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Density, refractivity, and composition relations of some natural glasses.

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WHILST some attention has in the past been devoted to the question of the properties of natural glasses, very little has been done to indicate the interrelations of the properties determined. In the case of the natural volcanic glasses, this is unfortunately only too evident from a study of the standard text-books. A determination of such properties as density, refractivity, and chemical composition, and the interrelations of these for particular glasses, is rarely to be found in the literature. The chief contribution to this subject is due to Stark,[1] who has examined the refringence of a large number of volcanic glasses. Unfortunately, there is no record of the density of these rocks, and the interrelation of these remained unknown. In the present paper a contribution on these lines is attempted. The work of previous investigators, where of import in the present studies, has been freely incorporated.

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[1] M. Stark, Tschermaks Min. Petr. Mitt., 1904, vol. 23, p. 536.

The densities of the glasses studied were in all cases corrected to water at 4° C. The actual temperature at which they were determined ranged from 13 to 15° C., and their accuracy is within the limits $\pm 0{\cdot}002$.

The refractivity was determined on polished plates, using a Herbert Smith refractometer with sodium-light, the determinations allowing an error not exceeding $\pm 0{\cdot}001$. In the case of *certain* basalt-glasses, however, the nature of the material necessitated resorting to the immersion method for refractivity determination, the refractive index being matched by a liquid, and this determined on the refractometer in the usual way. It should be remarked, that in the case of the refractometer determinations, for accurate values to be obtained, not only must the plate of material be plane, but a highly polished surface is also necessary. This latter prerequisite can be accomplished by use of a rouge plate.

The natural glasses investigated fall into two groups: (*a*) Tektite glasses, (*b*) Volcanic glasses. These will be considered in order.

(*a*) TEKTITE GLASSES.

A considerable literature has grown around those glass bodies which Suess has grouped together under the general name of tektite. Opinion has been divided as to their mode of origin, but considerable evidence has of late years been presented in favour of their meteoric origin, and some of the most potent evidence for this view is afforded by their chemical composition. An admirable summary of the present knowledge of these bodies has been quite recently given by F. E. Suess[1] in his paper 'Rückschau und Neueres über die Tektitfrage'. The present study is concerned with the two types of tektites, namely, moldavites and australites.

(i) *Moldavites.*

The specific gravity and refractive index of five specimens of moldavites (of unknown locality) in the Mineralogical Museum of Cambridge University were determined. It was not till these had been examined that the writer discovered that the density and refractivity of a large number of moldavites, from various localities in Bohemia and Moravia, had been determined by Ježek.[2] As these have been published in a journal not widely accessible, and moreover as they are of some impor-

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[1] F. E. Suess, Mitt. Geol. Gesell. Wien, 1914, vol. 7, pp. 51–121, 3 pls.

[2] B. Ježek and J. Woldřich, Bull. Intern. Acad. Sci. Bohême, 1910, vol. 15, pp. 232–245.

tance in connexion with the present investigation, they have been collected together with the present moldavites in Table I.

Table I. Moldavites.

[The first five are new determinations; the remaining twenty-eight are quoted from Ježek.]

Density.	n_{Na}.	K (Sp. Refr.).	Density.	n_{Na}.	K (Sp. Refr.).
2·337	1·488	0·2088	2·331	1·4893	0·2099
2·347	1·490	0·2088	2·333	1·4834	0·2072
2·350	1·489	0·2081	2·338	1·4858	0·2078
2·352	1·492	0·2092	2·339	1·4888	0·2090
2·367	1·492	0·2079	2·342	1·4863	0·2076
			2·346	1·4901	0·2089
2·303	1·4812	0·2089	2·347	1·4880	0·2079
2·304	1·4812	0·2089	2·348	1·4897	0·2086
2·305	1·4798	0·2081	2·352	1·4920	0·2092
2·309	1·4812	0·2084	2·354	1·4917	0·2089
2·317	1·4861	0·2098	2·355	1·4920	0·2089
2·321	1·4886	0·2105	2·356	1·4930	0·2093
2·321	1·4925	0·2122	2·357	1·4921	0·2088
2·323	1·4890	0·2105	2·359	1·4917	0·2084
2·325	1·4853	0·2087	2·360	1·4961	0·2102
2·326	1·4841	0·2081	2·362	1·4917	0·2082
2·326	1·4856	0·2088	2·364	1·4956	0·2096

Ježek determined the refringence for the wave-lengths corresponding to lithium-, sodium-, and thallium-light on an Abbe refractometer. For comparative purposes the refringence for sodium-light only is given in the table. In the third column is given the specific refractivity calculated from the relationship $K = (n-1)/d$, the values calculated from Ježek's results being given in addition. It will be seen that these moldavites show a considerable range both in density (2·303–2·367) and refractivity (1·4798–1·4961), with a range in the specific refractivity of 0·2072–0·2122.

The density, refractivity, and specific refractivity of the glasses of the isomorphous group of the albite-anorthite felspars, and of the mixtures of composition $CaSiO_3$—$MgSiO_3$, have been the subject of study by E. S. Larsen.[1] These results are of considerable utility in any consideration of the relations of natural glasses whether tektite or volcanic. In fig. 1 are plotted the relations, density and refractive index, for both these groups of artificial glasses. It will be seen that the *Ab-An* group of

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[1] E S. Larsen, Amer. Journ. Sci., 1909, vol. 28, pp. 263-274.

glasses fall on a curve approximately linear, but with a definite higher gradient near the anorthite end, as is evident from the higher value of the specific refractivity of the anorthite glass. The curve for the lime-magnesia metasilicate glasses is also approximately linear, but with a more pronounced gradient between the points corresponding to diopside and wollastonite glasses. The point for silica glass is also inserted, and falls above the prolongation of the *Ab-An* curve.

The moldavites on this graph group themselves along a line which lies above the prolongation of the *Ab-An* curve.

(ii) *Australites.*

Three specimens of these singular tektites have been examined, and with a fourth examined by Ježek constitute Table II. These plotted on the graph (fig. 1) again arrange themselves above the *Ab-An* curve, and conform closely in position to the extension of the moldavite locus parallel to the felspar-glass curve.

The last specimen in Table II represents an obsidianite from Pahang, Malay States.[1] It lies in close proximity to the *Ab-An* curve.

Table II. Australites (and Obsidianite).

Locality.	Density.	n_{Na}.	*K* (Sp. Refr.).
(Locality unknown. Ježek)	2·386	1·4981	0·2088
Mt. William, Victoria	2·393	1·504	0·2106
Mt. William, Victoria	2·443	1·520	0·2128
(Locality unknown)	2·453	1·519	0·2116
Obsidianite Pahang, Malay States	2·433	1·505	0·2076

(*b*) VOLCANIC GLASSES.

In Table III are collected fifteen examples of acid obsidians, the last eight of which are quoted from Ježek. They range in density from 2·330 to 2·413, in refractivity from 1·482 to 1·500, and in specific refractivity from 0·2044 to 0·2082, with the abnormal value of 0·2146 for the Newry pitchstone. Graphically plotted they range themselves in a field largely above the *Ab-An* curve but encroaching across it, the anomalous Newry rock being excepted. The field is distinct from that of the moldavites. The examples of marekanites which are also given in Table III fall normally in the obsidian field.

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[1] J. B. Scrivenor (Geol. Mag., 1909, p. 411) has given specific gravity determinations of obsidianites from this locality.

Table III.

Rhyolite-Obsidians.

Locality.		Density.	n_{Na}.	K (Sp. Refr.).
Newry, Ireland [Pitchstone]		2·330	1·500	0·2146
Yellowstone Park, Wyoming		2·353	1·482	0·2049
Mexico		2·361	1·490	0·2075
Forgia Vecchia, Lipari		2·363	1·490	0·2074
Rocche Rosse, Lipari		2·370	1·488	0·2059
Egypt [A scarab]		2·390	1·491	0·2054
Easter Island, Pacific		2·400	1·490	0·2046
Guamani, Ecuador	(Ježek)	2·386	1·4863	0·2082
Cali, Colombia	,,	2·344	1·4855	0·2070
Papayan, Colombia	,,	2·352	1·4852	0·2068
Clifton, Arizona	,,	2·355	1·4871	0·2068
Tokaj, Hungary	,,	2·379	1·4863	0·2044
Real del Monte, Mexico	,,	2·394	1·4912	0·2052
Otumbo, Mexico	,,	2·402	1·4917	0·2047
Greenland	,,	2·413	1·4956	0·2054

Marekanites.

Locality.		Density.	n_{Na}.	K (Sp. Refr.).
Okhotsk, Siberia	(Tilley)	2·353	1·482	0·2048
	,,	2·354	1·487	0·2069
	(Ježek)	2·358	1·4875	0·2067
	(Tilley)	2·359	1·489	0·2073
Nicaragua?	(Ježek)	2·383	1·4863	0·2041

In Table IV are collected the density and refractivity values of the remaining glasses, comprising the trachyte-obsidians and basalt-glasses.

Table IV.

Trachyte-Obsidians.

Locality.	Density.	n_{Na}.	K (Sp. Refr.).
Ascension	2·435	1·506	0·2078
Pantelleria	2·454	1·508	0·2078
Teneriffe	2·467	1·512	0·2076

Basalt-Glasses.

Locality.	Density.	n_{Na}.	K (Sp. Refr.).
Gallanach, Island of Muck	2·704	1·583	0·2156
Am Bile, Portree, Skye	2·716	1·576	0·2121
Rowley Regis, Staffs. [Dolerite, artificially fused.]	2·721	1·579	0·2128
Vesuvius [Lava of 1805]	2·769	1·586	0·2116
Caisteal an Duine Bhain, Muck	2·773	1·598	0·2157
Ardtun, Mull	2·811	1·598	0·2123
Reunion [' Pele's Hair ']	2·825	1·608	0·2152
Kau desert, Kilauea	2·841	1·603	0·2122
Halemaumau, Kilauea	2·851	1·605	0·2122
Kildonan, Island of Eigg	3·003	1·649	0·2161

The trachyte-obsidians from Teneriffe and Pantelleria fall completely on the *Ab-An* curve, but that of Ascension lies slightly above this curve. The basalt-glasses show a considerable range in density (2·704–3·003), refractivity (1·583–1·649), and specific refractivity (0·2116–0·2161). As a group, these glasses fall in a field intercepted by a prolongation of the *Ab-An* curve and the curve of the lime-magnesia metasilicate glasses. Two are, however, located above the prolongation of the felspar-glass curve, and the densest tachylyte falls below the extension of the curve of the metasilicate glasses.

The mean specific refractivities of the various glasses given in the tables is shown hereunder:

Tektites	Moldavites	0·2089
	Australites	0·2109
Volcanic Glasses	Rhyolite-obsidians	0·2065 [1]
	Marekanites	0·2060
	Trachyte-obsidians	0·2076
	Basalt-glasses	0·2136

THE RELATIONS BETWEEN DENSITY, REFRACTIVITY, AND COMPOSITION.

For the purpose of discussion of the interrelations of chemical composition, density, and refractivity, a second graph (fig. 2) has been constructed in which the density and specific refractivity of the various glasses have been plotted. In this manner the fields demarcating the various types of glasses are more clearly indicated.

It is possible to obtain a calculated value for the specific refractivity for any glass provided the specific refractivities of the individual constituents is known. Now the K values of silica, albite, and anorthite glasses are accurately known, that of hypersthene is taken from the values for $MgSiO_3$ and $FeSiO_3$, and that of corundum from the crystalline oxide. Gladstone long ago showed that the specific refractivity of a solution is additively related to the K values of its constituent members, and he further showed that the passage from the liquid to the crystalline state affected only very slightly the K value for a particular substance. This is also brought out in the studies of Larsen on the silicate glasses. There appears to be no record of the K value for orthoclase glass, but it may be calculated with some confidence from a determination of the specific refractivity of orthoclase itself (0·2056). A value of K for FeO

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[1] Omitting the anomalous Newry pitchstone, the mean becomes 0·2060.

was obtained from fayalite, and from it those for magnetite and the $FeSiO_3$ molecule can be computed. A value for Fe_2O_3 is obtained from haematite. The following are the values that have been used in the calculations recorded below :

Specific Refractivities of 'Normative' Constituents.

	K.		*K.*
SiO_2	0·2074	Mg_2SiO_4	0·2027
$KAlSi_3O_8$	0·2056	Fe_2SiO_4	0·1964
$NaAlSi_3O_8$	0·2053	$FeO.Fe_2O_3$	0·330
$CaAl_2Si_2O_8$	0·2132	$(CaCl)Ca_4(PO_4)_3$ ...	0·201
$CaSiO_3$	0·2163	$FeO.TiO_2$	0·304
$MgSiO_3$	0·2103	Al_2O_3	0·191
$FeSiO_3$	0·2000	H_2O	0·333

(i) *Moldavites.*

The chemical composition of moldavites from Bohemia and Moravia have been cited and discussed by F. E. Suess (loc. cit.) and H. S. Summers,[1] and their anomalous composition when referred to normal terrestrial glasses is strikingly displayed in the calculated 'norms' of the various analyses. How these chemical characters are related to their density and refractivity remains for treatment. Plotted on fig. 2, the moldavites occupy a field distinct from the rhyolite-obsidians, characterized by a higher specific refractivity and lying to the left. In the analyses set down by Suess, the range of the silica percentage is 77·69–82·68. The 'normative' composition of six analyses shows the following limits :

Quartz	56·8 to 63·8 per cent.
Orthoclase	13·3 ,, 17·2 ,,
Albite	2·1 ,, 6·8 ,,
Anorthite	6·4 ,, 15·3 ,,
Corundum	2·3 ,, 6·0 ,,
Hypersthene	3·8 ,, 10·0 ,,

It is clear that the very high contents of silica glass ($K = 0·2074$), anorthite glass (0·2132), and hypersthene glass (for enstatite $K = 0·2103$), are the contributing factors to the position of the moldavite field. This will be the more apparent when the 'normative' constitution of the rhyolite-obsidians is considered.

In the case of a moldavite from Radomilitz, the 'norm' calculated from an analysis given by Suess is : quartz 63·8, orthoclase 13·3, albite

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[1] H. S. Summers, Proc. Roy. Soc. Victoria, 1909, vol. 21, pp. 423–443.

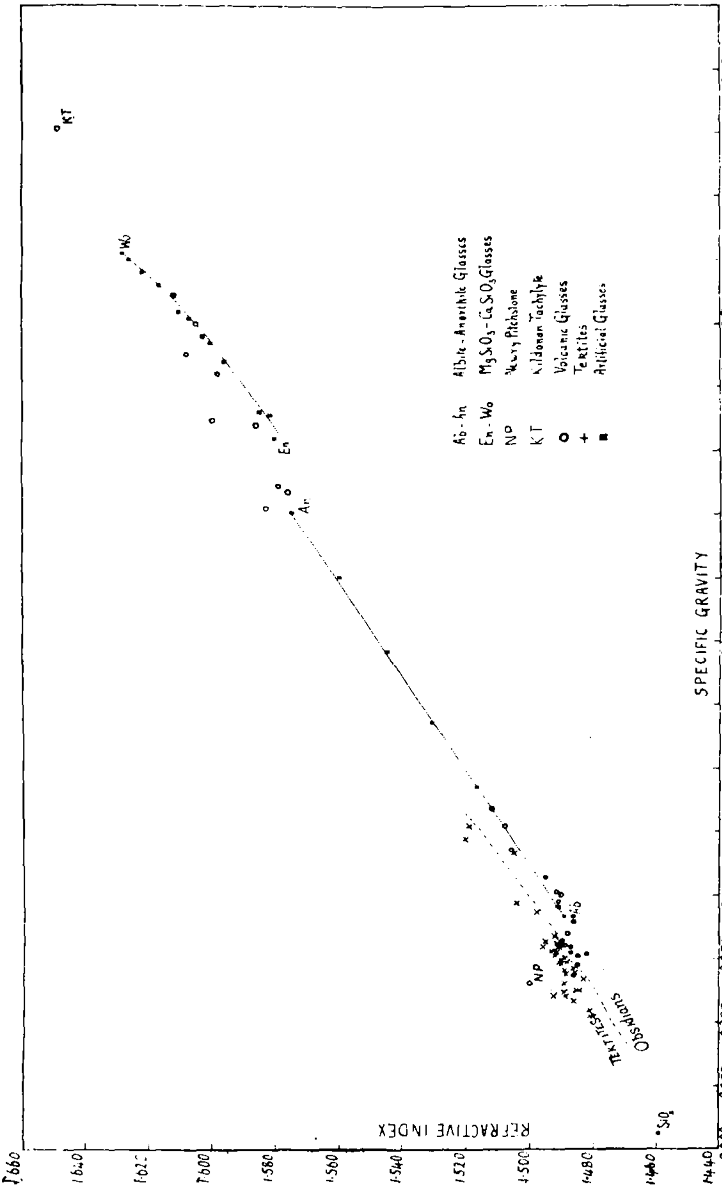

FIG. 1.—Graph plotting density and refractivity of glasses (See p. 277.)

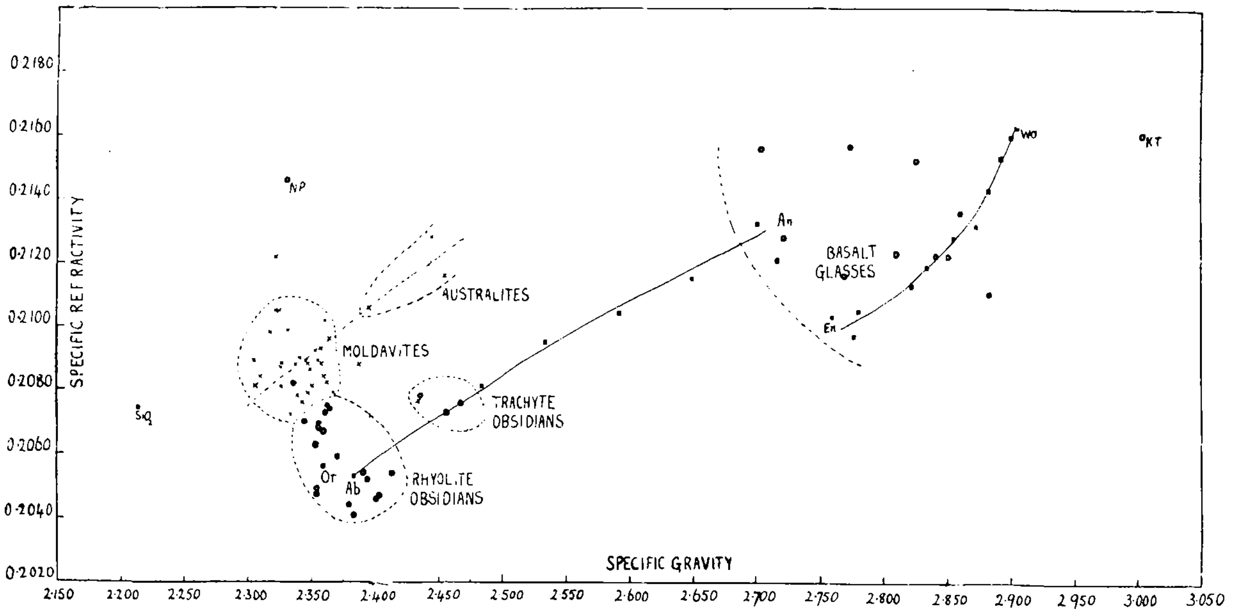

FIG. 2.—Graph plotting density and specific refractivity of glasses. (See p. 280.)

2·6, anorthite 11·1, corundum 3·1, hypersthene 6·2 per cent. From this the specific refractivity of the glass is calculated to be 0·2074. Three other moldavite analyses calculated in this way all gave $K = 0{\cdot}2072$.

(ii) *Australites.*

The chemical composition of a series of australites has been discussed by Summers (loc. cit.). He clearly shows that, compared with terrestrial volcanic glasses, these bodies possess an abnormal composition. This is reflected in the plotted positions of the australites in fig. 2. The limits of the 'normative' compositions of eight analysed specimens is as follows:

Quartz	38·6 to 52·7 per cent.
Orthoclase	10·6 ,, 15·0 ,,
Albite	8·9 ,, 14·7 ,,
Anorthite	12·0 ,, 18·6 ,,
Corundum	0·9 ,, 4·3 ,,
Hypersthene	9·1 ,, 15·6 ,,
Iron-ores	1·2 ,, 2·8 ,,

and the density limits of these specimens 2·385–2·454. The density of the analysed australites varied inversely as the silica percentage, the density in fact being an index of the approximate composition. The density limits of the four australites whose K values have been here determined are 2·386–2·453. A calculated value of K obtained for an analysed australite ($d = 2{\cdot}454$) from Coolgardie gave 0·2099. A second analysed australite ($d = 2{\cdot}454$) gave the value 0·2090.

Ježek considered, as a result of his studies of the refringence and density, that the moldavites could not be separated from the rhyolite-obsidians. It is clear, however, that this view cannot be maintained. The importance of the specific refractivity as a means of separation into fields is made manifest by the graphical plot of fig. 2. The same features are illustrated in the case of the australites, when they are compared with trachyte-obsidians of similar specific gravity.

The characteristics of the tektite glasses amply confirm their divergence from terrestrial glasses—a divergence which was first indicated in their chemical composition. This abnormality of chemical composition of the tektites, which is reflected in their fields in the graphical plots of figs. 1 and 2, although evidence of a negative kind, points—apart from other lines of study, as Suess and Summers have maintained—to their ultimate meteoric origin.

(iii) *Rhyolite-Obsidians.*

In fig. 2 the rhyolite-obsidians group themselves in a field which is intersected by the albite end of the *Ab-An* curve. The majority fall above this line. The 'norms' of rhyolite-obsidians show over 90 per cent. of quartz, orthoclase, albite, and anorthite. The positions of three of these mineral glasses are already marked on the graph. The point for orthoclase may be obtained from the determination of Tesch[1] that the refringence of orthoclase glass is 1·485. The specific gravity is therefore 0·485/0·2056 = 2·359.

From the specific refractivity of these glasses it is clear that normally the positions of the obsidians should be above the albite end of the *Ab-An* curve. The mean value of *K* given by fourteen obsidians is 0·2060. This point can be illustrated by a calculation of the specific refractivity of two Lipari obsidians from the 'norms' given by Washington's analyses,[2] and of which the specific refractivities are known from actual determination:

	Lipari Obsidians from	
	(*a*) Forgia Vecchia.	(*b*) Roccho Rosse.
Quartz	29·8	28·8
Orthoclase	28·6	29·7
Albite	34·9	35·3
Anorthite	1·7	2·2
Diopside	1·7	1·0
Hypersthene	1·3	1·9
Magnetite	0·9	0·7
Ilmenite	0·1	0·1
Water	1·0	0·3
K (calculated) ...	0·2088	0·2074
K (observed)	0·2074	0·2059–0·2063

The influence of water content on the specific refractivity is evident in the case of the obsidians just now treated, for the excess water in specimen (*a*) has contributed 23 units to its specific refractivity, whilst the variation in all the remaining constituents involves only a displacement of 9 units. The most remarkable instance, however, of the influence of water on the specific refractivity is afforded by the case of the Newry pitchstone, which, as seen from the graph, occupies an anomalous position quite beyond the obsidian field. An early but imperfect analysis of this rock by Knox showed it to contain an abnormally high content of water. A determination of the water content on the material actually utilized for the specific refractivity estimation

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[1] P. Tesch, Proc. Sci. K. Akad. Wetens. Amsterdam, 1903, vol. 5, pp. 602–605.

[2] H. S. Washington, Amer. Journ. Sci., 1920, vol. 50, p. 449.

confirms this point. The total water amounts to 7·04 per cent. Herein lies the explanation of the high K value of this glass. The specific refractivity of the glass minus its contained water can be calculated from the K value of water:

$$100\,(0{\cdot}2146) = 93\,(X) + 7\,(0{\cdot}333),$$

whence X is equal to 0·2057, a value quite close to the mean of the K values for obsidians (0·2060).

The discrepancies which are most likely to occur in the determination of K for these rhyolite-obsidians are those consequent on the development of small crystals of quartz or felspar as crystallites within the glass. As these constituents have a higher density than the glass, the apparent density of the glass will be in excess of the real, hence the K value will be too low, and the low K values for some of these obsidians are probably to be explained in this way. Gas cavities react in the opposite sense.

The Siberian marekanites derived from a perlitic obsidian at Okhotsk are inseparable from the rhyolite-obsidians, and are enclosed within the same field (fig. 2).

(iv) *Trachyte-Obsidians.*

The trachyte-obsidians are characterized by a higher specific gravity and a higher mean K value when compared with the rhyolite-obsidians. This is in accord with the lower content of silica and higher content of iron oxides. A pantellerite-obsidian from Costa Zeneti, Island of Pantelleria, has been analysed by Washington. The quartz in the 'norm' has fallen to 15 per cent., whilst the 'normative' ferromagnesian minerals total 16 per cent. Similarly, in the Ascension Island obsidian, the percentage of ferromagnesian minerals equals 12. In the normal rhyolite-obsidians, the content of ferromagnesian minerals does not exceed 4–6 per cent.

(v) *Basalt-Glasses.*

Of the ten basalt-glasses examined, four represent lavas, a fifth an artificially-fused dolerite, whilst the remainder are tachylyte selvages to Tertiary dykes from the Western Isles of Scotland. Specimens of the lava from the crater of Halemaumau, and a lava from the Kau desert, Kilauea, were very kindly forwarded to me by Dr. H. S. Washington, and the densities and refractive indices were determined on these specimens. The Halemaumau lava is that analysed by Ferguson for

Messrs. Day and Shepherd.[1] The Pele's hair from the Island of Reunion was derived from the eruption of 1860. The analyses of Boiteau, quoted by Lacroix, show that the Reunion basalt-lavas are closely analogous to the Halemaumau type (cf. lava of 1863, Reunion Island).[2] The Vesuvian obsidian is a potassic type, the vitreous equivalent of the leucite-tephrites.

Analyses of the tachylytes of Ardtun, Mull, and Am Bile, Skye, have been quoted by Judd[3] and Cole.[4] The analyses show that these two glasses are not true basalt types, but have mugearitic affinities, and they are so separated by Harker.[5] Their refractive indices and densities are lower than the true basalt-tachylytes. The same would appear to apply to the tachylytes of Gallanach and Caisteal an Duìne Bhaìn, Muck.[6]

The specific gravities of the tachylytes, and so their specific refractivities, tend to be somewhat vitiated by the presence of crystallites in the glass. If these be felspar the actual density of the glass will in general be greater than the apparent value, and the apparent specific refractivities therefore will be above the true value. On the other hand, in the true basalt-tachylytes, if the crystallites are magnetite or pyroxene, the apparent specific refractivities will be below the true values, and this is probably the case for the Kildonan tachylyte, which contains crystallites of iron-ore.

The majority of the basalt-glasses fall in the field intercepted by the pyroxene curve and the line joining anorthite glass with wollastonite glass, a feature to be expected when the dominance of the pyroxenes and felspars in the minerals of the 'norm' is considered. The iron-ores tend to locate their points to the right and above this field. The high values for the Gallanach and Caisteal an Duìne Bhaìn glasses, together with their comparatively low densities, suggest that water is here responsible. The glassy lavas, which may be expected to be practically anhydrous, fall within the field, and in the case of the Kilauean examples are close to the pyroxene curve. An analysis of the Kildonan tachylyte has been made, and is given hereunder.

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[1] A. L. Day and E. S. Shepherd, Ann. Rep. Smithson. Inst., 1913, p. 286.

[2] A. Lacroix, Compt. Rend. Acad. Sci. Paris, 1912, vol. 154, p. 253.

[3] J. W. Judd and G. A. J. Cole, Quart. Journ. Geol. Soc., 1883, vol. 39, p. 455.

[4] G. A. J. Cole, ibid., 1888, vol. 44, p. 303.

[5] A. Harker, Tertiary Igneous Rocks of Skye, Mem. Geol. Survey, Great Britain, 1904, p. 342.

[6] A. Harker, Geology of the Small Isles, Mem. Geol. Survey, Scotland, 1908, p. 157.

Tachylyte selvage of basic andesite, Kildonan, Eigg.

SiO_2	48·25
Al_2O_3	14·06
Fe_2O_3	3·39
FeO	9·38
MgO	6·60
CaO	10·82
Na_2O	2·54
K_2O	0·61
H_2O (above 105°)	2·08
H_2O (below 105°)	0·56
TiO_2	2·47
	100·76

Calculated 'Norm'.	
Orthoclase	3·87
Albite	21·34
Anorthite	24·84
Diopside	23·12
Hypersthene	13·12
Olivine	1·62
Magnetite	4·84
Ilmenite	4·68
Water	2·63
	100·06

The molecular ratio FeO/Fe_2O_3 for this glass is 6·2, whilst the value for the crystalline basic andesite of similar composition is only 3·0. This is in agreement with the results of Washington,[1] who finds 'that in many effusive lavas, ranging from rhyolites to basalts, ferrous oxide dominates very greatly over ferric oxide in the glassy forms, while ferric dominates over ferrous oxide, or at least is in much greater relative amount, in the docrystalline or holocrystalline forms'. In the following table the specific refractivities calculated from two analyses—that of the Kilauean basalt lava,[2] and that of the Kildonan tachylyte—are given.

	Glassy Lava, Halemaumau, Kilauea.		*Tachylyte of basic andesite, Kildonan, Eigg.*	
	Per cent.	*K.*	Per cent.	*K.*
Orthoclase	3·3	0·00678	3·9	0·00801
Albite	21·0	0·04311	21·4	0·04393
Anorthite	20·9	0·04456	24·8	0·05287
$CaSiO_3$	12·9	0·02790	11·8	0·02552
$MgSiO_3$	7·6	0·01598	7·0	0·01472
$FeSiO_3$	4·6	0·00920	4·3	0·00860
$MgSiO_3$	10·4	0·02187	8·1	0·01703
$FeSiO_3$	6·3	0·01260	5·0	0·01000
Mg_2SiO_4	2·9	0·00588	0·9	0·00182
Fe_2SiO_4	1·9	0·00373	0·7	0·00137
Magentite	2·3	0·00759	4·8	0·01584
Ilmenite	4·7	0·01429	4·7	0·01429
Apatite	1·0	0·00201	—	—
Water	0·2	0·00066	2·6	0·00865
K (calculated)	...	0·2161	...	0·2226
K (observed)	...	0·2122	...	0·2161

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[1] H. S. Washington, Amer. Journ. Sci., 1920, vol. 50, p. 458.

[2] A. L. Day and E. S. Shepherd, Ann. Rep. Smithson. Inst., 1913, p. 286.

The specific refractivities calculated from the analyses of the four rocks quoted in the foregoing are now collected for comparison with the actual K values as directly determined.

	K (obs.).	K (calc.).
Obsidian: Rocche Rosse, Lipari	0·2061	0·2074 [1]
Obsidian: Forgia Vecchia, Lipari	0·2074	0·2088 [1]
Basalt-glass: Halemaumau, Kilauea	0·2122	0·2161
Tachylyte: Kildonan, Eigg	0·2161	0·2226

The calculated values of K are throughout somewhat higher than the observed values, and this difference increases with the basicity of the rock. The larger discrepancies in the case of the basalts suggests that the errors arise in the K values taken for the iron-bearing minerals as magnetite and ilmenite. The differences are perhaps not greater than might be expected when it is considered that the K values for these denser minerals are not known with any great degree of accuracy, the values for FeO, Fe_2O_3, and TiO_2 being taken from those of fayalite, haematite, and rutile in the crystalline state. It is to be expected therefore that the specific refractivity of any glass is related to its constituent 'normative' minerals by the relation

$$K = \frac{k_1 w_1 + k_2 w_2 + k_3 w_3 + \ldots}{100},$$

where w_1, w_2, w_3 represent the percentage weights of these minerals and k_1, k_2, k_3 their respective specific refractivities. Or the refringence of the glass is given by

$$n = 1 + d\left[\frac{k_1 w_1 + k_2 w_2 + k_3 w_3 + \ldots}{100}\right],$$

where d is the specific gravity of the glass.

As silica is the preponderating oxide of natural glasses it is to be expected that the refractivity should show some relation to the silica content. Tesch has even suggested that silica percentage might be gauged by a determination of the refractive index, and gives a graphical plot correlating these variables for plutonic rocks in the fused state. Stark has likewise correlated the refractive index of natural glasses with their silica percentage. It is not to be expected, however, that this relationship should be more than a rough generalization. Certain constituents, although present in quite small amount, may influence in a high degree both the refringence and the specific refractivity. Of these, water, magnetite, and ilmenite are outstanding examples. In this

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[1] The K value for orthoclase using the specific gravity of the St. Gotthard adularia is 0·2035, in which case substituted here would reduce these values to 0·2067 and 0·2082 respectively without affecting the values for the basic glasses.

connexion the acid pitchstone of Newry, with a K value of 0·2146 and $n = 1{\cdot}500$, is a conspicuous example of the influence of water in solid solution in the glass. Quite as remarkable is the Lipari pumice glass analysed by Washington,[1] which, with a silica content of 71·7 per cent. and 3·35 per cent. of water, has a refractive index of 1·499, whilst the related Lipari obsidians showing 74 per cent. of silica have a refractive index not exceeding 1·490. The state of oxidation of the iron in the glass is also an important factor, influencing the specific refractivity and the refractive index, for while ferrous oxide contributes a K value of 0·192, ferric oxide has the much higher value of 0·393.

The influence of water on the Refraction of Rhyolite-Glasses.

Apart from the influence of water in raising the aggregate specific refractivity of glasses containing this constituent, it has a marked effect, as already noticed, on the refraction. For the three analysed and closely comparable rhyolite-glasses of Lipari its influence is seen in the following table:

Glass.	Water content.	Refraction.	Density.
Rocche Rosse	0·31 per cent.	1·4885	2·370
Forgia Vecchia	1·01 ,,	1·490	2·363
Monte Pelato	3·35 ,,	1·499	2·320

The composition of these glasses is so closely similar that practically the whole of these differences of refraction are to be attributed to the addition of water.

Provided no change of volume is involved, the refraction of any mixture is related to the refractions of its constituents in the proportion of their volumes. This can be simply shown as follows. Firstly, the Gladstone and Dale relationship gives

$$(1) \qquad \frac{n_1-1}{d_1}(w_1) + \frac{n_2-1}{d_2}(w_2) = \frac{n-1}{d}(w_1+w_2),$$

where n_1, n_2, n and d_1, d_2, d are the respective refractions and densities of components and mixture, and w_1, w_2 the weights of the components. This equation may be written

$$(2) \qquad n_1-1\,(v_1)+n_2-1\,(v_2) = n-1\,(v).$$

Also

$$(3) \qquad v_1+v_2 = v.$$

Whence by addition of (2) and (3) we have

$$(4) \qquad n_1v_1+n_2v_2 = nv, \quad \text{or} \quad n = \left(\frac{n_1v_1+n_2v_2}{v_1+v_2}\right).$$

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[1] H. S. Washington, loc. cit., p. 462.

In the case of a volume change, equation (2) yields

$$(5) \qquad n-1 = \frac{n_1-1\,(v_1)+n_2-1\,(v_2)}{v}, \text{ or}$$

$$(6) \qquad n = 1 + \frac{n_1v_1+n_2v_2-v_1-v_2}{v}$$

Equation (6)[1] shows that the magnitude of departure of a mixture from additive relations of its refractive index to that of the components is dependent on the value of v. If there is any marked volume contraction, this is accompanied by refractivity values, which may exceed the value of either of the components. Among liquids, such relations are not unknown, and it may be noticed here that the binary systems, ethyl alcohol, methyl alcohol, and water, show such relationships.[2]

Through the kindness of Dr. H. S. Washington, in forwarding a specimen of the rhyolite-pumice of Monte Pelato for specific gravity determination, the following table can now be drawn up:

Locality.	Density of			Expansional volume change (*b*) to (*c*).
	Anhydrous 'norm' (*a*).	Hydrous 'norm' (*b*).	Vitreous rock (*c*).	
Rocche Rosse	2·685	2·622	2·370	10·6 per cent.
Forgia Vecchia	2·686	2·593	2.363	9·7 ,,
Monte Pelato	2·623	2·485	2·320	7·1 ,,

The calculation of the 'normative' densities has been shown by Iddings to give very concordant results in the case of holocrystalline rocks when compared with their actual densities, and the specific gravities here used are those given by Iddings in his paper.[3] The difference between the 'normative' density and the actual density afford a measure of the volume change in the passage from the vitreous to the crystalline state for rocks of this composition.

When the Rocche Rosse and Forgia Vecchia rocks are compared, the densities of the anhydrous 'norms' are seen to be identical, but with the entry of water the higher content of the Forgia Vecchia specimen reduces its specific gravity to a much greater extent. When, however, the actual densities of the vitreous rocks are compared, the same hiatus is not apparent. Water in the hydrous 'norm' is calculated as a

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[1] Two slightly different relations to that expressed in equation (6), are derived by C. Pulfrich (Zeits. phys. Chem., 1889, vol. 4, pp. 561–569), and F. Schütt (Zeits. phys. Chem., 1892, vol. 9, pp. 349–377), in which the specific refractivities are considered as not being strictly additive.

[2] J. Holmes, Trans. Chem. Soc., 1915, vol. 107, p. 1471.

[3] J. P. Iddings, Amer. Journ. Sci., 1920, vol. 49, pp. 363–366.

component mechanically mixed. That a proportionate hiatus in density is not observed in the actual vitreous densities, can only be accounted for on the assumption that the addition of water to the glass is accompanied by a contraction of volume. We may consider the Forgia Vecchia specimen to consist of a rock glass of the Rocche Rosse composition to which is added water—without introducing any appreciable errors due to actual differences in the remaining constituents. We have then 99·3 parts by weight of glass of density 2·370 and refr. index 1·4885, and 0·7 parts by weight of water, giving a glass of refraction 1·490. If there were no contraction, the resultant density should be 2·347, whereas the actual density is 2·363. The figures for the Monte Pelato pumice are even more striking. This rhyolite may be considered as composed of ninety-seven parts of glass of Rocche Rosse type, plus three parts of water. Mechanically mixed, the resultant density is 2·276, whereas the actual density is 2·320.

The 'normative' compositions of the three rhyolites, and those of the mechanical mixtures regarded as the equivalents of the Forgia Vecchia and Monte Pelato rocks, are given in the subjoined table.

'Norm.'	Roche Rosse. (I)	Forgia Vecchia. (II)	Mixture of 99·3 per cent. of I + 0·7 per cent. water.	Monte Pelato.	Mixture of 97.0 per cen of I + 3·0 pe cent. water.
Quartz	28·8	29·8	28·6	26·9	27·9
Orthoclase	29·7	28·6	29·5	30·9	28·8
Albite	35·3	34·9	35·0	35·5	34·2
Anorthite	2·2	1·7	2·2	0·6	2·1
Diopside	1·0	1·7	1·0	—	1·0
Hypersthene	1·9	1·3	1·9	1·9	1·9
Magnetite	0·7	0·9	0·7	0·7	0·7
Ilmenite	0·1	0·1	0·1	0·1	0·1
Water	0·3	1·0	1·0	3·4	3·3
	100·0	100·0	100·0	100·0	100.0
Refraction	1·4885	1·490	1·486	1·499	1·478
Density	2.370	2·363	2·347	2·320	2·276
K (observed)	0·2061	0·2074	—	0.2150	—
K (calculated)	0·2074	0·2088	—	0·2113	—

The differences observed in the silicate contents of the two rocks and the mixtures derived by addition of water to the original Rocche Rosse type are quite small; and from the closeness of the refringence and density values for the 'normative' mineral glasses, it follows that the variations will not significantly modify the values themselves. The observed and calculated *K* values have been placed, together with the densities and refractions, in the adjoining columns. The relations here observed—refringence and specific volume—are graphically plotted in the accompanying diagram (fig. 3). The marked volume contraction associated with the presence of water is also expressed in the diminishing expansional volume change (10·6 per cent.–7·1 per cent.) shown in the table on p. 291.

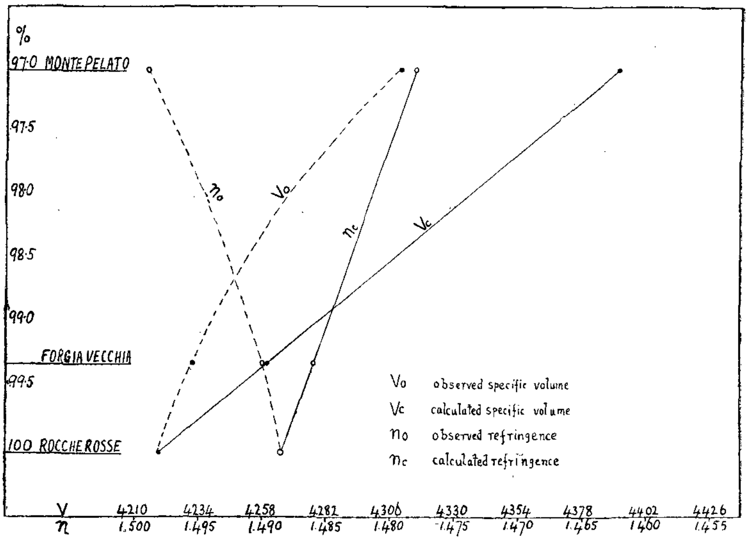

Fig. 3.—Graph showing observed and calculated refractive indices and specific volumes of the three analysed Lipari glasses.

Some apology is necessary for the incompleteness of the present study. Owing to the pressure of other work, however, its continuation has been impossible, and it is hoped that its presentation at this stage is not unwarranted. The writer is indebted to Dr. A. Harker, who first proposed this work, for encouragement and advice during its progress, and to Dr. H. S. Washington for valuable material. The work was carried out in the Mineralogical Department of the University, and the writer wishes to thank Dr. A. Hutchinson for the facilities placed at his disposal and for his kindly interest.

Summary.

(1) The natural glasses investigated fall into two groups: (*a*) Tektite glasses, embracing moldavites and australites, and (*b*) Volcanic glasses, including rhyolite-, trachyte-, and basalt-obsidians.

(2) When their physical properties, density, and refringence are graphically plotted, these glasses are seen to group themselves into distinct fields. For comparison, the glasses of the albite-anorthite felspars and the glasses of composition $CaSiO_3$—$MgSiO_3$ are similarly treated. The field of the tektites is sharply demarcated from those of the rhyolite- and trachyte-obsidians. The basalt-glasses including tachylytic types form a field which is enclosed by the $CaSiO_3$—$MgSiO_3$ curve and the prolongation of the *Ab-An* curve.

(3) In the same way the graphical plot of the relations, density and specific refractivity ($K = (n - 1)/d$), is broken up into fields separating the tektite and volcanic glasses. The mean specific refractivities of the various groups of glasses is given by—moldavites 0·2089, australites 0·2109, rhyolite-obsidians 0·2065, marekanites 0·2060, trachyte-obsidians 0·2076, and basalt-obsidians 0·2136.

The characteristics of the tektite glasses amply confirm their divergence from terrestrial glasses, established on other grounds, and support the contention of Suess and Summers with regard to their meteoric origin.

(4) The specific refractivities of five analysed glasses are compared with the values calculated from the specific refractivities of the component oxides, or 'normative' minerals. A notable correspondence is revealed, when the imperfection of the optical data with regard to the K values of a number of these oxides is considered.

The presence of water in a glass has a marked effect on the K value of the glass, as is shown by the Newry pitchstone, which with a water content of 7·04 per cent. has a specific refractivity of 0·2146. Water has a further influence, however, in that addition of water to certain glasses not only raises the K value of the glass, but has a marked effect on the refringence. This is to be observed in three analysed rhyolite-glasses from Lipari. It is shown that together with an increase in the refringence, a notable degree of contraction is involved. The three analysed glasses can be regarded as mixtures of one type, with varying amounts of water, and the departure from a mechanical mixture relation is indicated by their specific gravities (1) 2·370, (2) 2·363 instead of 2·347, (3) 2·320 instead of 2·276.

Lastly, in certain cases, an approximate estimate of the volume change accompanying the passage from the vitreous to the crystalline state is given by a comparison of the actual vitreous density and the density calculated from the 'norm'. For the three rhyolite-obsidians mentioned above the expansional volume changes (on fusion) are 10·6, 9·7, and 7·1 per cent. respectively.