carried out on clear isotropic jade which contains few scattered chromite grains and traces of zoisite.

It is concluded from the chemical analyses and physical properties that South African Jade is not true grossularite but hydrogarnet that

**Table VI. Chemical Analyses of Jade**

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<td>a₀</td>
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<td>(±.002)</td>
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2. “Green Jade,” Buffelsfontein. Total includes P₂O₅ 0.05%, H₂O — 0.20% (Hall, 1925).
has slight variations in composition. Hutton (1943) gave data for several specimens of the grossularite-hydrogarnet series which he named "hydrogrossular." Both Hutton and Yoder (1950) have suggested that the majority of garnets described as grossularite are really hydrogarnet, and Hutton says that the usual constituent of altered gabbros and related basic intrusive rocks is a member of the garnet-hydrogarnet series near the grossularite end. The South African hydrogrossular has the same variation trends of physical properties with change in water content as the New Zealand specimens.

### Table VII. Variation of Specific Gravity and Refractive Index with Combined Water Content in South African Jade

<table>
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<tr>
<th>% H₂O+ (samples dried at 105° C.)</th>
<th>0.83</th>
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<th>1.56</th>
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<td>1.728</td>
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<td>% SiO₂</td>
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<td>36.55</td>
<td></td>
<td></td>
<td>34.82</td>
</tr>
</tbody>
</table>

The D.T.A. diagram (Fig. 5, No. 2) shows that no change takes place during heating until the temperature reaches 1100° C. approximately. At this temperature the mineral probably begins to decompose in the solid state (c.f. Yoder, 1950, p. 239). There is no clear indication of the temperature at which water is lost. The diagram affords no evidence of an inversion from birefringent to isotropic jade.

Hall put forward two modes of origin for the jade—(1) magmatic and (2) metamorphic.

1. The material of the jade, derived from the norite magma, was later concentrated into bands, in a manner analogous to that of magnetite or chromite seam formation.

2. The garnet is a metamorphic-metasomatic product resulting from intensive alteration of an original calcareous aluminous sediment (like a calcareous/dolomitic marl) upstopeed from the sedimentary floor of the Complex and brought into the present position as xenoliths within the Norite body.

Alteration into massive lime garnet from an original aluminous calcareous rock implies an increase in silica and alumina, and removal of carbon dioxide. Hall suggested that carbon dioxide could be lost because the thin bands of the rock were engulfed in a great thickness of magma and the addition of other elements could be due to a metasomatic replacement caused by magmatic infiltration.

van Biljon (1955, p. 132) stated that the composition of the jade suggests that it is an intermediate product of the metasomatism which trans-
formed sedimentary carbonate rocks to anorthosite. Thin section, however, has convincingly revealed that feldspar laths \((\text{An}_{63})\) are transformed progressively with crystal outlines preserved, through zoisite to hydrogrossular. If the feldspar had formed from the hydrogrossular it is unlikely that well-defined crystal boundaries would develop before the whole of the enclosed volume had been converted to feldspar. In any event, the palimpsest textures of anorthosite and norite do not support van Biljon's theory.

The fairly uniform thickness of the jade layers "interbedded" with anorthosites, pyroxenites and chromite horizons, points simply to their being metasomatized equivalents of such rocks \textit{in situ}; access for calcium-bearing solutions being provided by the planes of pseudo-stratification or rifting. The dominant cations in the original minerals were Al, Ca with less of Na in the feldspar, and Mg with some \(\text{Fe}^{2+}\) in the orthopyroxene. Additional lime with a small amount of water could displace magnesia and silica. Both Yoder (1950) and Hummel (1950) stress that water acting as a catalyst may possibly assist the formation of even anhydrous grossularite in hydrothermal transformations.

Many authors have pointed out (Harker, 1939, p. 174) that saussuritization of feldspar belongs to a late stage in the cooling down of an igneous rock, so that grossularite and zoisite may have formed during this period when some stress may also have operated. This would afford a simple explanation for jade formation. However, it is difficult to understand why only certain zones of the anorthosites and pyroxenites should have been saussuritized, and a source of additional lime would still be necessary for the production of jade from pyroxenite and norite.

Several kinds of lime metasomatism are listed by Turner and Verhoogen (1951, p. 488) and the type described in the present paper has been previously recorded from New Zealand. Dykes containing diallage, prehnite and grossularite have been shown by Grange (1927) to be altered gabbrros really, with grossularite and prehnite secondary after feldspar. He suggested that grossularite-diopside veins were formed by solution which took up lime, magnesia and alumina from pyroxene, but direct evidence of the transformation to grossularite was lacking.

Turner (1933) suggested that in pyroxenites the reaction products are stress minerals, and with aqueous solutions OH group minerals would form. Turner quotes Benson who thought that "rodingite" was due to the action of concentrated magmatic water under high pressure, and that lime-silicate rocks from New South Wales were transformed by calcium-rich aqueous solutions which came from a cooling peridotite mass; monoclinic pyroxene was suggested as the source of the calcium.

Carbonate rocks in the floor of the Bushveld Complex below the jade
occurrence on Buffelsfontein are a nearby source for the additional calcium required in the transformation to "garnet." This calcium could have been introduced by heated waters.

It is considered, therefore, that the South African Jade is a metamorphic replacement of anorthosite, pyroxenite, and of the gangue minerals of the chromitite seams.

Conclusions

Both uvarovite and hydrogrossular have developed within the same zone of the Norite body, although the modes of occurrence differ. Uvarovite is intimately associated with chromitite and pyroxenite whereas the jade lies in anorthosite. It would appear, therefore, that the original composition of the rocks and the subsequent metasomatic/metamorphic processes have determined the variety of garnet developed.

Yoder failed to produce grossularite synthetically, however, and found as other workers had, that only hydrogrossular formed relatively easily in a hydrous environment. Hummel was able to prepare anhydrous uvarovite without difficulty. Undoubted anhydrous grossularite does exist in nature; it is possible that at high temperatures and elevated pressures hydrogrossular may be made over to grossularite.

In the formation of the jade, fairly uniform thicknesses of anorthosite have been saussuritized and converted to hydrogrossular in situ. The pale green tint is due to chromium from small amounts of dissolved chromite grains. In rare instances chromite is rimmed by true uvarovite which suggests local temperature increase. Jade formation is not dependent on the presence of chromium.

It is unlikely that, as suggested by Hall, the fairly uniform jade layers represent actual limestone horizons torn up from the floor of the Norite body and brought to their present position where they were transformed. Rather, it is visualized that anorthosite bands were transformed by heated calcium-bearing solutions.

Hummel found that a hydrous environment is not necessary for uvarovite formation, and none of the Bushveld uvarovite contains a significant amount of combined water. The associations show that the uvarovite formed essentially in pyroxenite and pyroxene-bearing chromitite as a result of deuteric alteration and lime metasomatism, followed by thermal metamorphism. Uvarovite formation was entirely dependent upon the presence of adequate amounts of chromium.

The chromite in the jade is relatively unattacked whereas it is generally corroded in the uvarovite-bearing rocks. It is suggested that this also points to higher temperatures at the time that the uvarovite was formed.

The complete digestion of small carbonate xenoliths within the chro-
mitnite zone may explain the irregular and erratic distribution of the uvarovite and such a possibility, of course, would allow of an additional source of lime.

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