GEM-QUALITY GROSSULAR GARNETS

By D. Vincent Manson and Carol M. Stockton

The gemological classification and identification of gem grossular garnets is examined through the study of 105 gem-quality grossulars. These specimens were measured for refractive index, specific gravity, absorption spectrum, color, and chemical composition. From these data, the authors were able to reexamine the ranges of physical and optical properties that are characteristic of the gem species grossular. In addition, they discuss the problems encountered in defining the two gem varieties of grossular, tsavorite and hessonite.

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Grossular garnet occurs in a diversity of colors, including tones of orange, yellow, and green. Until recently, this garnet species encountered little popularity as a gemstone, and then only in its brownish orange variety hessonite. About 10 years ago, however, a large deposit of vivid green grossular was discovered in east Africa (Bridges, 1974). Popularization of this material followed the coining of the trade name "tsavorite" (now accepted as a variety by mineralogists), and grossular emerged as a significant gem species. More recently, another new east African garnet, tagged with the trade name "malaya" and easily confused with hessonite, has brought new attention to bear on grossular (Stockton and Manson, 1982).

Hessonite and tsavorite illustrate the inconsistency that surrounds the definition of many gem varieties. *Hessonite* is the name traditionally applied to the yellow, orange, or brown transparent variety of grossular. However, color descriptions of hessonite are varied and vague: "cinnamon-colored" and "yellow" (Dana, 1911), "yellowish and brownish red" (Deer et al. 1963), "brownish-yellow, through a brownish-orange to aurora-red" (Webster, 1975), "orange-brown" (Anderson, 1959), "light yellow to dark yellow shades" (Arbuniés-Andreu, 1975), "yellow-brown to orangy-brown" (Shipley, 1974), "orangy-yellow to orangy-brown" (Liddicoat, 1981). In addition to the lack of agreement we encountered on the precise range of hues associated with this variety, nowhere could we find mention of the saturation of color to which these various hues referred.

The original description of tsavorite as being similar in color to emerald (Bridges, 1974) is also rather vague. At what point does green grossular have sufficient depth and intensity of color to be considered tsavorite? This is much the same as the familiar question, At what stage does green beryl become emerald? The net result of this lack

Figure 1. This selection of stones used in the grossular portion of the garnet study illustrates the range of colors examined.

of agreement with regard to the colors of the gem varieties of grossular is confusion among gemologists, understandably, as to the correct applications of these varietal terms.

The final goal of our study and review of garnets (Manson and Stockton, 1981; Stockton and Manson, 1982) is to provide clear, functional definitions of all the gem species and gem varieties of the garnet group. The size of this problem required that we divide the subject into a series of descriptive papers to be followed by a concluding article that proposes a cohesive gemological classification of the garnets on the basis of the data gathered. This study of the transparent grossular garnets continues the garnet project. Inasmuch as grossular has been well documented both mineralogically and gemologically, this portion of our study serves primarily to confirm previous work on the grossulars and to supply data (such as color description) not provided in earlier reports.

DATA COLLECTION

Stones were selected for study on the basis of gem quality, transparency, and colors that are typically associated with grossular. Chemical analyses from a previous study (Stockton and Manson, 1982) confirmed the identity of stones that might visually be mistaken for some other type of garnet. The resulting collection contained 105 specimens ranging from colorless through pale to intense hues of yellow, green, and orange (figure 1). Although orange grossulars were examined as part of the paper just cited, we included them here as well in order to compare all colors of grossular. Not included in this study are the massive, translucent materials (including pink and green) that in many cases actually belong to the separate species hydrogrossular (Deer et al., 1963). There have been cases of translucent grossular reported (Bank, 1980), but this material is ornamental rather than gem quality.

Refractive index, specific gravity, spectrum, color, and chemical composition were determined for the 105 garnets selected. The instruments and methods used to obtain these data were described fully in a previous article in this journal (Manson and Stockton, 1981). The data gathered on the 105 gem garnets used in this study will be published at a later date, upon completion of the entire GIA garnet project, and are summarized below. Specific data will be provided on request to the authors.

DISCUSSION OF DATA

Physical and Optical Data. Comparison of the ranges we obtained for refractive index and specific gravity with the values quoted by three other gemological references (table 1) reveals that the ranges we found in our 105 samples are somewhat broader than one might presume on the basis of the gemological literature. A graph of the relationship between refractive index and specific gravity among our 105 specimens (figure 2) suggests that, of the nongrossular components present, andradite appears to have the strongest effect on the departure of these properties from the values for pure grossular: 1.731 (McConnell, 1964) and 3.594 (Skinner, 1956), respectively. However, the scattering of points to either side of the grossular-andradite trend suggests that other influences, such as other end-member components, are present, as one might expect (Ford, 1915; Fleischer, 1937).

Comparison of our data with references in the mineralogy literature sometimes reveals discrepancies that result from the particular nature of our samples. Gem specimens are relatively inclusion-free, transparent, single crystals, while mineralogical specimens are frequently fractured and included such that they are not transparent even as single crystals. For example, data cited by Ford (1915) resulted in a low estimate of the specific gravity of pure grossular (3.530 rather than the 3.594 found by Skinner in 1956). It is recognized now that, as stated by Deer et al. (1963, p. 81), "garnets commonly have small inclusions of quartz or other minerals which cause the composite grain to have a low specific gravity." In fact, their data reflect this bias when compared with our results.

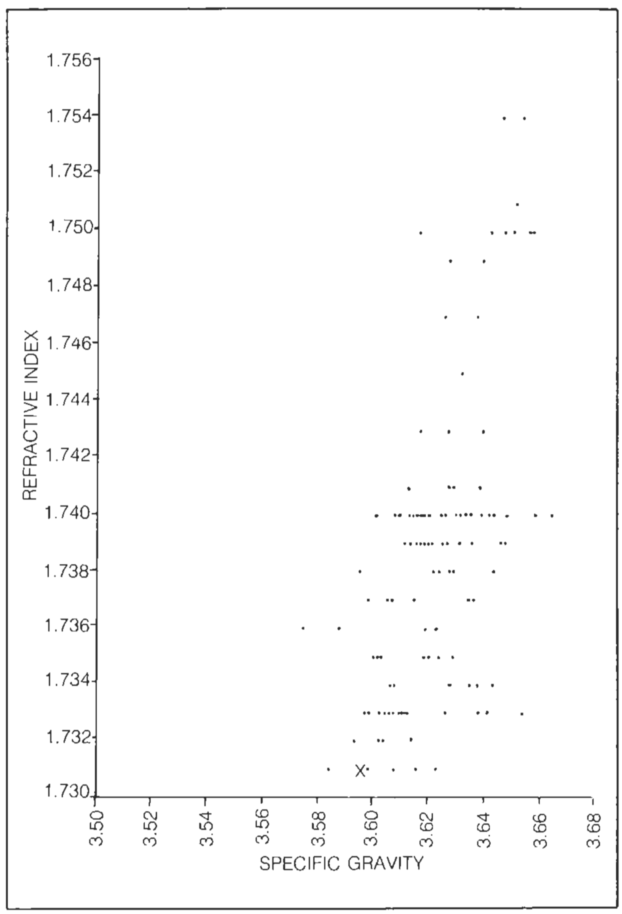
Absorption Spectra. We observed two basic shapes of spectral curves for grossular garnets on the recording spectrophotometer. Green stones exhibited two broad regions of absorption and two of transmission (figure 3). Absorption occurs from the short wavelength end of the visible region to about 460 nm and again in a broad region centered around approximately 603.5 nm and varying in width. However, these broad bands or regions of absorption can be discerned with the hand spectroscope only rarely, in some dark green stones. Transmission is centered around 525 to 550 nm in the green region, as might be expected, and in the far red around 700 to 735 nm. This is virtually identical to the spectra observed by Gübelin and Weibel (1975) and by Amthauer (1975) for similar grossulars.

The second type of spectral curve, associated with colorless, yellow, and orange stones, shows gradually increasing transmission from the short wavelength region to maximum transmission in

TABLE 1. Refractive index and specific gravity ranges for grossular garnets.

Reference	Refractive index	Specific gravity
Shipley (1974)	1.74 -1.75	3.57–3.73
Webster (1975)	1.742–1.748	approx. 3.65
Liddicoat (1981)	approx. 1.735	approx. 3.61
Manson and Stockton (present study)	1.731–1.754	3.57–3.67

Figure 2. Refractive index plotted against specific gravity for the 105 grossulars studied. The coordinate point for the ideal end member grossular is also shown (X).



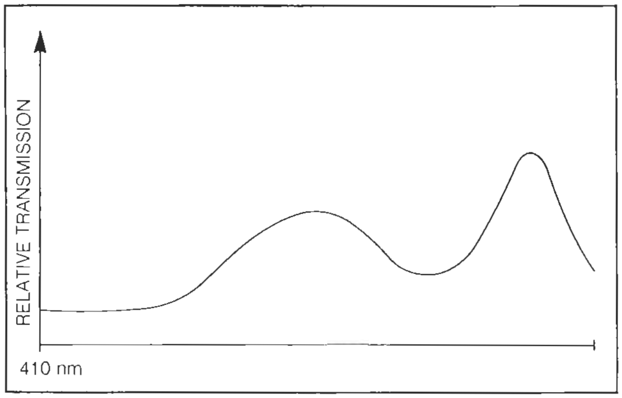


Figure 3. Representative spectral curve of a green grossular as observed with the spectrophotometer.

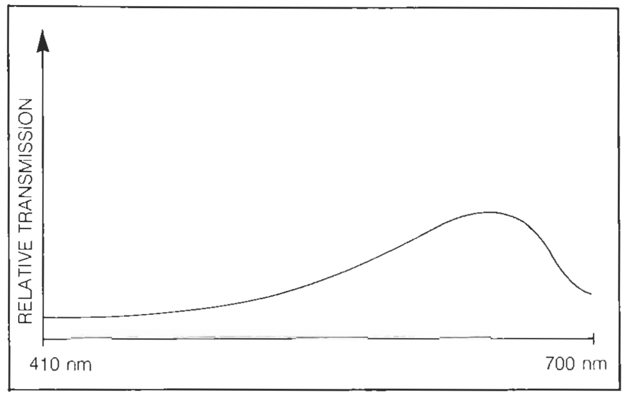


Figure 4. Representative spectral curve of a yellow-orange grossular as observed with the spectrophotometer.

the yellow-to-orange region, depending on the color of the stone, followed by a rapid decrease in transmission to the end of the long wavelength region of the visible spectrum (figure 4). Most of the stones examined showed no sharp absorption bands, but a few showed a faint band around 434 nm that is probably due to Fe^{3+} (Amthauer, 1975; Slack and Chrenko, 1971). One light orange stone also had very faint bands at 418.5, 489.5, 503.5, and 529.5 nm that to our knowledge have not been observed elsewhere in grossular. This stone, however, is the most heavily included one in our collection, and these absorption bands may be related to the inclusions that pervade the specimen. In any case, none of these bands is strong enough to be seen with the hand spectroscope, so they are purely of academic interest to most jeweler-gemologists.

Chemistry. It has long been considered that grossular is chemically continuous with andradite and uvarovite and forms a subgroup with them (Winchell and Winchell, 1951). While the amount of andradite in the 105 gems we analyzed ranged from approximately 0 to 19%, the most uvarovite we encountered was 1.5%. In fact, no single end-member component other than andradite exceeded 7% in any of the gem grossulars we analyzed (table 2). The relative quantities of grossular and andradite compared to all the remaining components can be observed in the ternary diagram of these three divisions (figure 5).

Although examples of stones containing less than 75% and more than 20% grossular have been cited (e.g., Deer et al., 1963), we personally have observed no gem-quality stones in that range. Grossular appears to be chemically less inclined to mix with other end-member components to produce large single crystals of gem quality, so grossulars of gem quality are generally very high in grossular content. The net result is that, chemically, grossular is easy to isolate and identify as a gem species.

Color and Chemistry. CIE color coordinates for the 105 grossulars were derived from ColorMaster notations (Manson and Stockton, 1981) and plotted in a chromaticity diagram (figure 6). The stones themselves were then placed on their respective coordinate points and photographed to show the complete range of colors for these gem grossulars (figure 7). This also illustrates, coincidentally, the quality of color description we have been able to obtain using the ColorMaster.

Pure grossular is colorless. Departing from this pure (and rarely seen) grossular are two distinct color trends that are evident on the chromaticity diagram (again, see figure 6). One of these proceeds through yellow-green to a pure, vivid green at one extreme, and the other proceeds through

TABLE 2. Weight percentage ranges of garnet end members calculated for the 105 grossulars studied.

Formula	End member	Weight %
$Ca_3Ti_2^{4+}(Fe^{3+},Si)_3O_{12}$	Schorlomite	0.02 - 2.14
$Ca_3Fe_2Si_3O_{12}$	Andradite	0 - 19.07
$Mn_3V_2Si_3O_{12}$		0 - 3.05
$Ca_3V_2Si_3O_{12}$	Goldmanite	0 - 6.91
$Ca_3Cr_2Si_3O_{12}$	Uvarovite	0 - 1.51
$Mg_3Cr_2Si_3O_{12}$	Knorringite	0
$Mg_3Al_2Si_3O_{12}$	Pyrope	0 - 2.19
$Mn_3Al_2Si_3O_{12}$	Spessartine	0 - 3.15
$Ca_3Al_2Si_3O_{12}$	Grossular	77.71 - 97.91
$Fe_3Al_2Si_3O_{12}$	Almandine	0 - 3.35

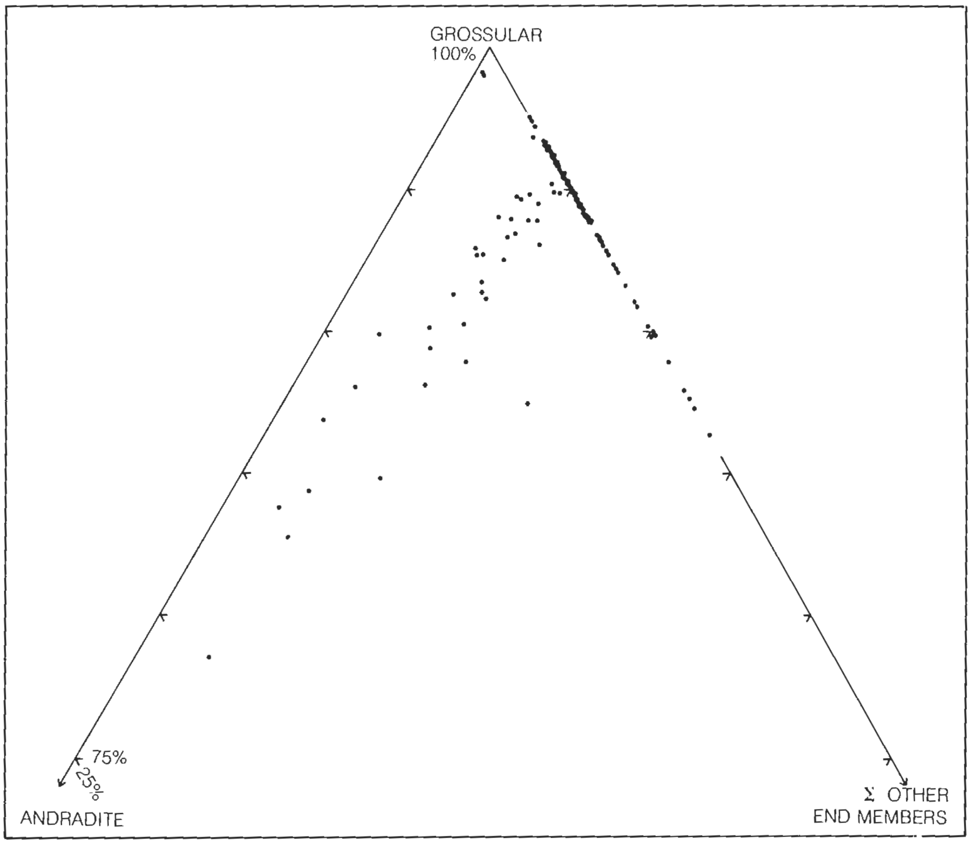
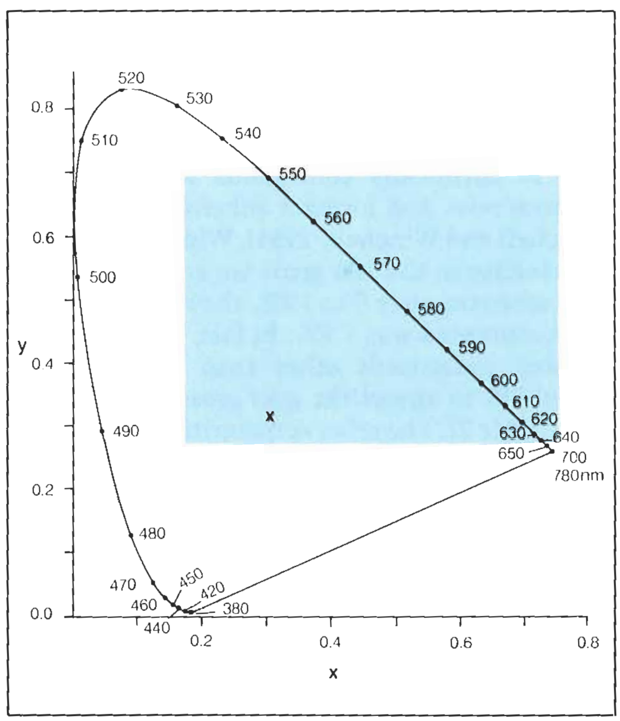


Figure 5. Points for the 105 grossulars plotted on a ternary diagram that displays the relationship between grossular, andradite, and the sum of the remaining end-member components. Grossular is clearly the dominant component among these garnets.

yellow and orange to a vivid red-orange at the other extreme. As the hue changes toward one of the extremes, the saturation increases. Each trend can be roughly represented by a curved line (figure 8). The change in color, corresponding to the changes in both hue and saturation, can then be measured as a percentage of the distance along the respective trend line.

To determine the relationships among the nine oxides present in these garnets, we used the statistical method of factor analysis as described earlier by Manson and Stockton (1981). This analysis of the oxide components for all 105 grossulars revealed the three important clusterings presented in table 3. (We used the original oxide figures from our microprobe analyses in order to avoid any bias that might have been introduced in our calculation of end members.) Factor 1 interprets as the basic grossular component, which is almost invariably accompanied by small amounts of pyrope (less than 2.20%), spessartine (less than 3.15%), and schorlomite (less than 2.15%). Factor 2 expresses the correlation between vanadium (V_2O_3) and chromium (Cr_2O_3) end-member components, and factor 3 is essentially an iron component that appears principally as Fe_2O_3, or

Figure 6. The CIE chromaticity diagram with an indication of the region reproduced in figure 7. X indicates the coordinates for colorless or neutral grey.



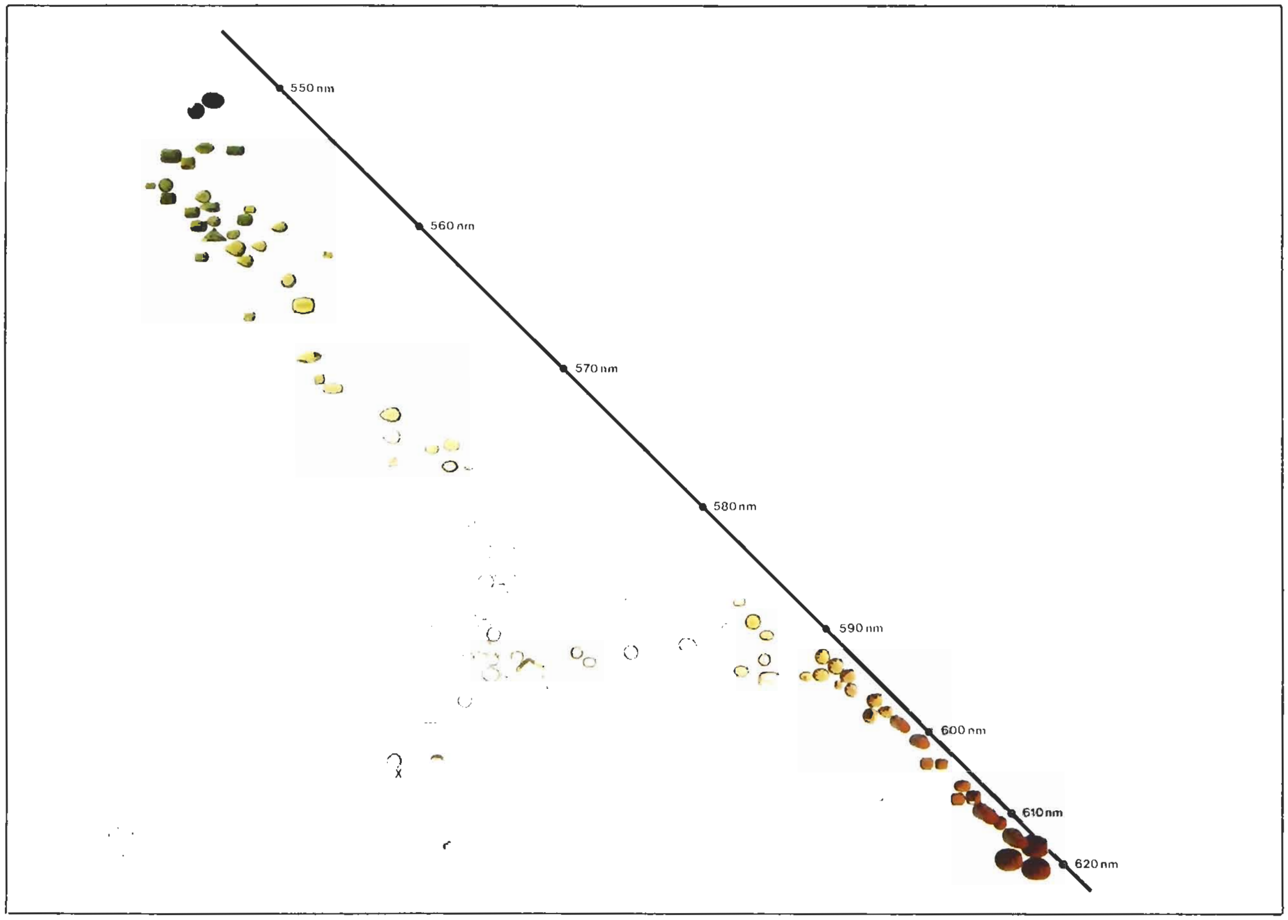


Figure 7. The green-through-orange region of the CIE chromaticity diagram with the 105 garnets positioned according to their x-y coordinates.

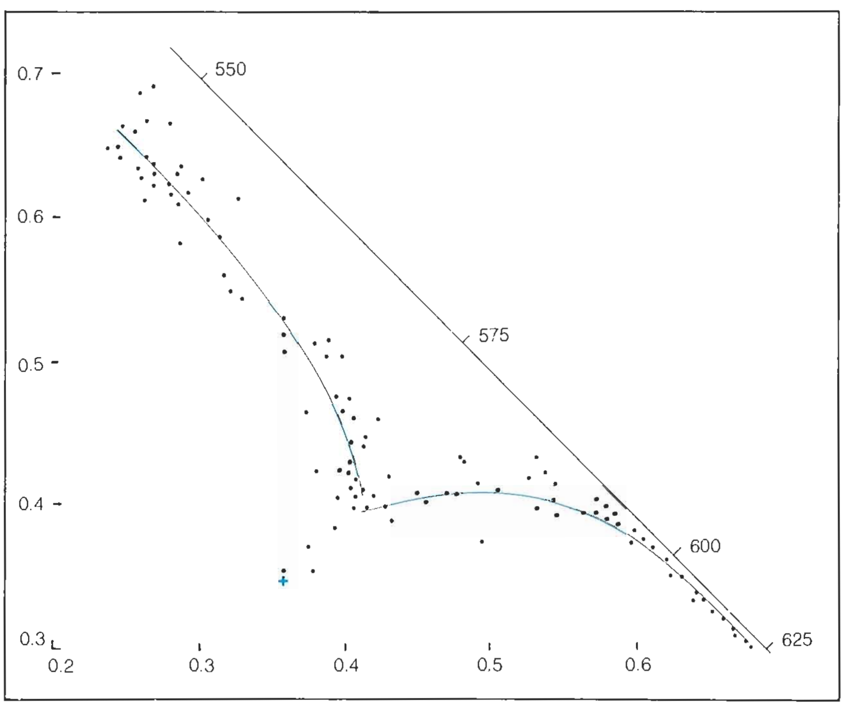


Figure 8. The green-through-orange region of the CIE chromaticity diagram with the color trend lines used to approximate changes in color of the 105 grossulars.

andradite. Factors 2 and 3 become especially significant when we compare the oxides that they represent with the two color trends in grossulars.

Specifically, the measure of color change described above can be plotted against the quantities of the color-related oxides as identified by our factor analysis. Figure 9 shows that there is a direct relationship between the amount of V_2O_3 in a stone and increasing green, but there is no correlation between V_2O_3 and yellow and orange grossulars. Cr_2O_3 (figure 10) behaves remarkably like V_2O_3, although in smaller quantities (only two stones with more than 30% green contained no chromium). This suggests that generally the V_2O_3:Cr_2O_3 ratio in green grossulars is considerably greater than 1:1 but that both vanadium and chromium are usually responsible for the green in grossular garnets. A similar relationship between V^{3+} and Cr^{3+} has been observed by Switzer (1974) and by Amthauer (1975). An exception to this relationship between vanadium, chromium, and green coloration apparently exists in grossulars that are colored principally or solely by Cr_2O_3 (Amthauer, 1976; Wight and Grice, 1982). We have not yet had the opportunity to examine any such material and so have no idea how the green of these stones compares to that of the gems in our study.

By examining how the quantities of FeO and Fe_2O_3 calculated by our end-member program behave with respect to changes in color, we can in-

TABLE 3. Factor analysis of the oxides present in the 105 garnets studied.

Oxide	Factors 1	2	3
SiO_2	+++		+
TiO_2	+++	−	−
Al_2O_3	+++		
V_2O_3		+++	
Cr_2O_3		+++	
MgO	+++	+	−−
CaO	+++		+
MnO	++	++	
FeO			+++

+++ *strong positive correlation*
++ *moderate positive correlation*
+ *weak positive correlation*
−− *moderate negative correlation*
− *weak negative correlation*

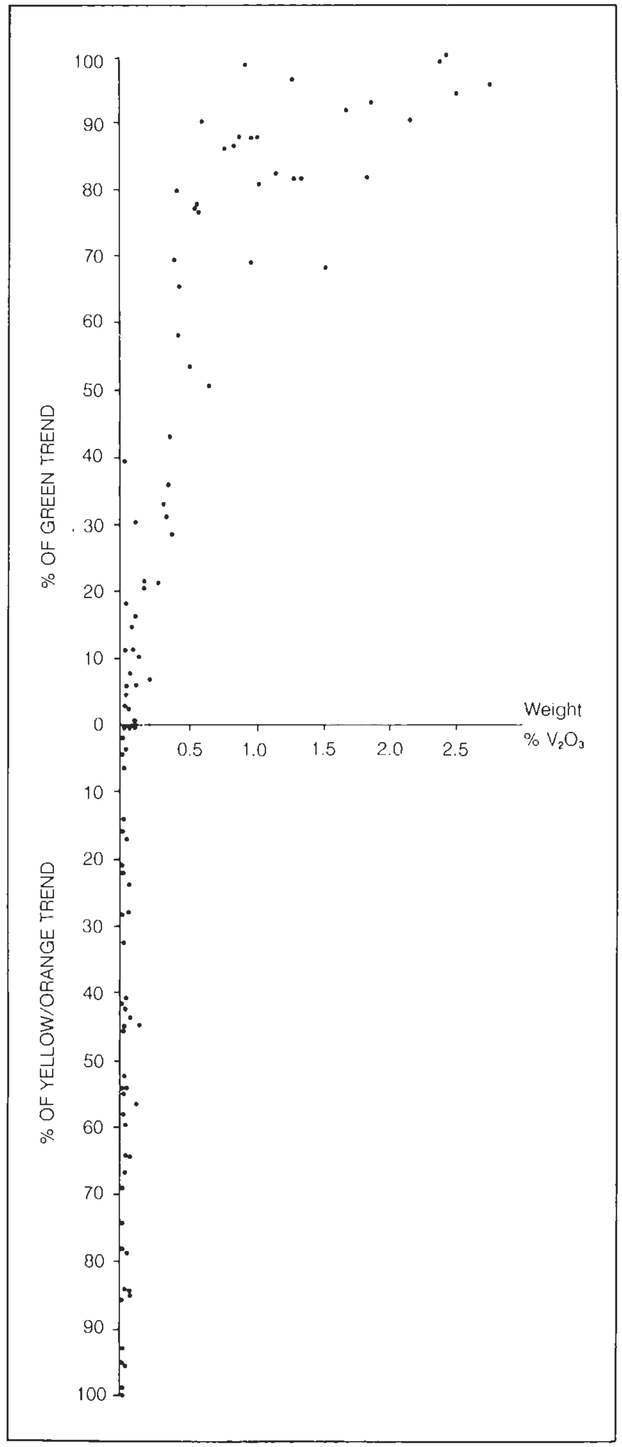


Figure 9. A comparison of the weight percentages of V_2O_3 in the 105 garnets with their respective positions along the color trend lines illustrated in figure 8. V_2O_3 shows a definite increase with respect to increasing green trend.

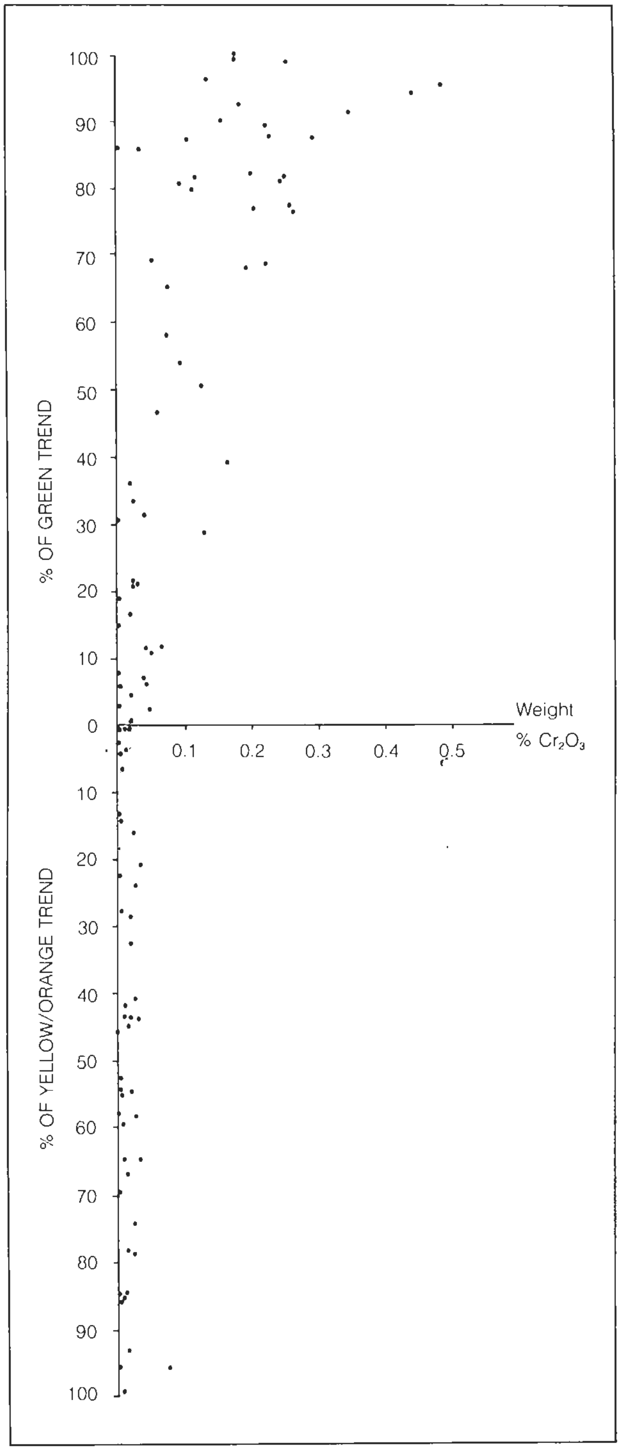


Figure 10. Weight percentage of Cr_2O_3 compared with changes along the color trend lines. Cr_2O_3 increases with the green trend here in much the same way as V_2O_3 does in figure 8. (Caution must be taken to consider the different scales along the x-axis in figures 9–12.)

fer in which valence state (Fe^{2+} or Fe^{3+})* iron is affecting color in grossulars. Figure 11 shows that the amount of Fe_2O_3 clearly increases with the change of color through yellow and orange to red-orange and decreases as green increases. While FeO is conspicuously absent from the green grossulars (figure 12), its role in the color of yellow and orange grossulars remains ambiguous. In order to resolve this question, determination of the valence states of iron in yellow and orange grossulars by means more accurate than stoichiometric calculation is required. In fact, numerous such studies have been performed on the roles of Fe^{2+} and Fe^{3+} in grossular (e.g., Amthauer, 1975; Manning, 1972 and 1973). These have supported the relationship we found between yellow and orange grossular and Fe^{3+}. A study by Manning and Tricker (1977) confirmed the role of Fe^{3+} in this context.

CONCLUSIONS

The gem species grossular presents few problems in description or identification. All of the gems we examined in this study contain more than 70% of the component $Ca_3Al_2Si_3O_{12}$, considerably more than the 50% required to assign them to a gem species. Mineralogical evidence based on non-gem-quality material has shown that there is indeed a continuous chemical series between grossular, andradite, and uvarovite, but the grossulars examined in this study suggest that there is a gap in this series with regard to gem-quality grossular garnets. Until we receive evidence to the contrary, then, we can regard gem grossulars as discrete with respect to any other gem garnet species. Gem grossulars have been observed in this study to possess refractive indices from 1.731 to 1.754 and specific gravities of 3.57 to 3.67. The hand spectroscope revealed no characteristic absorption bands for the 105 grossulars examined, in support of past observations.

Considering the effect that the application of varietal names has on the appreciation of gemstones, the precise definition of gem varieties is of some importance. The two gem varieties of grossular, hessonite and tsavorite, lack such def-

** Fe^{2+} is the valence state of iron in FeO; Fe^{3+} is the valence state of iron in Fe_2O_3.*

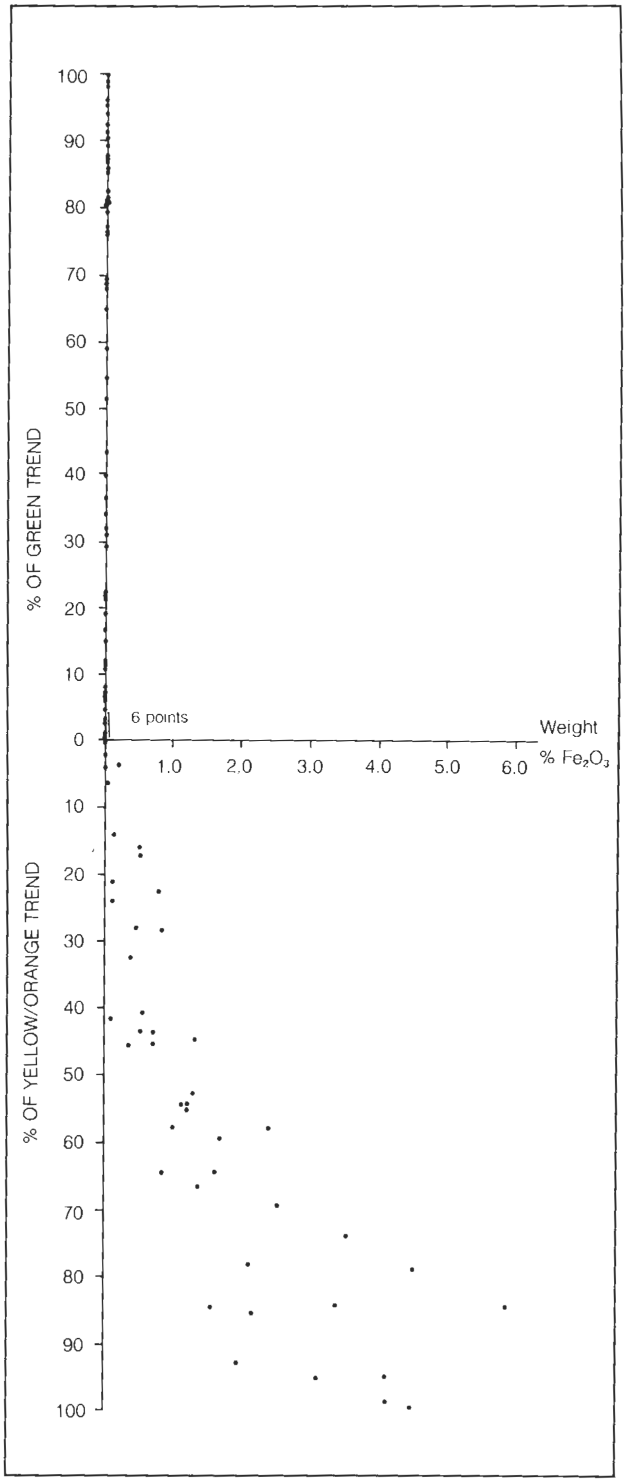


Figure 11. Weight percentage of Fe_2O_3 as determined by the end-member calculations compared to changes along the color trend lines for the 105 grossulars. Fe_2O_3 increases with the increase along the yellow/orange trend line.

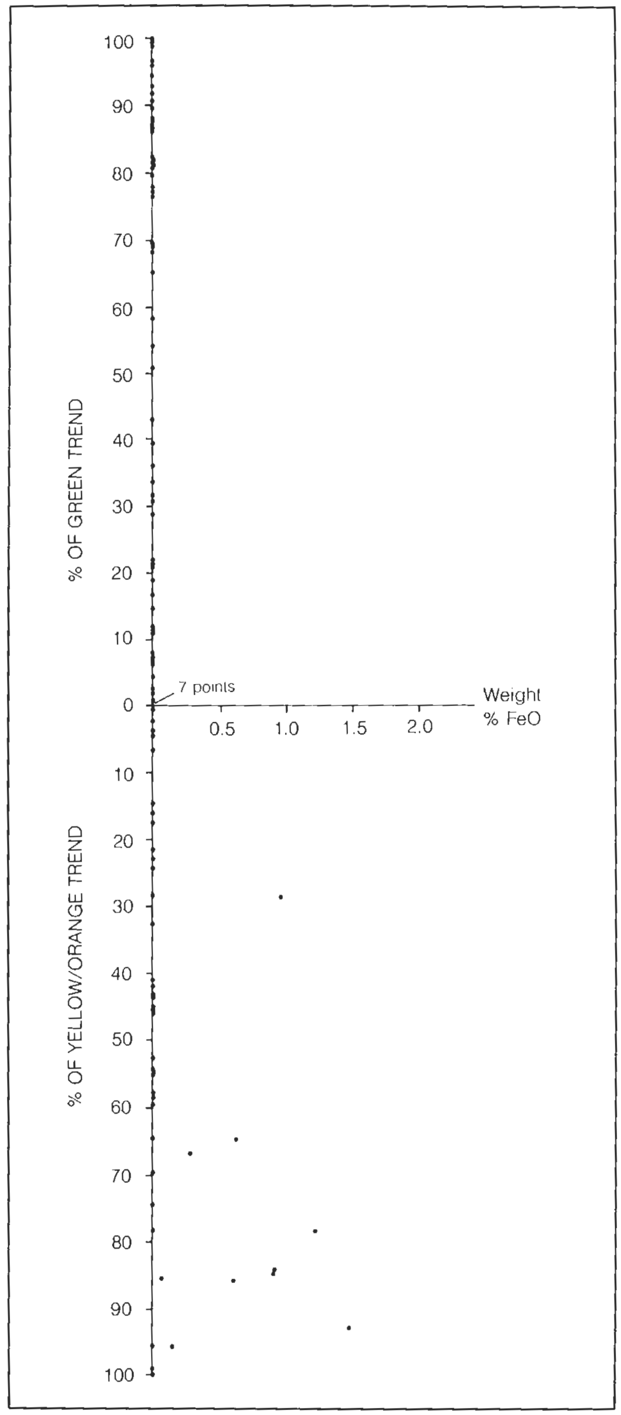


Figure 12. Weight percentage of FeO as determined by the end-member calculations compared with changes along the color trend lines. While FeO is present only in some stones located along the yellow/orange color trend line, no clear-cut correlation can be observed between changing yellow/orange color and the amount of FeO present.

inition, as do most varieties in gemology. In the case of grossular, we have been able to observe a correlation between color and certain chemical elements that strongly supports the use of varietal distinctions. However, as with red and violet garnets, including rhodolite, the causes of color are not always so readily determined. Before we can define any varieties of garnets, therefore, we must review all the variables related to the varietal classification of gem garnets in order to formulate general rules that can be applied uniformly across the group. The concluding paper of the garnet project will include our recommendations for the precise definition of the gem varieties hessonite and tsavorite.

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