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REFRACTIVE INDICES VERSUS ALKALI CONTENTS IN BERYL: GENERAL LIMITATIONS AND APPLICATIONS TO SOME PEGMATITIC TYPES*

P. ČERNÝ AND F. C. HAWTHORNE
Department of Earth Sciences, University of Manitoba, Winnipeg, Manitoba

ABSTRACT

Refractive indices of beryl may be influenced by replacement of (a) Si^{4+} by P^{5+} and B^{3+} in tetrahedra; (b) Al^{3+} by Fe^{3+}, Cr^{3+}, V^{3+}, Sc^{3+}, and Ti in octahedra; (c) Al^{3+} by Mg^{2+}, Mn^{2+}, Ni^{2+}, and Fe^{2+} in octahedra, balanced by alkali metals in channels; (d) Be^{2+} by Li^{+} in tetrahedra, balanced again by alkali metals and large alkaline earths in channels; the indices are also influenced by (e) molecular H_2O entering channels. Furthermore, data used for constructing a determinative curve may be affected by chemical analytical error, different optical methods, presence of submicroscopic channels, and by compositional zoning in beryl crystals. Derivation of an optical versus compositional relationship that would cover all genetic types of beryl is evidently impossible. A new relationship between n_ω and ΣR_2O+CaO is proposed here for primary (Na,Li)– and (Na,Li,Cs)–enriched pegmatitic beryls, in which the octahedral substitutions are negligible. The graph may serve for rapid but approximate estimates of alkali content which exhibits a distinct geochemical trend in this type of beryl.

SOMMAIRE

Les indices de réfraction du béryl subissent l'influence de divers remplacements: (a) Si^{4+} par P^{5+} et B^{3+} dans les tétraèdres; (b) Al^{3+} par Fe^{3+}, Cr^{3+}, V^{3+}, Sc^{3+} et Ti dans les octaèdres; (c) Al^{3+} par Mg^{2+}, Mn^{2+}, Ni^{2+} et Fe^{2+}, aussi dans les octaèdres, mais avec introduction de métaux alcalins dans les tunnels; (d) Be^{2+} par Li^{+} dans les tétraèdres, et métaux alcalins et alcalins-terreus dans les tunnels; les indices sont aussi influencés par (e) l'eau moléculaire à l'intérieur des tunnels. De plus, les données nécessaires à la construction d'une courbe déterminative peuvent être affectées par les erreurs de l'analyse chimique, les différentes méthodes optiques, la présence de tunnels submicroscopiques et de zones de composition variable dans les cristaux de béryl. Il est évidemment impossible d'établir, entre les constantes optiques et la composition, une relation unique valable pour tous les types génétiques. On propose ici une courbe nouvelle, donnant n_ω en fonction de (ΣR_2O+CaO) pour les béryls pegmatitiques primaires, enrichis en (Na,Li) ou en (Na,Li,Cs), mais dans lesquels les remplacements de cations octaédriques sont négligeables. Cette courbe permet une estimation approximative, mais rapide, de la teneur en alcalis, dont la variation dans les béryls de ce genre est d'allure nettement géochimique.

(Traduit par la Rédaction)

INTRODUCTION

Reliable correlation of composition with physical properties in a mineral species showing compositional variability is extremely useful for rapid estimates (or indirect determination) of composition; however, only rarely can such a correlation be established easily and accurately. Most mineral series, particularly complex silicates, have a long history of attempts to clarify these relationships. The obstructions to progress range from the seemingly trivial, such as a lack of match between chemically analyzed and physically tested materials, to complex interaction of several independent variables (e.g., several types of ionic substitutions, filling of interstitial space, and variable structural state).

In beryl, the relationships between the different chemical substitutions and unit-cell dimensions were successfully elucidated by Bakakin *et al.* (1970). Since the early work of Ford (1910) and Lacroix (1911), there have been many attempts to develop relationships between chemical composition and refractive indices. Alkali content has long been recognized as the major factor influencing the variation in optical properties; however, the role of other compositional variables has frequently been neglected, possibly giving rise to the diversity of relationships proposed by different authors. For example, Figures 1a and 1b show the relationship developed, respectively, by Winchell (1951) and Deer *et al.* (1962); the regressions lines are strikingly dissimilar and there is considerable scatter in the data. In addition, the formula "$R_3Al_2Si_6O_{18}$" used to denote compositional variation does not conform to the present understanding of ionic substitutions in beryl. Figures 1c and 1d show the relationships proposed by Dorfman (1951) and Feklitchev (1964). Dorfman's data are almost perfectly aligned in a

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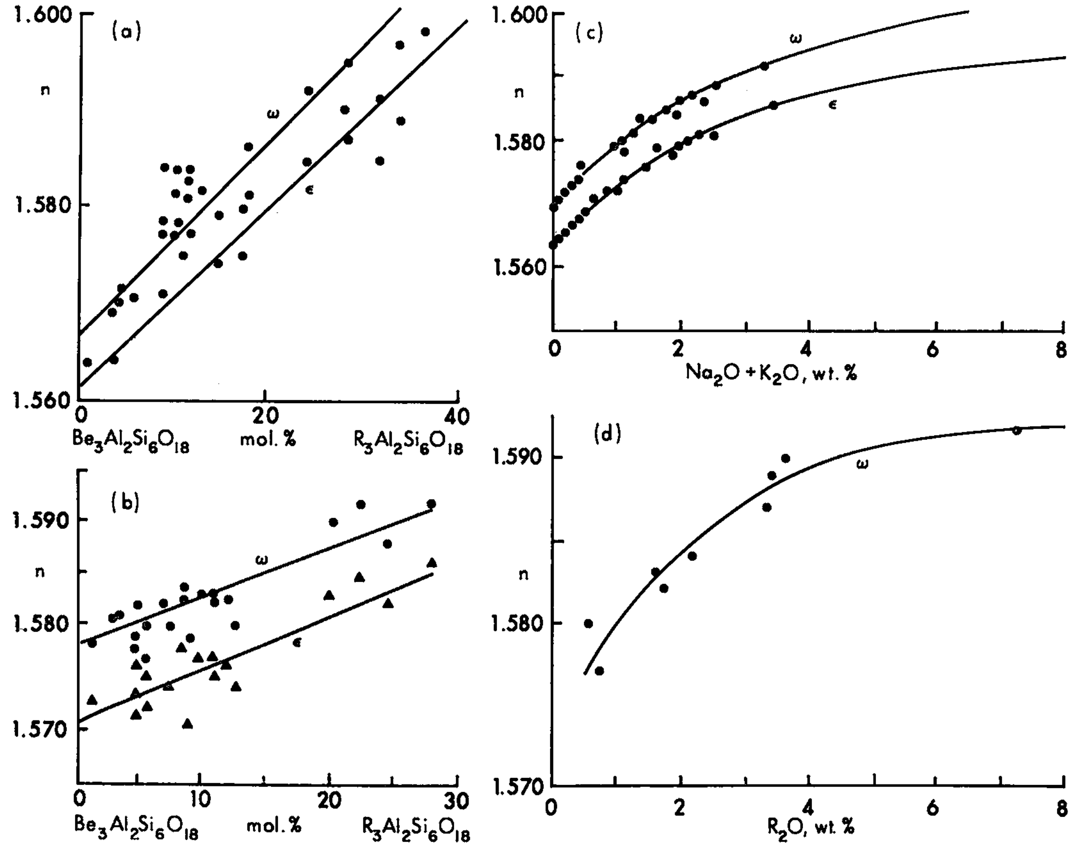

FIG. 1. Correlation of alkali contents with refractive indices of beryl by (a) Winchell (1951), (b) Deer *et al.* (1962), (c) Dorfman (1951), and (d) Feklitchev (1964). Plots of n_ϵ and n_ω are not distinguished in Winchell's (1951) original graph. Note the inconsistencies in the extrapolated values for $Be_3Al_2Si_6O_{18}$, in the slopes of the regression lines, and in the changes of birefringence with increasing "$R_3Al_2Si_6O_{18}$" between (a) and (b).

plot involving only one of the major alkalis (Na) and one of the minor components (K) entering the beryl structure. Feklitchev's graph is based only on 9 data points and hence is, as shown below, of questionable validity. The reasons for these inconsistencies should be examined, as a rapid characterization of beryl compositions is very useful in reconnaissance studies of its various parageneses.

FACTORS INFLUENCING REFRACTIVE INDICES

Based on knowledge of the structure and crystal chemistry of beryl (Gibbs *et al.* 1968; Bakakin *et al.* 1969, 1970; Morosin 1972; Hawthorne & Černý in prep.), the following factors affect the refractive indices:

(a) Tetrahedral substitution of Si^{4+} by P^{5+} or B^{3+}, and possibly minor Al^{3+}. Although this is extremely unusual in any large amount (e.g., Feklitchev & Razina 1967), on a small scale it may be much more common than is generally realized as beryl is only rarely analyzed for phosphorus or boron. The effect of this substitution on optical properties cannot be predicted as it would depend on the type of charge balancing mechanism; as yet, this has not been sufficiently characterized.

(b) Octahedral substitution of Al^{3+} by Fe^{3+}, Cr^{3+}, V^{3+}, Sc^{3+} and Ti. This substitution is common in most genetic types of beryl and may attain considerable levels in some varieties (Feklitchev 1964, Table 6). As the ionic refractivities of all these species are higher than that of Al^{3+} (Feklitchev 1964), this type of substitution will raise the refractive indices of beryl.

(c) Octahedral substitution of Al^{3+} by Mg^{2+}, Mn^{2+}, Ni^{2+} and Fe^{2+}. As with (b), this substitution is also very common in many types of beryl. This substitution produces in the structure a charge imbalance that is generally compensated by monovalent cations entering the channel sites. As shown by the data of Schaller *et al.* (1962), the increase in refractive indices is much steeper than for substitutions of type (b) because of the additional effect of the alkalis.

(d) Tetrahedral substitution of Be^{2+} by Li^+, coupled with alkalis, Ca^{2+}, Ba^{2+} and Sr^{2+} in the channels to balance the charge deficiency produced by the substitution. Although K^+,Rb^+,Li^+ and the alkaline-earth elements are generally minor components in the channels, both Na^+ and Cs^+ attain considerable levels.

Alkali enrichment in pegmatitic beryl shows a distinct geochemical trend (Černý 1975) but there is no apparent crystallochemical constraint that would make possible an accurate prediction of alkali ratios. In alkali-poor pegmatitic beryl and in beryl from other parageneses, the alkali contents are known to be rather variable (Beus 1960; Feklitchev 1964; Černý 1975). In addition, the channel alkalis are not exchangeable and thus the more complex beryls of different origin cannot be transformed into simple, e.g., Li^++Na^+–substituted phases, to simplify mutual comparison.

Most authors do not take Ca^{2+} into consideration when plotting alkali contents vs. refractive indices. However, Ca must occupy the channel sites in the beryl structure, as clearly shown by the formulae of those rare beryl varieties that contain substantial Ca (Simpson 1948; Feklitchev 1964; Yurgenson 1965). The ionic refractivity of Ca indicates that CaO should increase the refractive indices at about triple the rate of Na_2O (Feklitchev 1964).

(e) Molecular water in channels. As pointed out by Hall & Walsh (1971), its importance has not been recognized by most authors, although it has a prominent effect. The close structural analog, Mg-cordierite, shows n_m increases by about 0.009 for each wt. % of channel H_2O (Schreyer & Yoder 1964). Hydrogen bonding of Na^+ to the channel walls requires at least 1 H_2O, ideally $2H_2O$, per Na^+ (Hawthorne & Černý in prep.). Figure 2a shows that most beryls contain water in excess of the ideal configuration; the similarity in shape of the Na^+ and H_2O fields in Figure 2b suggests that this excess is truly random, and is not influenced by other alkali species. Thus, water represents an unpredictable quantity which again cannot be separated from other factors. In the generally alkali-poor and Cs-free cordierite, channel water can be driven out easily by heating, and refractive indices can be compared for anhydrous material of about equivalent structural state (as standardized by Iiyama 1960). However, the channels of the beryl structure are effectively plugged by alkalis, particularly Cs, and dehydration proceeds very sluggishly even in pulverized material at high temperatures (Ginzburg 1955; Matsuda 1963).

The correlation of all the ionic substitutions outlined above with refractive indices is also plagued by experimental problems. Besides the omnipresent influence of chemical analytical error and variable experimental conditions and

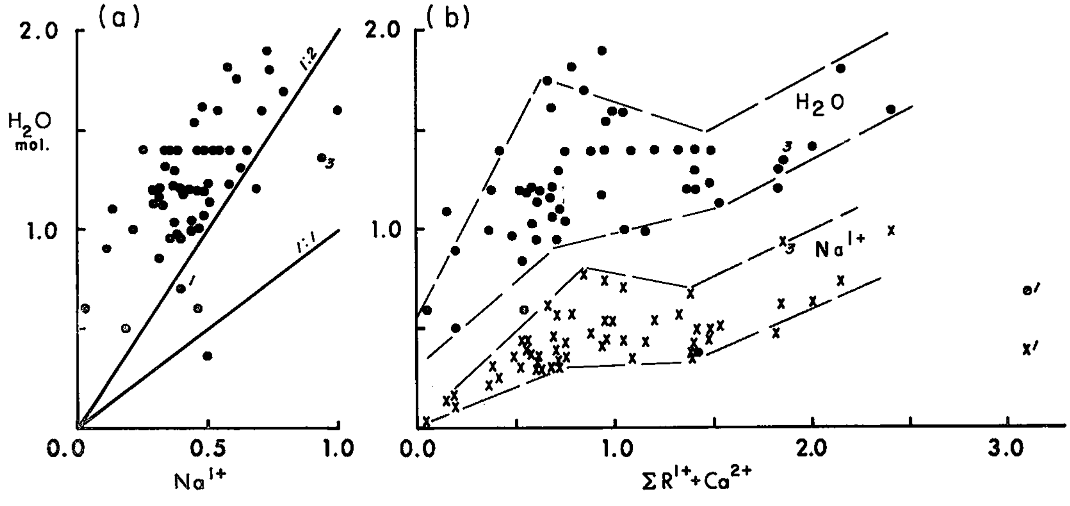

FIG. 2. H_2O content of beryl (mol. %) compared to the Na^+ content (a) and to the ΣR^++Ca content (b). Data from Sosedko (1957) 3 samples with the richest in alkalis marked 3; Deer *et al.* (1962), 8 samples; Vorma *et al.* (1965), 9 samples; Evans & Mrose (1966), 1 sample marked 1; Gordiyenko (1970), 5 samples; Bakakin *et al.* (1970), 22 samples; Hall & Walsh (1971), 6 samples; and Černý & Simpson (in prep.), 3 samples. Note that in (a) most beryls contain H_2O in excess of the 1:2 ratio, and that the general shape of the main H_2O field (dots) in (b) corresponds to that of Na^+ (X).

accuracy of determining the optical constants, two other points deserve mention:

(f) Submicroscopic channels parallel to *c*, of average thickness of about 20 μm, may be filled with gaseous or liquid substances, and may contain solid particles. Baier & Pense (1962) report that the channels are common in different varieties of beryl. If abundant, they may affect the relationship between composition and optical properties.

(g) Compositional zoning, a feature very common in beryl crystals, occasionally involves rather steep gradients (Hurlbut & Wenden 1951; Heinrich 1952; Jahns 1953; Beus 1960). Determination of optical properties on fragments representative of the bulk of chemically analyzed material is often difficult to achieve (Černý & Simpson in prep.), and yet is rarely discussed.

LIMITATIONS AND POSSIBILITIES

The array of mutally independent variables (with no chance of experimentally eliminating any) may easily convince us that any attempt to correlate alkali contents of beryl with its refractive indices is impossible. This would be the case if we tried to include all kinds of beryl. However, consideration of individual genetic types of beryl may simplify the problem, since in specific cogenetic groups some ionic substitutions are often negligible. This approach was used by Pavlova (1963) who distinguished nine optical types having different paragenetic affiliations in vein-type beryllium occurrences; however, pegmatitic beryl was not included in her study. As pegmatitic beryls are of our immediate interest, and because alkali-rich pegmatitic beryls are characterized by the near-absence of octahedral substitutions, we restrict the following discussion to this particular group, the (Na,Li)– and (Na,Li,Cs)–enriched beryls from Li-bearing and Li,Rb,Cs-rich pegmatites.

No attempt was made to compile all available analyses for Figure 3, but the sampling from recent literature gives a fair representation of this genetic type. The sample is large enough to

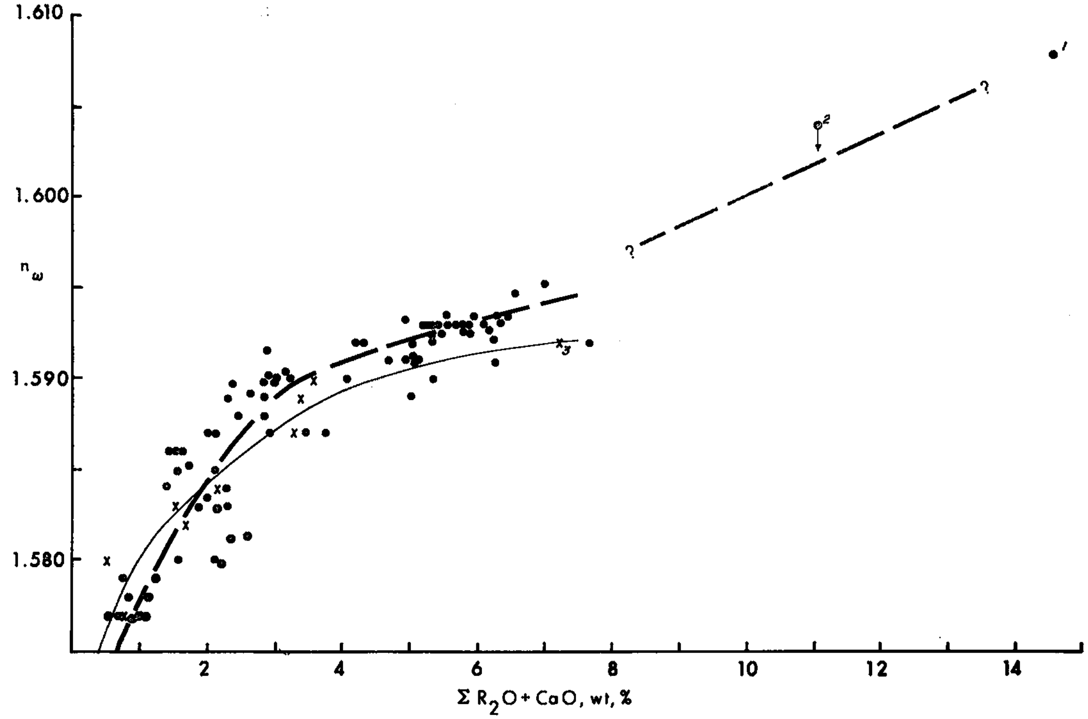

FIG. 3. Plot of n_ω vs. ΣR_2O+CaO (wt. %) in (Na,Li)– and (Na,Li,Cs)–rich pegmatitic beryls. Specimens marked by X and the thin line of best fit are those of Feklitchev (1964, Fig. 12; CaO is not included by him). Data from Quensel (1937), 2 samples; Sosedko (1957), 3 samples, his sample richest in alkalis as plotted by Feklitchev marked 3; Deer *et al.* (1962), 2 samples; Vorma *et al.* (1965), 9 samples; Evans & Mrose (1966), 1 sample marked 1; Gordiyenko (1970), 9 samples; Černý (1972), 1 sample marked 2; Černý & Turnock (1975), 18 samples; and Černý & Simpson (in prep.), 41 samples. Note the differences between our and Feklitchev's (1964) trends, the bend in our trend line at about 3 wt. % ΣR_2O+CaO, and the deviation of samples 1 and 2 from this trend.

eliminate the influence of distinctly anomalous and/or erroneous data.

The optical data used were limited to n_ω since it is the most practical to determine, and birefringence (known to increase with alkali content) changes at a very slow rate.

In the bulk of the data plotted in Figure 3, the influence of factors (a) — tetrahedral substitution, and (f) — submicroscopic channels, is probably negligible. Octahedral substitutions (b) and (c) are almost nonexistent in the alkali-rich specimens, and an arbitrary limit of 0.5 wt. % of substituting R_2O_3 was used in selecting analyses for the alkali-poor part of the graph. Thus Figure 3 illustrates mainly the effects of the alkali enrichment due to mechanism (d), with a scatter introduced by partly covariant H_2O (e) and erratic errors introduced by zoning (g). Figure 3 also contains Feklitchev's (1964) curve based on nine beryl analyses, with Li_2O, Na_2O, K_2O, and Cs_2O taken as ΣR_2O.

DISCUSSION

Figure 3 shows an alignment of data in a trend that is expected from the general shape of Feklitchev's (1964) curve. However, the scatter of data is much larger than that of his selected analyses, and our visually estimated trend line is distinctly steeper at high alkali contents. Feklitchev used Sosedko's (1957) most alkali-enriched beryl with $\Sigma R_2O=7.23$ wt. % as the only anchoring point above ΣR_2O of 3.5 wt. %. Figure 3 shows this point on the low-n_ω side of the data set, suggesting that the analyzed material was richer in alkalis than that examined optically. Also, this beryl contained less than the usual (and crystallochemically preferred) amount of H_2O (cf. Fig. 2a).

The conspicuous change in the slope of the trend line at about 3 wt. % ΣR_2O+CaO is caused by a prominent shift in alkali ratios. As shown by Černý (1975, Figs. 1 to 4), the initial increase in total alkalis in this type of beryl is due mainly to Na and Li, with very slow increase in Cs. At about 3 wt. % ΣR_2O+CaO (and approximately 0.5 wt. % Cs_2O) cesium starts increasing at a much greater rate, whereas sodium and H_2O remain at about the same level (Fig. 2b). This change should cause a slower increase in n_ω per unit of wt. % ΣR_2O+CaO because of the high molecular weight of Cs_2O.

Thus the compositional vs. optical relationship of Figure 3 reflects a geochemical trend in evolution of beryl from Li-bearing and Li,Rb, Cs-rich pegmatites, which involves a prominent change in the alkali substitution with increasing total alkalis. Initially, $Be^{2+}/Li^{+}+Na^{+}+2H_2O$ predominates; at higher alkali levels, $Be^{2+}/Li^{+}+Cs^{+}$ takes over almost exclusively. Because of these two mainly consecutive but somewhat overlapping mechanisms, no attempt was made at mathematical expression of their combined effects.

A quantitative confirmation of the above interpretation was obtained by calculating n_m [$=(n_\alpha+n_\beta+n_\gamma)/3$] for selected beryl compositions listed in Table 1. They include ideal $Be_3Al_2Si_6O_{18}$ (A), two compositions of alkali-poor NaLi-beryls (B and C, very close to beryls described by Černý & Turnock 1975), a composition with high Na+Li but low Cs (D, similar to beryls studied by Vorma *et al.* 1965 and Gordiyenko 1970), and finally a close approximation to the most cesian primary Tanco beryls (E, Černý & Simpson in prep.). The increase in total alkalis from A to B and C is due only to the substitution $Be^{2+}/Li^{+}+Na^{+}+2H_2O$, and that from D to E is controlled only by $Be^{2+}/Li^{+}+Cs^{+}$; the step C-D is achieved by a combined effect of both mechanisms.

TABLE 1. HYPOTHETICAL BERYL COMPOSITIONS AND CALCULATED PHYSICAL PROPERTIES

	A	B	C	D	E	F	G
SiO_2	67.04	66.34	65.64	64.59	62.75	65.52	65.43
Al_2O_3	19.00	18.80	18.60	18.30	17.78	18.24	18.26
Fe_2O_3	-	-	-	-	-	.51	-
FeO	-	-	-	-	-	-	.40
BeO	13.96	13.35	12.76	12.00	11.11	12.74	12.80
Li_2O	-	.28	.55	.87	1.18	.55	.50
Na_2O	-	.57	1.13	1.67	1.62	1.13	1.21
Cs_2O	-	-	-	.63	3.68	-	-
H_2O+	-	.66	1.32	1.94	1.88	1.31	1.40
	100.00	100.00	100.00	100.00	100.00	100.00	100.00
ΣR_2O	-	.85	1.68	3.17	6.48	1.68	1.71
Si	12.00	12.00	12.00	12.00	12.00	12.00	12.00
Al	4.00	4.00	4.00	4.00	4.00	3.93	3.94
Fe^{3+}	-	-	-	-	-	.07	-
Fe^{2+}	-	-	-	-	-	-	.06
Be	6.00	5.80	5.60	5.35	5.10	5.60	5.63
Li	-	.20	.40	.65	.90	.40	.37
Na	-	.20	.40	.60	.60	.40	.43
Cs	-	-	-	.05	.30	-	-
H_2O	-	.40	.80	1.20	1.20	.80	.86
ΣR^{1+}	-	.40	.80	1.30	1.80	.80	.80
$\underline{a}$ (Å)	9.210	9.210	9.210	9.210	9.210	9.210	9.210
$\underline{c}$ (Å)	9.190	9.200	9.210	9.222_5	9.235	9.210	9.210
V (Å³)	675.1	675.8	676.5	677.5	678.4	676.5	676.5
density	2.641	2.666	2.692	2.732	2.808	2.697	2.700
n_m	1.561_6	1.569_3	1.577_2	1.586_6	1.595_4	1.579_5	1.578_9

Unit-cell dimensions of beryl with the above compositions were derived from the graphs of Bakakin *et al.* (1970), and after calculating the density, n_m was obtained using the Gladstone-Dale equation and specific refractive energies

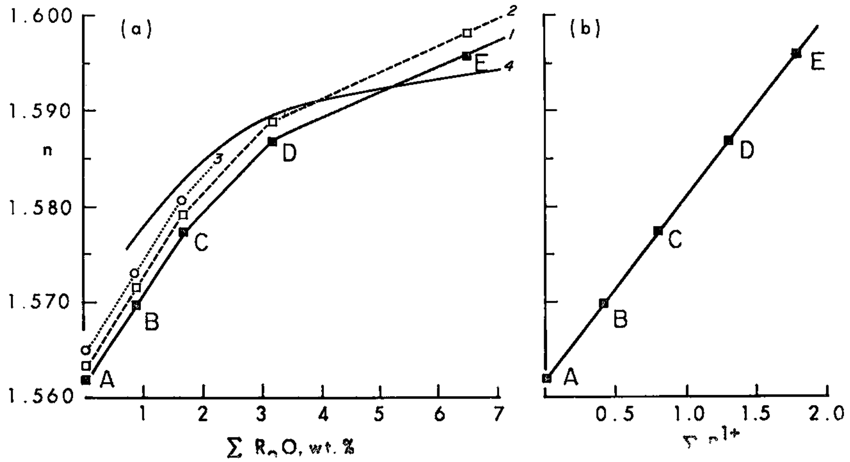

FIG. 4. Calculated n_m of simplified beryl compositions vs. ΣR_2O in wt. % (a) and vs. ΣR^+ in atomic contents (b). In (a), calculated n_m values from Table 1 (line 1, solid squares), adjusted to probable n_ω using $n_m=(2n_\omega+n_\epsilon)/3$ (line 2, empty squares), and corrected for about 0.5 wt. % Fe_2O_3 (line 3, empty circles), are compared to the same relationship in natural beryls (line 4, redrawn from Fig. 3); see text for comments. In (b), calculated n_m shows almost the same rate of increase with Cs^+ (points D to E) as with $Na^+ + 2H_2O$ (points A to C).

quoted by Larsen & Berman (1934), except for Jaffe's (1956) value of 0.290 for Fe_2O_3 in silicates and Mandarino's (1964) value of 0.120 for Cs_2O.

Figure 4a shows that the main change in the slope of our trend from Figure 3 coincides with that of the $n_{m(calc.)}$ line 1. (Fig. 4b demonstrates that the change in slope of n_m plotted against atomic ΣR^+ is not visually perceptible). The calculated values shift even closer to the pegmatitic beryl trend when n_m is recast to n_ω (line 2), using the probable relationship $n_m = (2n_\omega + n_\epsilon)/3$ (Larsen & Berman 1934), and assuming $\epsilon - \omega = 0.004$ and 0.007 for alkali-poor and alkali-rich beryl, respectively. However, the calculated trend is slightly steeper than that defined by the natural specimens.

Calculated values are low in the alkali-poor part of the graph because the corresponding pegmatitic beryls have some of the highest water in excess of the 1 $Na/2H_2O$ ratio (Fig. 2b), and they usually carry some Fe substituting for Al^{3+}. As shown in Table 1, the presence of 0.5 wt. % Fe_2O_3 (Al^{3+}/Fe^{3+} substitution in F) or 0.4 wt. % FeO ($Al^{3+}/Fe^{2+} + Na + 2H_2O$ in G) produces n_m higher by about 0.002 than in the otherwise comparable composition D (line 3 in Fig. 4a).

There is no simple explanation available for the relatively high values of calculated n_m and n_ω in the alkali-rich side of the graph. It is possible that the trend from Figure 3 (line 4 in Fig. 4) is strongly influenced by a large group of beryls from a single locality (Tanco) that are not fully representative of beryls containing 3 to 4 wt. % Cs_2O. However, the few beryls from this pegmatite that were analyzed in full (Černý & Simpson in prep.) do not show any compositional deviations that might support this assumption.

The two samples showing the highest total alkali contents in Figures 2b and 3 are the secondary beryl from Tanco (Černý 1972) and the Madagascar sample of Evans & Mrose (1966). Both samples plot higher than would be indicated by extrapolation of the Cs-rich part of the trend between 4.0 and 7.5 wt. % $\Sigma R_2O +$ CaO (Fig. 3) but lower than required by the calculated trend in Figure 4a. It is not possible to isolate the main factor(s) responsible for this deviation, since the data come from incompletely described specimens. However, both samples represent secondary phases from leaching cavities, genetically different from the primary rock-forming beryls which constitute the bulk of the specimens used in constructing the graph; the Tanco sample contains appreciable K_2O, and the Madagascar specimen has very low Na_2O and H_2O. Also, the optically characterized Tanco sample was probably contaminated by some material from the adjacent alkali-poorer zone of the crystal. All this suggests that the secondary beryls richest in alkalis are chemically not comparable with primary rock-forming beryls, and may possibly show an optical trend of their own.

We may conclude that only rough estimates of alkali contents are possible from Figure 3. The probable error should be between ±0.5 and ±1.0 wt. %. Despite this lack of accuracy, the graph yields useful geochemical information. Beryls with $n_\omega < 1.580$ contain subordinate amounts of Cs, and $n_\omega > 1.590$ is characteristic of Cs-rich varieties from inner zones of pegmatites that contain not only abundant lithium, but also tend to contain pollucite.

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