THE GLADSTONE–DALE RELATIONSHIP. PART III:
SOME GENERAL APPLICATIONS

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ABSTRACT

Although the Gladstone–Dale relationship is most often used to calculate either the mean refractive index or the density, the most useful quantities derived from the relationship are the two values of K. The chemical refractive energy, K_C, is calculated from the weight percentages of the constituents and their specific refractive energy constants: the physical refractive energy, K_P, is calculated from the mean refractive index and the density. The ratio K_P/K_C serves as a measure of the compatibility of the refractive index, density and chemical data. Where $1 - (K_P/K_C)$ is from ± 0.000 to ± 0.019, the compatibility of the data is considered superior; from ± 0.020 to ± 0.039 it is excellent, from ± 0.040 to ± 0.059 it is good, from ± 0.060 to ± 0.079 it is fair, and greater than ± 0.079 it is poor.

SOMMAIRE

Quoique la loi de Gladstone–Dale serve généralement à calculer soit l'indice médian de réfraction soit la densité, ce sont les deux valeurs de K dérivées de cette relation qui sont les plus utiles. L'énergie réfractive chimique K_C d'un composé est la moyenne arithmétique pondérée des énergies réfractives spécifiques de ses composants, et l'énergie réfractive physique K_P égale $(n-1)/D$, où n est l'indice médian de réfraction et D la densité. Le rapport K_P/K_C reflète la compatibilité entre l'indice médian, la densité et la composition. Cette compatibilité est qualifiée de "supérieure" lorsque le rapport s'écarte de l'unité de moins de ± 0.020; elle est "excellente" pour un écart de ± 0.020 à ± 0.039, "bonne" de ± 0.040 à ± 0.059, "passable" de ± 0.060 à ± 0.079, et "piètre" si l'écart excède ± 0.079.

(Traduit par la Rédaction)

INTRODUCTION

In the two previous parts of this series, new specific refractive energy constants were presented (Mandarino 1976) and the relationships among these constants were discussed (Mandarino 1978). This part deals with some practical applications of the Gladstone–Dale relationship.

The most common applications seen in the literature are: (1) the calculation of the mean refractive index (n) from measured density (D) and the specific refractive energy (K) derived from the chemical analysis; (2) the calculation of density from the mean refractive index and the specific refractive energy. The equations used for these calculations are, respectively, $\bar{n} = 1 + KD$ and $D = (\bar{n} - 1)/K$. The results of such calculations should be assessed very carefully before any use is made of them, especially in the first case. For example, a compound with D (meas.) = 2.95 g/cm³ and $K = 0.200$ calculated from the chemical composition would give a calculated $\bar{n}$ of 1.590. A error of ± 0.002 in K would result in an error of ± 0.006 in $\bar{n}$. If, in addition, there was an error of ± 0.02 in the density, the error in the calculated value of $\bar{n}$ could be as high as ± 0.010. Although errors in D of ± 0.02 and in K of ± 0.002 are quite acceptable, an error in $\bar{n}$ of ± 0.010 or even ± 0.006 is high by most standards. On the other hand, in the second application, for a compound in which $\bar{n} = 1.590$ and $K = 0.200$, an error of ± 0.002 in both $\bar{n}$ and K could result in a maximum error in D of ± 0.04, which is not unreasonable for a D of 2.95 g/cm³. In other words, calculation of $\bar{n}$ can result in much larger errors than the calculation of D.

Calculation of $\bar{n}$

A value of $\bar{n}$ calculated from K and D should be considered only as an approximate refractive index; in general, it would be of little use. One possible use is the selection of an appropriate refractive index liquid at the start of an optical study of a new mineral. A recent example at the Royal Ontario Museum will serve to illustrate the approach. The chemical formula of a new mineral had been determined by Mr. Pete J. Dunn of the Smithsonian Institution as $AlCa_2F_7 \cdot H_2O$ with D(meas.) =

2.86 g/cm^3 and D(calc.) = 2.89 g/cm^3. The specific refractive energy calculated from the chemical formula is 0.136. Using K and D(meas.), $\bar{n}$ = 1.389; using K and D(calc.), $\bar{n}$ = 1.393. By using the estimated mean refractive index as 1.39–1.40, my colleague, Mr. B. D. Sturman, saved much time by selecting as his first refractive index liquid one with $\bar{n}$ = 1.400. He quickly found that the refractive indices of the mineral were somewhat higher than 1.400.

Calculation of D

Densities calculated from the Gladstone–Dale equation can be used to check measured densities and those calculated from unit-cell data. Mandarino *et al.* (1976) gave an example of the use of two sets of calculated densities (from the unit-cell data and from the Gladstone–Dale equation) to shed light on the chemical composition of zemannite. More generally, however, a density calculated from the Gladstone–Dale equation probably will be of greatest use in detecting gross errors.

SPECIFIC REFRACTIVE ENERGY

In the author's opinion, the most useful quantity calculated from the Gladstone–Dale relationship is the specific refractive energy K. Because values of K may be calculated from two different equations, it is important to distinguish between these. It is proposed that the specific refractive energy calculated from the chemical composition be called the "chemical refractive energy", represented by K_C, and that the specific refractive energy calculated from the mean refractive index and density be called the "physical refractive energy", represented by K_P. The equations for these constants are:

$$K_C = \frac{k_1 \, p_1}{100} + \frac{k_2 \, p_2}{100} \cdots + \frac{k_n \, p_n}{100}$$

$$K_P = \frac{\bar{n} - 1}{D},$$

where k_1, k_2, k_n are the specific refractive energy constants for the constituents in the compound, p_1, p_2, p_n are the weight percentages of the constituents, $\bar{n}$ is the mean refractive index and D is density.

Ideally, K_P should equal K_C, and so the ratio K_P/K_C serves as a measure of the combined accuracy of $\bar{n}$, D and the chemical composition. A large deviation from 1.000 indicates either an error (or errors) in the data, or suggests that the Gladstone–Dale relationship does not apply to the compound in question. With respect to the latter point, note that the Gladstone–Dale relationship is not a law, as it has often been incorrectly termed, but an empirical relationship. Many compounds do not conform to the relationship. Notable among the exceptions are many simple oxides; I can offer no complete explanation for this. The reason that some compounds do not conform to the Gladstone–Dale relationship can certainly be attributed to the individual k values of some of the constituents; it has been shown in Part I of this series that different values of k for certain constituents must be used for different kinds of compounds. In the case of MgO, for example, k is 0.200 for most compounds and 0.225 for sulfates. In their list of specific refractive energy constants, Larsen & Berman (1934) noted many multiple values. It is possible that a different set of constants should be used for oxides and certain other compounds. Some additional alternative constants will be given in part IV of this series; part IV will also include data suggesting that the Gladstone–Dale relationship can be applied successfully to most minerals.

Before presenting some examples of calculations, it is important to discuss the determination of the mean refractive index, $\bar{n}$. Throughout this series of papers, the value of $\bar{n}$ is defined as the arithmetic mean of the principal refractive indices. In an optically isotropic material, $\bar{n} = n$. For uniaxial compounds, $\bar{n} = (2\omega + \epsilon)/3$, and for biaxial compounds, $\bar{n} = (\alpha + \beta + \gamma)/3$. Larsen & Berman (1934) defined the mean refractive index the same way, but warned that where the birefringence is very strong, the following values should be used for uniaxial and biaxial crystals, respectively: $\bar{n}' = \sqrt[3]{\omega^2 \epsilon}$ and $\bar{n}' = \sqrt[3]{\alpha\beta\gamma}$. For the refractive index range 1.300 to 2.000, the difference between $\bar{n}$ and $\bar{n}'$ is less than 0.001 for birefringences less than 0.100. Even with a birefringence of 0.200, this difference is less than 0.003 for the same refractive index range. As slight orientation errors of highly birefringent grains can cause relatively large errors in the usual immersion methods of refractive index determinations, the errors in these measurements may exceed the difference between the two calculated means. For this reason and because most minerals have birefringences well below 0.100, the arithmetic mean of the refractive indices is adequate for use with the Gladstone–Dale relationship.

TABLE 1. DATA FOR SYNTHETIC $Rb_2Mg(SO_4)_2 \cdot 6H_2O$

A Constituent	B Mol. Wt.	C ΣMol. Wt.	D Wt. %(p)	E k	F $\frac{kp}{100}$
Rb_2O	1 x 186.94 = 186.94		37.73	0.128	0.0483
MgO	1 x 40.30 = 40.30		8.13	0.225	0.0183
SO_3	2 x 80.06 = 160.12		32.32	0.177	0.0572
H_2O	6 x 18.02 = 108.12		21.82	0.340	0.0742
	Total	495.48	100.00		0.1980

α = 1.4672 $\bar{n}$ = 1.4713 $K_C = \Sigma\frac{kp}{100}$ = 0.198
β = 1.4689
γ = 1.4779 D = 2.38 g/cm³ $K_P = \frac{\bar{n}-1}{D}$ = 0.198

Compounds Without Halide or Sulfide Ions

The first example is a simple compound, $Rb_2Mg(SO_4)_2 \cdot 6H_2O$, one of Tutton's salts. Table 1 lists all the pertinent data for this compound. In the upper part of the table are the data necessary for the calculation of K_C. Column A lists the constituents, column B gives the molecular weight of each oxide constituent multiplied by the number of that "oxide" in the formula, column C gives the total molecular weight of each oxide, column D gives the weight percentages calculated from the figures in column C, column E lists the specific refractive energies of the constituents, and column F lists the products of columns D and E divided by 100. The summation of the values in column F is the chemical refractive energy (K_C) of $Rb_2Mg(SO_4)_2 \cdot 6H_2O$. The lower part of the table contains the refractive indices and density of the compound. The mean index of refraction ($\bar{n}$) and the density D are used to calculate the physical refractive energy K_P. In this example, $K_C = 0.198$, $K_P = 0.198$, and the ratio $K_P/K_C = 1.000$, which indicates a high degree of compatibility of the optical data, density and chemical data.

A mineralogical example is illustrated in Table 2. The analytical data pertain to an olivine (analysis no. 6, Deer et al. 1962) given by Hawkes (1946) for a forsterite from California. In this example, a chemical analysis is used rather than a theoretical composition, as in the previous example. Consequently, the summation of the $(kp)/100$ values must be multiplied by the factor 100/total; this changes the apparent K_C from 0.2036 to 0.2028, which is rounded off to 0.203. The same answer results if the individual weight percentages are "corrected" to a total of 100.00 wt. % before they are multiplied by their k values. For this forsterite, $K_C = 0.203$, $K_P = 0.202$, and

TABLE 2. DATA FOR FORSTERITIC OLIVINE NUMBER 6[*]

Constituent	Wt. %(p)	k	$\frac{kp}{100}$
SiO_2	40.84	0.208	0.0849
TiO_2	0.04	0.393	0.0002
Al_2O_3	0.19	0.207	0.0004
Fe_2O_3	0.13	0.268	0.0003
FeO	8.18	0.188	0.0154
NiO	0.19	0.176	0.0003
MnO	0.17	0.197	0.0003
MgO	50.27	0.200	0.1005
H_2O^+	0.37	0.340	0.0013
Total	100.38		0.2036
	100.00		0.2028

α = 1.649 $\bar{n}$ = 1.666 K_C = 0.203
β = 1.666
γ = 1.684 D = 3.30 g/cm³ K_P = 0.202

[*] Deer et al. (1962); original data of Hawkes (1946), M. Fleischer, analyst.

$K_P/K_C = 0.995$. The K_P/K_C value indicates a high degree of agreement for the data. In addition, the measured density (3.30 g/cm³) compares favorably with the density (3.324 g/cm³) calculated by the method outlined by Bloss (1952) and by the usual unit-cell method assuming a linear variation of unit-cell volume with respect to composition (3.31 g/cm³).

Another set of olivine data (Table 3) cited by Deer et al. (1962) will serve as an example of the use of the ratio K_P/K_C to discover errors. The data are listed under olivine analysis no. 19 and were taken from Ramdohr (1927). The value of K_C calculated from the chemical analysis (corrected to 100.00 wt. %) is 0.195; K_P from the physical data is 0.203. From these data, $K_P/K_C = 1.041$, which indicates

TABLE 3. DATA FOR FAYALITIC OLIVINE NUMBER 19[*]

Constituent	Wt. %(p)	k	$\frac{kp}{100}$
SiO_2	29.83	0.208	0.0620
FeO	69.48	0.188	0.1306
MnO	0.28	0.197	0.0006
H_2O^+	0.34	0.340	0.0012
H_2O^-	0.04	0.340	0.0001
Total	99.97		0.1945
	100.00		0.1946

α = 1.8235 $\bar{n}$ = 1.8551 K_C = 0.195
β = 1.8649
γ = 1.8770 D = 4.215 g/cm³ K_P = 0.203

[*] Deer et al. (1962); original data of Ramdohr (1927), J. Jakob, analyst.

TABLE 4. DATA FOR SYNTHETIC CARNALLITE, $KMgCl_3 \cdot 6H_2O$

Constituent		A Mol. Wt.	B Wt. %(p)	C k	D $\frac{kp}{100}$
0.5 K₂O		47.10	16.95	0.196	0.0332
MgO		40.30	14.50	0.200	0.0290
3 Cl		106.35	38.27	0.318	0.1217
6 H₂O		108.12	38.91	0.340	0.1323
	Total	301.87	108.63		0.3162
less 1.5(0) ≡ 3Cl		-24.00	-8.64	0.203	-0.0175
	Total	277.87	99.99		0.2987

α = 1.4665 β = 1.4753 γ = 1.4937	$\bar{n} = 1.4785$ $D = 1.60 \text{ g/cm}^3$	$K_C = 0.299$ $K_P = 0.299$

TABLE 5. DATA FOR CHONDRODITE NUMBER 3[*]

Constituent		Wt. %(p)	k	$\frac{kp}{100}$
SiO₂		33.60	0.208	0.0699
TiO₂		0.06	0.393	0.0002
Al₂O₃		0.24	0.207	0.0005
Fe₂O₃		0.05	0.268	0.0001
FeO		0.86	0.188	0.0016
MnO		0.16	0.197	0.0003
MgO		59.30	0.200	0.1186
H₂O⁺		1.46	0.340	0.0050
F		6.61	0.047	0.0031
	Total	102.34		0.1993
less O ≡ F		-2.78	0.203	-0.0056
	Total	99.56		0.1937
		100.00		0.1946

α = 1.600 β = 1.609 γ = 1.628	$\bar{n} = 1.612$ $D = 3.177 \text{ g/cm}^3$	$K_C = 0.195$ $K_P = 0.193$

[*] Deer *et al.* (1962); original data of Sahama *et al.* (1953),
H.B. Wiik, analyst.

possible errors among the data. Examination of the analysis shows that the mineral is almost pure end-member fayalite. Consequently, its physical properties should be close to those of pure Fe_2SiO_4. The measured density given for this olivine is 4.215 g/cm³, but for pure Fe_2SiO_4 it should be 4.392 g/cm³. A density of 4.39 g/cm³ and a mean refractive index of 1.854 to 1.858 will give a K_P of 0.195, which is identical to K_C. It seems reasonable to conclude that the measured density is wrong. The density calculated from K_C and $\bar{n}$ is 4.385 g/cm³.

COMPOUNDS WITH HALIDE IONS

The first example in this group is carnallite, $KMgCl_3 \cdot 6H_2O$. The pertinent data are shown in Table 4. Before proceeding with the calculations, particular attention should be paid to the computation of the weight percentages. Column A lists the molecular weights of the constituents multiplied by the number of units of each constituent in the formula. Note that a molecular weight equivalent to 1.5 oxygens is subtracted from the total. These 1.5 oxygens were added to the total molecular weight when K and Mg were expressed as oxides (0.5 oxygen for 0.5K₂O and 1.0 oxygen for MgO). The weight percentages in column B are obtained by dividing each figure in column A by 277.87 and multiplying by 100. Listed in column C are the values of $(kp)/100$. The value of $(kp)/100$ for the oxygen equivalent of Cl (O ≡ Cl) of 8.64 wt. % must be subtracted from the summation of $(kp)/100$ values for K₂O, MgO, Cl and H₂O. The result is a value of 0.299 for K_C. The value of K_P is also 0.299 and $K_P/K_C = 1.000$. Again, the data are compatible.

Another example is given in Table 5 for a fluorine-bearing chondrodite. The value of K_C calculated from the data is 0.194, K_P is 0.193, and K_P/K_C is 0.995, indicating compatibility of the data.

COMPOUNDS WITH S^{2-} IONS

The last example deals with minerals that contain sulfide ions: the helvite group. The end-members of this group are helvite, $Mn_4Be_3(SiO_4)_3S$, danalite, $Fe_4Be_3(SiO_4)_3S$, and genthelvite, $Zn_4Be_3(SiO_4)_3S$; they contain between 5.37 wt. % S (pure genthelvite) and 5.78 wt. % S (pure helvite). Dunn (1976) published 75 new analyses along with refractive indices for 72 of these and densities for 61 of them. Chemical, optical, and density data are given for 60 specimens. These 60 sets of data were used to calculate a specific refractive energy constant for S^{2-}. Using this constant of 0.628 as given by Mandarino (1978), values of K_C and K_P were calculated for the 60 specimens and the ratio K_P/K_C was obtained for each set of data. Forty-six (77%) of these sets of data have K_P/K_C ratios that depart by less than ± 0.019 from unity and are thus considered to have superior compatibility. Nine (15%) of the sets have K_P/K_C ratios between ± 0.020 and ± 0.039 from unity and only five (8%) of the sets of data have K_P/K_C ratios greater

TABLE 6. DATA FOR FOUR MEMBERS OF THE HELVITE GROUP

Dunn (1976)				This Study			Using D_{meas}		Using D_{calc}	
Analysis No.	Specimen No.	n	D_{meas} (g/cm^3)	K_C	a_0 Å	D_{calc} (g/cm^3)	K_P	K_P/K_C	K_P	K_P/K_C
13	H108172	1.747	3.37	0.212	8.133	3.63	0.222	1.047	0.206	0.972
47	127337	1.735	2.99	0.230	8.307	3.22	0.246	1.070	0.228	0.991
56	45634	1.731	3.00	0.227	8.294	3.24	0.244	1.075	0.226	0.996
60	123702	1.727	3.02	0.227	8.294	3.24	0.241	1.062	0.224	0.987

than $\pm$ 0.039 from unity. These are: nos. 13(1.047), 24(0.959), 47(1.070), 56(1.075) and 60 (1.062). It was felt that these larger deviations from unity indicated discrepancies among the data for these five specimens. Small fragments of each were requested from Mr. Dunn, who kindly supplied four of them (no. 24 could not be sampled as it is an exhibit crystal). X-ray powder diffraction patterns were produced from each fragment and the unit-cell edge was calculated from the d_{330} value from each pattern. The densities of the specimens, calculated from the unit-cell volumes and the chemical data given by Dunn (1976), are compared in Table 6 with Dunn's measured densities. In all four cases, the calculated densities are higher and result in higher values of K_P which, in turn, give values of K_P/K_C

much closer to unity. Interestingly, for specimen no. 45634, $a = 8.294$Å and D(calc.) = 3.24 g/cm^3; these data compare favorably with the data obtained by Holloway et al. (1972) for a specimen from the same locality: $a = 8.294(7)$Å and D(calc.) = 3.25 g/cm^3.

If one assumes that refractive index and density within the helvite group are additive properties, these can be calculated for each of the samples using the end-member values given by Glass et al. (1944) as modified by Dunn (1976). For the four specimens, the densities calculated in this way compare more favorably with those derived from unit-cell data than with the measured densities. The refractive indices calculated from the end-members are very close to the values measured by Dunn (1976). Clearly, some of the discrepancies indicated by the K_P/K_C ratios are due to errors in the measured densities, which probably reflect the presence of other phases.

TABLE 7. DATA FOR HELVITE*

Constituent	Wt. %(p)	k	$\frac{kp}{100}$
BeO	13.52	0.240	0.0324
CaO	0.08	0.210	0.0002
MnO	51.70	0.197	0.1018
FeO	0.96	0.188	0.0018
ZnO	0.55	0.158	0.0009
Al_2O_3	0.09	0.207	0.0002
SiO_2	31.79	0.208	0.0661
S	5.84	0.628	0.0367
Total	104.53		0.2401
less O ≡ S	−2.91	0.203	−0.0059
Total	101.62		0.2342
	100.00		0.2305

$n = 1.735$ $D = 2.99$ $K_C = 0.230$ $K_P = 0.246$

* analysis no. 47 of Dunn (1976).

TABLE 8. DATA FOR GENTHELVITE*

Constituent	Wt. %(p)	k	$\frac{kp}{100}$
BeO	13.02	0.240	0.0312
CaO	0.07	0.210	0.0001
MnO	5.40	0.197	0.0106
FeO	20.76	0.188	0.0390
ZnO	26.55	0.158	0.0419
SiO_2	29.90	0.208	0.0622
S	5.61	0.628	0.0352
Total	101.31		0.2202
less O ≡ S	−2.80	0.203	−0.0057
	98.51		0.2145
	100.00		0.2177

$n = 1.752$ $D = 3.45$ $K_C = 0.218$ $K_P = 0.218$

* analysis no. 12 of Dunn (1976).

Calculations involving compounds that contain S^{2-} ions are similar to those involving compounds with halide ions. One of Dunn's analyzed specimens of helvite (analysis no. 47) discussed earlier is used in Table 7 as an illustration. These data yield K_P and K_C values of 0.246 and 0.230, respectively. The K_P/K_C value of 1.070 indicates poor compatibility of the data. On the other hand, the data for a genthelvite (Dunn's analysis no. 12) listed in Table 8 give $K_P = 0.218$, $K_C = 0.218$, $K_P/K_C = 1.000$; the data show perfect compatibility.

CONCLUSIONS

The value of K_P/K_C can be used as a measure of the compatibility of $\overline{n}$, D and the chemical composition. The difference between K_P/K_C and 1.000 can be considered the "fractional error" (multiplication by 100 would give the "percentage error"). For example, a K_P/K_C of 0.978 represents an "error" of 0.022 (2.2%) and a value of 1.015 represents an "error" of 0.015 (1.5%). From the foregoing, one should not get the impression that a given value of K_C is free of error, because in some cases all or part of the error may lie in the chemical analysis or in the individual values of k.

The author uses the following arbitrary scale to characterize the compatibility of data:

$1 - (K_P/K_C)$	Compatibility
±0.000 to ±0.019	Superior
±0.020 to ±0.039	Excellent
±0.040 to ±0.059	Good
±0.060 to ±0.079	Fair
> ±0.079	Poor

The author has applied the K_P/K_C concept to numerous sets of data, both published and unpublished. Where the scale indicated poor compatibility, attempts were made to discover the source(s) of error. Often, as in the case of the helvite-group minerals, errors in density were detected; sometimes, the optical data were wrong; in a few cases, errors in the chemical analysis seemed most probable.

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