



TOWARD DEVELOPING IMPROVED MANDARINO COMPATIBILITY INDICES (K_p/K_c), BY USING STATISTICALLY SIGNIFICANT K_p VALUES

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ABSTRACT

A specific-refractivity (K_p) model for a whole-rock powder is represented. Successful, innovative completion of this reconnaissance to explore whether compatibility indices might be calculable even for whole-rock samples (not just for mineral separates) suggests that using only statistically-significant data sets when estimating specific refractivities would significantly improve precisions and accuracies of results. K_p and E% (percentages of fragments with RIs matching those of immersion liquids) profiles are drawn and interpreted. Automation of all related technologies and widespread cooperative work on the same standard samples are suggested, in order both to facilitate inter-laboratory instrument calibrations and to improve discrimination among samples.

Keywords: Refractive indices, whole-rock and mineral powders, emmons double-variation liquid-immersion refractometry, statistically significant data sets, 3D modeling, specific refractivity K_p , notation, Mandarino compatibility indices (MCI).

INTRODUCTION

In 1961, Gerald P. “Gerry” Brophy taught that geologists were ignoring refractive indices (RIs) to their own disadvantage. In almost the same breath, he said that some subjects had been completely removed from the literature. The combination of those instructions inspired this work.

The term “Specific Refractive Energy” was introduced by (Gladstone and Dale, 1863). Pointing to the preferred phrase “specific refractivity”, Donnay *et al.* (1980) nicely argued for dropping “energy” from the terminology and referenced (Gray, 1972). The preferred term is also found earlier, in both (Gray, 1957) and (Smith, 1909).

Mandarino wrote a series of papers (Mandarino, 1964, 1976, 1978, 1979, 1981, 2007). In (Mandarino, 1979), a distinction between K_p (physical) and K_c (chemical) specific refractivities was made. In this his paper, when without a subscript, K means K_p . A bulk chemical analysis is available for the subject sample (Langford, 1972, p. 36) but no attempt to calculate its K_c values has been made.

Larsen (1921) published what was to become (Larsen and Berman, 1934). Chapter 3 seems to be identical in both but K_p values were not listed in the tabulations of either book. Sadly, the (Fleischer *et al.*, 1984) “revision” of (Larsen and Berman, 1934) completely redacted Chapter 3 representation related to the law of Gladstone and Dale.

Mandarino (2007) wrote on page 1307: “It is discouraging to see the lack of optical data in descriptions of new species”. None of all that discouraged Mandarino from persevering in his amazingly gargantuan efforts to bring the chemical and physical aspects of that law into a cohesive and functional methodology, as realized by his K_p/K_c “compatibility index”, page 71 in (Mandarino, 1979).

Although such statements as

- 1) “Users of the Gladstone-Dale relationship should realize that wide differences between calculated and observed refractive indices and densities may be caused by the variation in k values” (Mandarino, 1964, where $k = K_p$),
- 2) “During the latter half of the nineteenth century several other refractivity-density relations were proposed but these had very restricted applicability and were little more than extrapolations from a limited number of experiments” (Kragh, 2018, p. 9; quoted by permission) and
- 3) “The practice of using the Gladstone-Dale relationship to minerals only gives an approximation because of the effects that different crystal systems have on the anisotropy of the crystal lattice and the resultant values of n (the index of refraction)” <http://webmineral.com/help/Gladstone-Dale.shtml>; tend to dissuade people from trusting K_p values (Nickel and Grice, 1998) included the requirement to report the Mandarino compatibility index when introducing and naming new, nonopaque minerals.

The central purpose of this paper is to encourage a general appreciation of the precisions and accuracies available from statistically-significant refractive index ($RI = n$) data sets, by demonstrating that K_p is a statistic that can be extremely-well determined. In addition to the Lorenz-Lorenz work discussed by (Kragh, 2018), many other formulations suggesting improvements to the simplest $K = \text{Refractivity/Density}$ approach (e.g., Sun *et al.*, 1940) have been encountered during literature reviews. To compare and contrast all such approaches is beyond present purposes.

Langford (1972, 1991, 2021a, 2021b) has worked extensively to investigate how much more might be learned from creating statistically-significant RI data sets; by studying RIs displayed by powdered rock and mineral fragments, via the double-variation, liquid-immersion methodology outlined by Emmons (1943) in Chapter 5; while ignoring all aspects related to optical orientations. Creation of statistically-significant RI data sets is made

possible by rapidly collecting RI data without optically orienting any fragment before comparing its RI to that of an immersion liquid (Tsuboi, 1923; Langford, 1972, 2021).

The experiment

A complex Density Model was created by calculating the K_p quotient of dividend mass (m ; colored red in contradistinction to the m used for meters) by Langford (2021b) over divisor E% (Langford, 2021a). Intricate and complex modeling efforts produced both cross-hatched Kriging and Radial-Basis, Thin-Plate-Splining models, which were then combined into one model of Density. A model of Refractivity ($RI - 1$) was also created. The $K_p = \text{Refractivity/Density}$ calculation was then performed via the GRID|MATH module of Surfer (Golden Software, Golden, Colorado, USA). However, the resulting K_p Model required rectification, so that its values would fall within the K_p bounds to be expected among the minerals known to be present in the sample.

Table 1. The K_p values calculated from USGS Bull. 1627 (Fleischer *et al.*, 1984).

	Mineral Species	α	β	γ	$(\alpha + \beta + \gamma)/3$	Refractivity	S.G.	Kcalculated	Bull1627 pp.
1	Tridymite	1.482	1.484	1.486	1.4840	0.4840	2.25	0.2151	128
2	Sanidine	1.52	1.525	1.525	1.5233	0.5233	2.56	0.2044	219
3	Sanidine	1.523	1.529	1.53	1.5273	0.5273	2.56	0.2060	220
4	Orthoclase	1.519	1.524	1.525	1.5227	0.5227	2.57	0.2034	219
5	Albite	1.528	1.532	1.539	1.5330	0.5330	2.61	0.2042	136
6	Albite	1.527	1.534	1.535	1.5320	0.5320	2.6	0.2046	221
7	Oligoclase	1.536	1.541	1.546	1.5410	0.5410	2.64	0.2049	138
8	Oligoclase	1.536	1.541	1.546	1.5410	0.5410	2.64	0.2049	223
9	Oligoclase	1.541	1.546	1.55	1.5457	0.5457	2.65	0.2059	224
10	Andesine	1.546	1.55	1.554	1.5500	0.5500	2.67	0.2060	140
11	Andesine	1.546	1.55	1.554	1.5500	0.5500	2.65	0.2075	225
12	Andesine	1.549	1.552	1.556	1.5523	0.5523	2.66	0.2076	226
13	Labradorite	1.555	1.558	1.562	1.5583	0.5583	2.69	0.2076	142
14	Labradorite	1.558	1.562	1.566	1.5620	0.5620	2.69	0.2089	143
15	Bytownite	1.563	1.568	1.573	1.5680	0.5680	2.71	0.2096	143
16	Bytownite	1.565	1.569	1.574	1.5693	0.5693	2.72	0.2093	144
17	Bytownite	1.563	1.568	1.573	1.5680	0.5680	2.71	0.2096	230
18	Bytownite	1.565	1.569	1.574	1.5693	0.5693	2.72	0.2093	231
19	Bytownite	1.571	1.577	1.583	1.5770	0.5770	2.74	0.2106	233
20	Anorthite	1.575	1.584	1.589	1.5827	0.5827	2.76	0.2111	236
21	OI Forsterite	1.653	1.664	1.686	1.6677	0.6677	3.35	0.1993	165
22	OI Chrysolite	1.661	1.68	1.697	1.6793	0.6793	3.45	0.1969	169
23	OI Chrysolite	1.661	1.68	1.697	1.6793	0.6793	3.15	0.2157	267
24	OI Chrysolite	1.683	1.704	1.722	1.7030	0.7030	3.53	0.1992	275
25	OI Hyalosiderite	1.71	1.733	1.75	1.7310	0.7310	3.69	0.1981	282
26	OI Tephroite	1.75	1.766	1.779	1.7650	0.7650	3.87	0.1977	287
27	OI Hortonolite	1.742	1.77	1.786	1.7660	0.7660	3.9	0.1964	289
28	OI Hortonolite	1.758	1.786	1.804	1.7827	0.7827	4.1	0.1909	291
29	OI Ferrohorton...	1.777	1.818	1.828	1.8077	0.8077	4.21	0.1918	295
30	OI Fayalite	1.803	1.843	1.851	1.8323	0.8323	4.3	0.1936	297
31	OI Fayalite	1.816	1.85	1.863	1.8430	0.8430	4.36	0.1933	298
32	OI Liebenbergite	1.82	1.854	1.888	1.8540	0.8540	4.6	0.1857	298
33	Augite	1.714	1.723	1.774	1.7370	0.7370	3.55	0.2076	179
34	Augite	1.725	1.73	1.75	1.7350	0.7350	3.42	0.2149	181
35	Pigeonite	1.684	1.684	1.707	1.6917	0.6917	3.4	0.2034	170
36	Pigeonite	1.696	1.698	1.721	1.7050	0.7050	3.38	0.2086	173
37	Pigeonite	1.714	1.714	1.742	1.7233	0.7233	3.44	0.2103	176

So, one can have to that end and under the possibly incorrect assumptions that

- 1) specific refractivities are not additive in the data from a whole rock powder RI study and
- 2) the β' values derived from study of unoriented fragments (Langford, 1972) are close enough to those $(\alpha + \beta + \gamma)/3$ values, for the present purposes: The data listed in Table 1 (calculated from (Fleischer *et al.*, 1984)) were created and were then used to calibrate the K_p Model, by re-scaling it to the 0.1857 and 0.2157 limits listed in column Calculated.

RESULTS

Figure 1 shows how models of RIs and densities were used to create the first-known model of K_p for a whole-rock sample, which was the initial purpose for the work reported here. Prior work by Langford (2021a, 2021b) shows the results over the visible spectrum but this paper is restricted to the region most pertinent to the K_p calculations reported (Larsen, 1921; Larsen and Berman, 1934) for 589.3 nm Fraunhofer D-line data.

INTERPREATION OF RESULTS

The results shown in Figure 1 are interpreted in Figure 2, where profiles of both E% (Langford, 1972, 2021a) and K_p are plotted over the Refractivity (RI - 1) abscissa (E% and K_p are orthogonal factors). To test whether magnetic effects previously seen to have affected RI probability levels ((Langford, 2021a) and the results shown in Figure 3; “This seems to be due to the magnetics...”) might also significantly affect K_p signals; profiles are drawn for each of the 589.0 nm and 589.6 nm sodium (Na) Fraunhofer-doublet lines and for their median, 589.3 nm (at which “D line” optical mineralogists have traditionally reported most RIs, at a previous 25°C standard temperature, now raised to 30°C). Significantly, no magnetic effects similar to those noted in (Langford, 2021a) and the results shown in Figure 3 are apparent in the K_p model. The three K_p profiles shown in Figure 2 fall nicely within the 0.1857 - 0.2157 K_p range of the full model. The most-remarkable and satisfactory aspect of the results shown in Figure 2 is that the E% levels in no way affect K_p levels. Interestingly the 589.0 nm and 589.3 nm K_p profiles flip-flop on either side of the 589.3 nm profile, which might be due only to modeling uncertainties; but which might also be worthy of much-closer study, particularly by workers concerned with intricate details of the complicated physical chemistries within the magnetic augitic-clinopyroxene solid solutions. The widest spread of the K_p profiles lies between the 1.770RI and 1.779RI limits. That is interesting because there is no associated sparsity of data; but that spread remains unexplained here.

The sanidine mentioned by Macdonald (Langford, 2021a) may be reflected in the K_p rise starting at by about RI1.5175 but sanidine may have been mostly hydrothermally altered to phillipsite; which would account for the signal centered at RI1.5100. Macdonald also mentioned tridymite, for which confirmation has been found in unpublished work by the author of this paper. However, the K_p values of the three profiles studied here are at higher RIs than those of tridymite [up to 1.481RI (Tröger, 1979, p. 30)]. So, Figure 2 does not include RIs in the tridymite range.

Interestingly, a K_p “catenoid” (from ~ RI1.510 to ~ RI1.591) spans more than the published RI limits of the feldspars. It begins somewhere in the region attributed to possible sanidine and phillipsite and ends over a peak attributed to volcanic glasses. This suggests that the same chemical domain spreads beyond the solid-solution range traditionally attributed to the feldspars, into the RI domains of chemically-related hydrothermal alterations and volcanic glasses.

Although olivine phases extend way beyond the 1.790RI maximum in Figure 2, data in this experiment are too sparse above that RI for serious consideration. However, the broad K_p humps of olivine on each side of the more-magnetic, augitic-clinopyroxene signals are very different from those jittery signals attributed to strong magnetic effects. In this sample, the broadest K_p hump at the higher end of the possible pigeonite range seems to be due entirely to olivine. The Appendix below lists extremely-high-resolution versions of Figures 1 and 2, together with the other supplemental data.

DISCUSSION

Abrupt transitions of RI-probability-levels produce “meromorphic” topologies of RI probability levels are seen in the Density Model that is shown in Figure 1. The related singularities are due to optical-physics considerations explored by (Saylor, 1935) and (Lipson and Lipson, 1969, p. 310.)

In (Fleischer *et al.*, 1984) and Chapter 3 of both (Larsen, 1921) and (Larsen and Berman, 1934) was redacted:

“The present edition, in holding to the objective of mineral identifications, omits discussion of statistics of mineral optics, as covered in Chapter 3 of the 1934 edition.... The Commission on New Minerals and Mineral Names, International Mineralogical Association (American Mineralogist, v. 67, pp.191-192) states for new minerals, ‘It is recommended that the relationship between chemical composition, density, and refractive indices, be checked by the Gladstone-Dale rule’” (Fleischer *et al.*, 1984), see also in (Commission on New

Minerals and Mineral Names of the International Mineralogical Association. 1982).

chemical ranges, concomitantly displaying ranges of RIs (and by implication of densities) even within a single

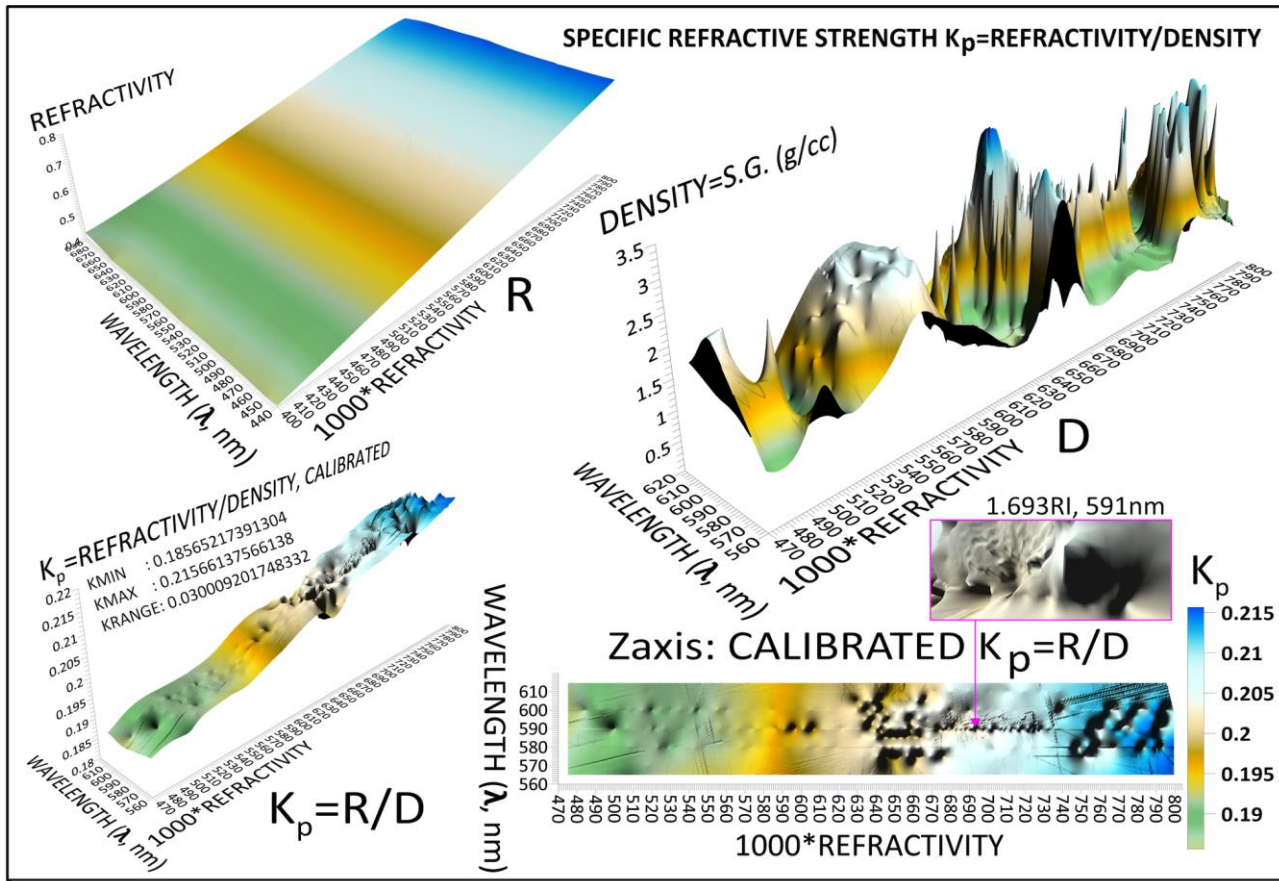


Fig. 1. The models of 1) The refractivity over most of the Emmons Surface within which data were taken during this study, 2) The densities estimated between 565 nm and 615 nm, and 3) The K_p values calculated over the same "Fraunhofer D Band" (ranging from 565 nm to 615 nm).

Interestingly, in work related to studying scatter of density data used in the denominator of the K_p calculation, Jaffe (1996; Figure 11.2, p. 123) drew a straight-line through measured densities and those calculated from the literature. But how statistically significant were the density values that Jaffe measured or drew from the literature? And how much would the respectably low level of scatter in Jaffe's Figure 11.2 be yet further reduced, were all data to have statistical significance; and were each element of each calculation to have been derived from studies of the same samples?

Some minerals (quartz, for example) have such narrow ranges of chemistry and related RIs that there is no need to obtain statistically significant estimations of their RIs and densities. However, many other mineral families (for instance, the feldspars; the most-common minerals in earth's crust) are comprised of solid solutions over

grain or grain fragment.

APPENDIX: THE SUPPLEMENTAL DATA

High-resolution images of Figures 1 and 2 are available at <https://tinyurl.com/ku6wet29> and <https://tinyurl.com/42epb2v4>, respectively. A copy of Figure 2, with finer divisions of the ordinate, is at <https://tinyurl.com/f2yuxrmd>. A high-resolution copy of Table 1 is at <https://tinyurl.com/5nd3ps3r>. A copy of statistics created by Surfer [v.16.6.484 (64-bit) Aug 22 2019], during modeling of the K_p surface and shown in Figure 1, is at <https://tinyurl.com/5d6k3rru>. [That two of those statistics are reported at the E^{-5} level reflects the fact that liquid-RI values were arbitrarily rounded to that level for purposes of work reported in this phase of the experiment.] The interactive Surfer file pictured in Figure 1 is at <https://tinyurl.com/djpd2uz7> (it must be manually downloaded to be seen). Logs of other work done since

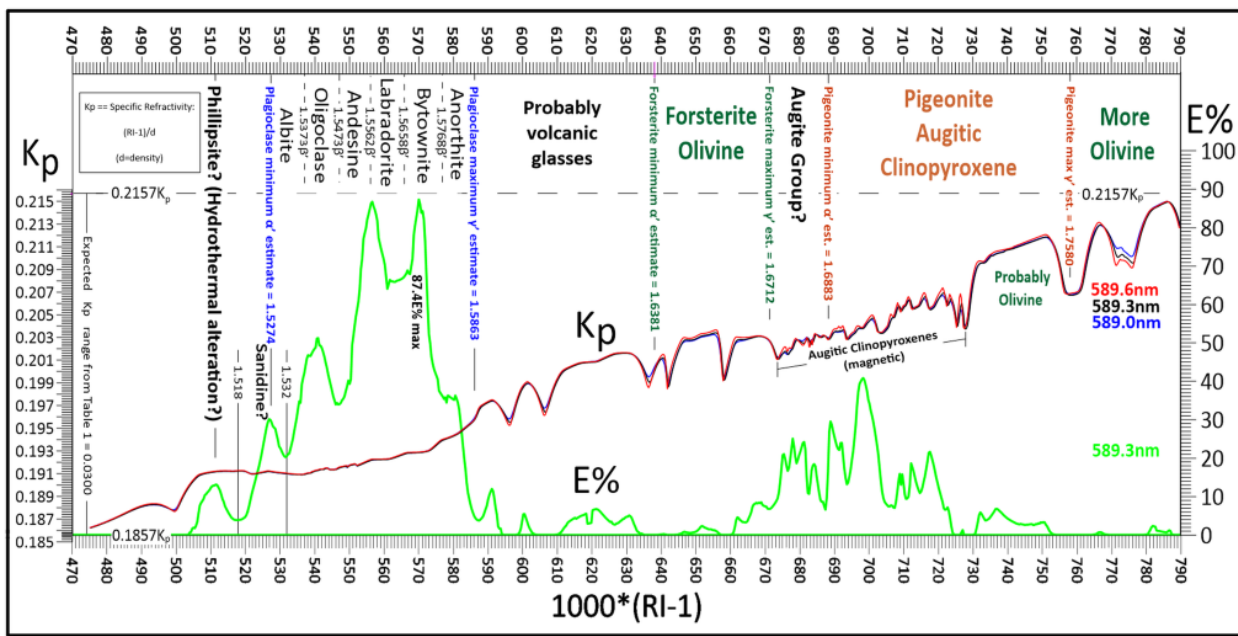


Fig. 2. The E% and K_p (orthogonal factors) versus the refractivity (RI-1).

13 February 2021 are available at <https://tinyurl.com/f68djxfh>, yielding many statistical reports from regressions, model formulas, and model images; as well as developments of thoughts and adaptations of modeling techniques to purposes at hand. Various Surfer models (with .SRF extensions) are there; they can be examined by those using recent Surfer versions.

CONCLUSION

A meaningful K_p model has successfully been created for a whole-rock powder, confirming the usefulness of the approach taken in (Langford, 2021b) and justifying the assumption that K_p values are not additive in whole-rock powders. Whether for studies of mineral separates (Langford, 1972) or of whole-rock powders (this work), data sets to estimate central tendencies of RI and density should be statistically significant. It makes no sense to expect less scatter among K_p estimates for such as plagioclase-feldspar separates, if the related estimates of RIs or densities have no statistical significance. Mineral separates, of all chemical-solid-solution minerals, should be examined as though they are whole-rock powders; because they are most likely to display RI variations that mimic the associated chemical variations sometimes optically to be seen even within very small (here, ~ 0.038 mm) fragments of any single grain (such as in zoned feldspar grains so often seen in thin sections).

Stating (Fleischer *et al.*, 1984) that others recommend that chemistries, densities, and RIs be checked by the GD rule is not a ringing endorsement of that advice. In this

author's considered opinion, a revision of (Fleischer *et al.*, 1984) is advisable. It would reinstate Chapter 3 (perhaps with appropriate updates) listing K_p values for every mineral. It would also list K_c , whenever those data are available for the same samples. Malcolm E. Back (who worked intimately with Joe Mandarino as a graduate student and who continues work relating K_c to K_p) in recent personal communications has convinced the author that at least some people have not lost sight of the importance of the Gladstone and Dale Law, though related literature is sparse. This reinforces the suggestion that a reinstatement of Chapter 3 be made, in a revision of (Fleischer *et al.*, 1984), wherein (for all appropriate tables) the empty spaces on the right side of the "Hardness, specific gravity, and fusibility" columns would be used to list all K_p and relevant K_c values. Future editions can include emboldened, K_p , K_c , and Mandarino Compatibility Index (hereby dubbed "MCI") values that would be emboldened when derived from statistically-significant data sets. Or, a set of entirely new publications can be initiated, to focus upon all data related to the MCI; and with distinctions being made between values from previous efforts and those based upon statistically-significant data for RIs, densities, and chemistries performed on the same samples.

Improvements to both data-collection methodologies and related data analyses (which in turn impact K_p/K_c calculations) might well facilitate revisions to petrographic, petrological, "mineralographic", and mineralogical classifications and nomenclatures (Mills *et al.*, 2009). This call for improvement is quite like some previous ones, for instance: "Such evaluation should have

been commonplace a generation ago” (Fairbairn *et al.*, 1951); and “Invariably, the organization that prepares a proposed standard cannot afford the time and effort to make all determinations necessary for standardization, and it must depend on the generosity of cooperating laboratories. Despite the large size of our organization, we must also depend on such assistance” (Flanagan, 1976).

Flanagan (1986) wrote on page 41 that the *Geostandards Newsletter* was first issued in January 1977. In 2004, the journal was renamed *Geostandards and Geoanalytical Research*; <https://tinyurl.com/p2yr346w> and flourishes <https://tinyurl.com/rajnt5nk>. However, no references pertinent either to the precision and accuracy of refractive-index data sets or to Gladstone and Dale specific refractivities are yet to be found in Web searches of that journal. Pointers to such articles found anywhere would be appreciated.

In order to create a truly practical, production-line system for coordinated physical and chemical descriptions and classifications applicable to all kinds of liquids and solids, automated data collection should be developed for all technologies related to the creation of K_p , K_c , and MCI data. Cooperative work applying various technologies to the same standard samples is encouraged, in order 1) further to improve precisions and accuracies of MCI and related data; 2) to facilitate sample descriptions, identifications, inter-sample discriminations, and inter-laboratory instrument calibrations; and 3) to discover whether improved RI sampling (in particular) might well prove to be the answer to the call (Paradis *et al.*, 2021) for a “*New Type of Sensor*”.

It has proven to be practicable to calculate a useful estimation of sample Densities by dividing the Mass (m) Model (Langford, 2021b) by a sufficiently well-determined E% Model (Langford, 2021a); then using that Density Model as the divisor into a well-determined Refractivity Model; in order to estimate K_p values that were calibrated against calculations based upon trustworthy data in the literature (Fleischer *et al.*, 1984). This is believed to be the first such work done on any rock sample.

The most-comprehensive previous published work focused upon studying RIs of particles in whatever positions they lay on the liquid-immersion slide was probably done by Tsuboi (1923): “... if grains of random orientation are used, ... repeat the determinations with a large number of grains”. It has not yet been possible to find a useful English translation of (Tsuboi, 1920), referenced in his 1923 paper and possibly pointing to a prior usage of the Beta-Prime notation (β') in (Langford, 1972) for the average RIs of “unoriented” fragments. Technologies now exist that should facilitate the

development of robots that would emulate the RI work reported here.

This work is not intended to suggest that optical mineralogists doing double-variation, liquid-immersion refractometry should work only with fragments in whatever positions they are first encountered when lying on a microscope slide. But the suggestion for generating only statistically-significant data is strongly linked to the implied suggestion for automation of all-possible optical-mineralogy processes; because to demand that people create statistically significant data sets for such attributes as 2V would result in production lines being brought to a halt. On the other hand, some workers might find the graphical approaches of (Langford, 1972) to be useful, when studying relatively small-sample RI distributions. Computer programs make analyses of such data much easier and more meaningful, whatever the number of sample counts may be. There will always be a need for traditional optical mineralogy.

In a personal communication, Malcolm Back wrote: “I think this [the appearance of optical data in descriptions of new mineral species] has improved somewhat in recent times and as I said the GD [Gladstone and Dale] compatibility of Mandarino is still used today and is very often quoted in new mineral descriptions”. This gives the present author reason to hope that this paper will be read by somebody who will recognize its significance and will improve K_p estimates; and thereby, MCI (K_p/K_c) precisions and accuracies.

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