Red and green labradorite feldspar from Congo

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Abstract: Two red and one green transparent cut feldspars from the Democratic Republic of Congo have been investigated and are compared with similar material from Oregon (USA). The specimens from Congo display a deeper colour saturation, which make these new findings very attractive gemstones. The studied specimens belong to the labradorite variety of plagioclase feldspar. Microscopically the stones are rather clean, and only some twin lamellae, hollow channels and fine inclusions are present.

The red and green colours in labradorites from both Congo and Oregon are due to copper, and the differences in colour are attributed to tiny copper colloids of different size. The Congo labradorites contain about 20× more copper than the Oregon stones.

General considerations about commercial terms and mineralogical nomenclature of feldspars are presented.

Keywords: Congo, copper, feldspar, labradorite, plagioclase

Introduction and sample description

Recently, the SSEF Swiss Gemmological Institute received two feldspars from Dr Laurent Sikirdji (Gemfrance, St.-Ismier, France) for testing. Specimen RC1 (1.967 ct) is intensely red and specimen GC1 (4.856 ct) is of a saturated green colour. A similar red specimen (RC2) of 2.969 ct was provided by Peter Salzmann (Lucerne, Switzerland). The location of the studied specimen is not yet exactly known; however, they are said to originate from the Democratic Republic of Congo (L. Sikirdji, pers. comm., 2003). The author has seen several faceted red feldspars from Congo up to 10 ct during the 2003 Tucson show. These feldspars (Figure 1) strongly resemble sunstone labradorite from Oregon in the USA (Pough, 1983; Hofmeister and Rossman, 1985; Johnston et al., 1991), but display a more vivid and saturated colour than the latter. The occurrence of red and green plagioclase is not uncommon. Hofmeister and Rossman (1985) and Johnston et al. (1991) described in detail that such colours may even occur within a single specimen, often accompanied by a spangling effect (auroscent or schiller) as a result of oriented small red platelets. The red and green samples examined in our study showed no spangling effect (auroscent), although Fritsch (2002) observed such an effect in stones from the same general location.
On the market (Tucson 2003), the red and green stones from Congo have been offered as red andesine (an intermediate member of the plagioclase feldspar solid solution series). In the literature, gem-quality red andesine from Congo was first mentioned by Fritsch (2002). Below, physical, chemical and spectroscopic data of the studied samples are compared to similar feldspars from other sources (Hofmeister and Rossmann, 1985; Johnston et al., 1991; Lairs, 2002). Then the nomenclature of sunstone feldspar is discussed, taking into account the controversy among gemmologists in 1992 (Gübelin, 1992; Hanneman, 1992; Hoover, 1992; Johnston, 1992; Liddicoat, 1992a, 1992b).

Table 1: Gemmological properties of labradorite and andesine from Congo and Oregon (USA).

<table>
<thead>
<tr>
<th>Labradore from Congo</th>
<th>Andesine from Congo</th>
<th>Labradore from Oregon</th>
</tr>
</thead>
<tbody>
<tr>
<td>(this study)</td>
<td>Fritsch (2002)</td>
<td>(this study)</td>
</tr>
<tr>
<td>weight</td>
<td>1.97 to 4.856 ct</td>
<td>1.04 to 8.95 ct</td>
</tr>
<tr>
<td>SG</td>
<td>2.68 to 2.70</td>
<td>2.67</td>
</tr>
<tr>
<td>RI</td>
<td>RIo: 1.553 to 1.555</td>
<td>1.551</td>
</tr>
<tr>
<td></td>
<td>RLy: 1.562 to 1.563</td>
<td>1.560</td>
</tr>
<tr>
<td>birefringence</td>
<td>0.007 to 0.011</td>
<td>0.009</td>
</tr>
<tr>
<td>optic nature</td>
<td>biaxial (positive)</td>
<td>biaxial (negative)</td>
</tr>
<tr>
<td>colour</td>
<td>red (RC1 and RC2)</td>
<td>red</td>
</tr>
<tr>
<td></td>
<td>and green (GC1)</td>
<td></td>
</tr>
<tr>
<td>pleochroism</td>
<td>very weak (red samples RC1 and RC2) to distinct (green sample GC1)</td>
<td>very weak</td>
</tr>
<tr>
<td>transparency</td>
<td>transparent</td>
<td>transparent</td>
</tr>
<tr>
<td>LW-UV</td>
<td>weak to distinct orange</td>
<td>weak to medium orange</td>
</tr>
<tr>
<td>SW-UV</td>
<td>none</td>
<td>weak red</td>
</tr>
<tr>
<td>observations</td>
<td>milky turbidity</td>
<td>‘schiller’ effect</td>
</tr>
<tr>
<td></td>
<td>green labradorite: red under incandescent light</td>
<td></td>
</tr>
</tbody>
</table>
Gemmological properties

The samples were first studied using classical gemmological equipment and methods, namely, the refractometer, hydrostatic balance, polariscope, long- and short-wave ultraviolet light (LW-UV and SW-UV) and microscope. RC1, RC2 and GC1 showed more or less equal values for refractive indices, birefringence and specific gravity (Table 1). All samples were determined as optically biaxial positive. The red specimens (RC1, RC2) showed only very weak pleochroism, whereas the green sample (GC1) presented a distinct pleochroism from greenish-yellow to bluish-green. Hofmeister and Rossman (1985) also found only minor pleochroism in the red sunstone labradorites from Oregon (USA), but a distinct dichroism in their green samples. Under LW-UV, the red samples showed a weak orange glow, whereas the green stone appeared distinctly orange. Under SW-UV, no fluorescence was observed in RC1, RC2 and GC1, in contrast to Fritsch (2002) who reported a weak red fluorescence and even some surface related bluish luminescence in his stones from Congo.

Specimen RC1 shows a step-like healing fissure along a cleavage direction. RC2 (Figures 2 and 4) contains several distinct twin lamellae and a few parallel hollow channels; in some parts along but not within the twin lamellae, very tiny particles were observed. These particles are responsible for a general milky turbidity, which may be concentrated in zones. No reflecting inclusions producing aventurescence were observed.

The colour of the green stone (GC1) is distinctly lamellar zoned. With high magnification, small parallel fluid channels and trails of tiny inclusions are visible. Furthermore, the specimen shows a milky turbidity and a reddish hue in white incident light (Figure 3).

Microscopic observations

All samples are rather clean with only a few inclusions. The red stones show a relatively uniform colour distribution, which was confirmed in immersion liquid.

UV-Vis spectroscopic analyses

Spectra were recorded with a Hitachi U-4001 spectrophotometer from 800 to 280 nm (UV-Vis) and from 2500 to 280 nm to record absorption in the near-infrared region.
The red specimens (Figure 5a) were measured without a polarizing filter, as only very weak pleochroism was observed. The UV-Vis spectra are characterized by a general increase in absorption towards ultraviolet (strong UV tail). However, two distinct absorption bands at 565 and 380 nm are present. In Figure 5a the spectra of RC1 and RC2 are compared with a pale red sunstone labradorite (RO1) from Oregon (USA).

The green sample (Figure 5b) shows two different absorption curves for the yellowish-green and bluish-green directions. The yellowish-green direction is a rather flat curve with a broad minimum at 620 nm and a small absorption band at about 430 nm, partly masked by the steep increase of absorption towards ultraviolet (total absorption at ~370 nm). The bluish-green direction shows no distinct minimum at 620 nm, and a broad absorption band is centred at 680 nm. The spectra of green labradorite from Oregon (sample GO1) are somewhat similar. However, if the bluish-green and green direction spectra are compared (Figure 5b), the broad absorption band is shifted towards 620 nm and is less pronounced. In the light red direction, no minimum is found at 620 nm, but there is a general decrease in absorption towards the near-infrared. Furthermore, the green labradorite from Oregon shows not only absorption at 420 nm, but also two bands at 450 nm (weak) and 380 nm.

Discussion of UV-Vis spectra and colour cause

According to Hofmeister and Rossman (1985) the colour in labradorites from Oregon (USA) is correlated with the concentration of copper. Red labradorites show the highest copper content (150-200 ppm CuO), whereas yellow varieties contain the lowest concentrations (0-40 ppm).
The colour is further dependent on Cu exsolution temperature, rate of Cu diffusion and aggregation (Cu\(^0\) colloids) and state of copper (Cu\(^0\) or Cu\(^+\)).

(a) Red labradorites

After Hofmeister and Rossman (1985), the band at 560 to 585 nm combined with a general increase of absorption towards ultraviolet is attributed to intrinsic absorption by tiny Cu\(^0\) particles which are too small to scatter light (< 22 nm), and produces a red colour. The band-shift from 560 nm towards shorter wavelengths, coupled with a broadening of the absorption band is related to increasing size of the Cu\(^0\) colloids. As the spectra of the red samples from Congo (RC1 and RC2) are very similar to that from Oregon (RO1) in the 565 nm spectral region, the copper colloids are most probably of a very similar size and measure only a few nanometres (Hofmeister and Rossman, 1985).

According to Hofmeister and Rossman (1985), the bands at 380, 420 and 450 nm are due to Fe\(^3+\) in tetrahedral sites in the feldspar structure. In the samples from Congo, the band at 380 nm is present but smaller, and those at 420 nm and 450 nm are absent. The latter, however, may be masked by the strong UV absorption, probably resulting from a much higher copper concentration in stones from Congo (up to 0.329 wt.% CuO in RC1) compared to material from Oregon (0.015 wt.% CuO in RO1). When extending the spectra into the near-infrared (NIR) up to 2000 nm, the samples from Congo show a broad absorption band at about 1260 to 1310 nm. According to Hofmeister and Rossman (1985), this absorption band can be attributed to Fe\(^2+\).

(b) Green labradorites

In 1985, Hofmeister and Rossman presented several models involving tiny copper clusters (i.e. pairs of Cu\(^0\) atoms) or Cu\(^0\) – Cu\(^+\) intervalence charge transfer to explain the green colour, but still the cause of the green colour in copper-bearing labradorite remains rather an enigma.

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**Figure 5b**: Polarized UV-Vis spectra of green labradorite from Congo and Oregon (USA). Both labradorites are characterized by distinct pleochroism.
Table II: Chemical compositions of labradorites from Congo and Oregon, USA.

<table>
<thead>
<tr>
<th>Sample Colour Origin</th>
<th>RC1* red</th>
<th>RC2* red</th>
<th>GC1* green</th>
<th>RO1 pale red</th>
<th>Johnston 1991 pale red</th>
<th>GO1* green</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO₂</td>
<td>53.865</td>
<td>55.958</td>
<td>57.570</td>
<td>51.56</td>
<td>51.83</td>
<td>52.370</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>30.182</td>
<td>28.139</td>
<td>25.612</td>
<td>30.23</td>
<td>30.51</td>
<td>31.018</td>
</tr>
<tr>
<td>Na₂O</td>
<td>5.051</td>
<td>4.950</td>
<td>4.914</td>
<td>3.78</td>
<td>3.74</td>
<td>3.744</td>
</tr>
<tr>
<td>K₂O</td>
<td>0.195</td>
<td>0.097</td>
<td>0.098</td>
<td>0.02</td>
<td>0.11</td>
<td>0.197</td>
</tr>
<tr>
<td>MgO</td>
<td>b.d.</td>
<td>b.d.</td>
<td>b.d.</td>
<td>0.16</td>
<td>b.d.</td>
<td>b.d.</td>
</tr>
<tr>
<td>TiO₂</td>
<td>0.149</td>
<td>0.160</td>
<td>0.141</td>
<td>0.02</td>
<td>0.043</td>
<td>0.081</td>
</tr>
<tr>
<td>Fe₂O₃</td>
<td>0.448</td>
<td>0.470</td>
<td>0.549</td>
<td>0.48</td>
<td>0.03</td>
<td>0.641</td>
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<tr>
<td>MnO</td>
<td>0.001</td>
<td>0.002</td>
<td>0.004</td>
<td>0.06</td>
<td>0.010</td>
<td>0.021</td>
</tr>
<tr>
<td>Cr₂O₃</td>
<td>0.007</td>
<td>b.d.</td>
<td>b.d.</td>
<td>n.a.</td>
<td>n.a.</td>
<td>0.010</td>
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<tr>
<td>V₂O₅</td>
<td>0.007</td>
<td>b.d.</td>
<td>0.006</td>
<td>n.a.</td>
<td>n.a.</td>
<td>b.d.</td>
</tr>
<tr>
<td>CuO</td>
<td>0.329</td>
<td>0.205</td>
<td>0.260</td>
<td>0.015*</td>
<td>0.011</td>
<td>0.010</td>
</tr>
<tr>
<td>ZnO</td>
<td>b.d.</td>
<td>0.001</td>
<td>0.001</td>
<td>n.a.</td>
<td>n.a.</td>
<td>0.006</td>
</tr>
<tr>
<td>Total</td>
<td>100.00</td>
<td>100.00</td>
<td>100.00</td>
<td>99.83</td>
<td>99.73</td>
<td>100.00</td>
</tr>
<tr>
<td>anorthite%</td>
<td>52</td>
<td>53</td>
<td>55</td>
<td>66</td>
<td>66</td>
<td>64</td>
</tr>
<tr>
<td>albite%</td>
<td>48</td>
<td>47</td>
<td>45</td>
<td>34</td>
<td>34</td>
<td>36</td>
</tr>
</tbody>
</table>

* determined with ED-XRF
n.a. not analysed; b.d. below detection limit

The green colour in labradorite sample GC1 from Congo is also attributed to copper and, as in the red labradorites, this causes strong absorption towards the ultraviolet. The band at 430 nm is attributed to Fe³⁺, whereas the other iron bands at 450 and 380 nm, which are present in the UV-Vis spectrum of green labradorite from Oregon, may be masked by the strong absorption. Again, this is due to the higher copper content of the Congo stones compared to those from Oregon (0.26 vs. 0.01 wt.% CuO respectively).

The difference in pleochroism of green labradorite from Oregon (GO1) and the specimen from Congo (GC1) is clearly illustrated in the spectra. In sample GC1, the minimum at 620 nm combined with an increase in absorption towards the near infrared (800 nm) is responsible for the yellowish-green hue, whereas the green specimen from Oregon (GO1) shows a constant decrease in absorption towards the near-infrared, thus resulting in a pale red pleochroic colour.

When using incident light, the red specimens from Congo display an off-white milky turbidity (Figure 2), which is attributed to scattering (Steffen, 2000) by tiny particles (probably copper) apparently not large enough to produce aventurecence. Similar incident illumination of the green sample (GC1) produces a distinct red, most probably due to the presence of relatively larger copper colloids (a few nm to 22 nm in diameter); these are also thought to be responsible for the colour of the red labradorites RC1 and RC2. These colloid clusters add their red component to the overall green colour, produced by smaller Cu⁰ clusters and possibly Cu⁰ – Cu⁺ intervalence charge transfer.

In conclusion, it can be stated that the causes of colour in these feldspars are quite complex. Hofmeister and Rossman (1985) and Johnston et al. (1991) established that 100 ppm copper is sufficient to cause a red colour in sunstone labradorite from Oregon. In the studied samples from Congo, the Cu con-
concentration exceeds this value by a factor of >20 (2000 - 3300 ppm Cu), and could explain the much more pronounced UV absorption of these stones.

Chemical investigations

Semi-quantitative chemical analyses were carried out with a Spectrace 5000 Tracer ED-XRF on three cut stones from Congo. The results are listed in Table II, together with analyses of Oregon stones. Apart from the main constituents Si and Al, the specimens reveal calcium dominating sodium (about 10 wt.% CaO versus 5 wt.% Na₂O). Potassium is only a minor constituent. Furthermore, all samples contain minor amounts of copper (up to 0.33 wt.% CuO), iron (0.55 wt.% Fe₂O₃), titanium, and traces of Mn, Cr, V and Zn.

Most feldspars may be classified in the ternary K-Na-Ca-diagram (Figure 6) with the end members orthoclase (potassium-feldspar), albite (sodium-feldspar) and anorthite (calcium-feldspar). The classification of feldspar is rather complex, involving variation of both chemical composition and structural state (ordering of Si and Al on distinct tetrahedral sites), the latter depending on the temperature of crystallization and on subsequent thermal history (Deer et al., 1992; Hänni, 1989). At high temperature, there is solid solution between albite and orthoclase (alkali feldspar series) and between albite and anorthite (plagioclase series), respectively. At lower temperature, there are some miscibility gaps, which result in exsolution of different feldspar phases, often causing optical effects such as iridescence and light scattering. The plagioclases are classified chemically into the varieties albite, oligoclase, andesine, labradorite, bytownite and anorthite, characterized by a gradual increase in RI and SG towards the calcium end member anorthite. The intermediate plagioclase varieties are basically defined for convenience and have no structural significance (Deer et al., 1992).

The investigated feldspars from Congo are very low in potassium and thus belong to the plagioclase series. When calculating their composition and their place in the plagioclase solid solution series, the Congo feldspars lie in a range slightly over the 50% mark of the anorthite component, which is the limit between andesine and labradorite. In Figure 6 they are plotted together with labradorite samples from Oregon. Their position in the labradorite field is near the andesine boundary, and thus they may be considered as sodic labradorite, in contrast to the calcic labradorites from Oregon. This conclusion is supported by RI and SG values, which are at the lower end of the labradorite range. As these samples from Congo are very close to the andesine/labradorite boundary, it is possible that further sampling of red and green feldspars from Congo may reveal a wider range of chemical compositions which could extend into the andesine field of the ternary feldspar plot (Fritsch, 2002; L. Sikirdji, pers. comm., 2003).

Figure 6: Ternary feldspar plot indicating the composition of the samples from Congo and Oregon (USA).
The labradorites from Congo are in many aspects very similar to the material from Oregon (USA), but have a lower anorthite content (less Ca, see Table II), also reflected by slightly lower RI values (Congo: RI 1.553 to 1.563; Oregon: RI 1.560 to 1.572).

**Discussion of feldspar nomenclature in gemmology**

There are many commercial names for different feldspar varieties such as sunstone, moonstone, spectrolite, amazonite, etc. These names are generally based on the appearance of the feldspar, but more or less connected also to the chemistry, structure and inclusions. The discussions about sunstone or heliolite as a commercial name for the material from Oregon (Gübelin, 1992; Hanneman, 1992; Hoover, 1992; Johnston, 1992; Liddicoat, 1992a, 1992b) have illustrated that the use and correct designation of names remains a matter of discussion in the gemmological community.

Since the term sunstone is merely a matter of appearance, describing golden to red feldspars with more or less schiller (auratescence) due to tiny inclusions (e.g. hematite platelets, copper colloids), it is well fitted to be used in the trade for many varieties of feldspar in both the plagioclase and alkali feldspar ranges (see Hänni et al., 2003). It is suggested, however, that the commercial name should be followed by the correct mineralogical name, e.g. sunstone labradorite, variety of feldspar.

The term moonstone, however, should be restricted to alkali feldspars (Deer et al., 1992; Hänni, 1989) showing a filmy white to pale blue sheen; this is due to scattering of light from tiny domains caused by perthitic exsolution (irregularly-shaped intergrowth of potassium-rich and sodium-rich feldspar). Stones resembling these but with a plagioclase composition (also called peristerites) may display a white sheen or a rainbow-coloured iridescence, depending on the sizes of the submicroscopic intergrowths of the calcium-rich and calcium-poor plagioclase lamellae (Deer et al., 1992). When the lamellae are thick enough, multiple reflections produce iridescence (labroaescence). If all spectral colours are visible, these iridescent plagioclases (labradorites) may be called spectrolite, regardless of how much ore mineral inclusions are providing a dark background. Such iridescent plagioclases should not be called moonstones for three reasons, they differ not only in appearance (pale blue sheen vs. iridescence), but also in their structure (perthitic exsolution vs. peristeritic lamellar intergrowth) and chemical composition (alkali feldspar vs. plagioclase feldspar).

In conclusion: it is recommended that commercial names for feldspar varieties should be linked with the correct mineralogical names to avoid any confusion.

**Acknowledgements**

The author would like to thank Dr Laurent Sikirdji (Gemfrance, St.-Ismer, France) and Peter Salzmann (Lucerne, Switzerland) for the loan of samples. ED-XRF analyses were kindly done by Peter Giese at SSEF Swiss Gemmological Institute. Further thanks go to Professor Dr Henry A. Hänni who provided reference samples from Oregon and other sources and to Dr Lore Kiefert who reviewed this manuscript (both from SSEF Swiss Gemmological Institute).

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