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Volatility of oxides from silicate melt and the origin of moldavites

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SUMMARY. The volatilities of SiO_2, Na_2O, and Fe_2O_3[1] increase with increasing thermal function within the range 1600 to 2100 °C during the melting of a sandy-silty clay. The remaining melt is to some extent enriched in Al_2O_3, K_2O, CaO, and MgO. Chemical analyses of the sandy-silty clay, of the glasses melted from the latter, of the silicate material evaporated and newly condensed and their correlation with increasing thermal function and bulk density of glasses, as well as with the hitherto known quantitative data on moldavites, testify to the fact that in moldavites evaporated and newly condensed silicate material substantially prevails over material that passed directly from a solid to a fluid state. Evaporation of the silicate material from the rocks of the impact crater caused a significant change in relations between major oxides compared to the original relations in the source rocks. Thus, the hypothesis that tektites are directly remelted rocks of the Earth's crust, based on over-all chemical similarities of tektites and some rocks in major oxides, appears incorrect.

MOLDAVITES are inhomogeneous silicate glasses belonging to tektites; they occur in southern Bohemia and Moravia, Czechoslovakia. Studies that contributed to a more detailed knowledge of moldavites were made by Barnes (1964, 1968, 1969), Bouška and Povondra (1964, 1966), Bouška, Faul, and Naeser (1968), Rost (1964, 1966, 1967), Konta (1966, 1971a, b, c, 1972), and Konta and Mráz (1969). Most moldavites consist of chemically more or less different glassy schlierens and portions and contain a small admixture of lechatelierite. If the material was more heated moldavites are generally more homogeneous and contain less or no lechatelierite. Moreover, they enclose bubbles of different sizes and shapes (Konta, 1972). Sizes of the bubbles vary from the microscopic to a maximum of about 15 mm in diameter. The shape of moldavites and the quality of their surfaces also differ, and so do the bulk density, density, and refractive index. Quantitative relations between chemical composition and bulk density as well as other quantitative petrographical relations were explored by Konta and Mráz (1969) and Konta (1972).

The shapes of 1666 moldavites from nineteen localities in Bohemia and of 617 moldavites from ten localities in Moravia were studied by Konta (1971b, c), who evaluated statistically the differences in the shapes of less-heated moldavites from Bohemia and of more-heated moldavites from Moravia by means of a digital computer. Less-heated, and therefore more viscous, tektite material containing a higher amount of lechatelierite grains and bubbles in the majority of Bohemian localities shows a relatively lower average sphericity than the more-heated tektite material from

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[1] 'Fe_2O_3' represents total iron oxides recalculated to Fe_2O_3.

the Moravian localities or from sporadic occurrences in Bohemia (especially the Radomilice area). According to Konta (1971b) the less-heated moldavite material was ejected from the impact crater on relatively short and flat trajectories (Bohemia) while the more-heated material was ejected into geographically more distant places (Moravia) or travelled on relatively long and steep trajectories (several Bohemian localities).

Rost (1972) compiled ideas concerning the origin of tektites and moldavites and their source material. Most of them are based on conjecture and assumption. Little experimental work has been done to support individual hypotheses. The papers that deal with the absolute age of tektites (e.g. Zähringer, 1963; Gentner, Lippolt, and Schaeffer, 1963; Faul, 1966; McDougall and Lovering, 1969) or record the kinship or dissimilarity of tektite substance with the common silicate materials of the Earth's crust are of high value for the problem of genesis. The majority of these papers show that the terrestrial origin of tektites is the most probable one.

In our opinion, many authors are wrong in postulating that tektites originated by a direct melting of the terrestrial silicate material to glass. The postulate requires that the chemical composition of tektites be similar to that of the source rocks (e.g. eruptive rocks according to Chapman and Scheiber, 1969, sedimentary rocks according to Bouška, 1968). On the other hand Rost (1972, p. 184), as well as other authors, points to the fact that there is an 'incompatibility between the chemical composition of the Ries glasses and moldavites, although the values of absolute age of both substances found at a distance of 300 km from each other are identical [14·4 Myr]. The paucity of occurrences of imperfectly melted terrestrial rocks and also of the transitions of tektite glass into source rocks is believed by Rost to speak against Spencer's (1933) theory'. On p. 181 he adds: 'It seems that the crater glasses from Ries are derived from the underlying gneiss. Moldavites may have originated from some of the overlying Jurassic argillaceous rocks (provided that the moldavites also came from the Ries crater). Nearly all the Jurassic rocks from the Ries crater show a high content of CaO! However, the moldavites contain only 2 % of calcium oxide. The disappearance of CaO in the formation of moldavites is difficult to explain!'

In the volume of abstracts of the Meteoritical Society 34th Annual Meeting, held at Tübingen, Germany, Gentner (1971) and Konta (1971a) presented, independent of one another, a hypothesis based on experimental data that moldavite substance originated from the silicate material evaporated during thermal shock after the impact of a big cosmic body on the Earth's surface. During the explosive ejection, the gaseous silicate phase seized some of the solid and liquid phases (quartz grains melted to lechatelierite in particular). In the opinion of both authors moldavites were formed by condensation of silicate vapours during their passage through the atmosphere.

Theoretical presumptions

Judging from the quantitative analytical data so far obtained on moldavites from Bohemian localities we arrive at three important and generally valid conclusions: with increasing SiO_2 content the content of $Al_2O_3+CaO+MgO$ decreases and the bulk density and density decreases roughly in the same manner; this principal relation between the chemical composition and density can be partly distorted by an increased content of Fe_2O_3 and K_2O. With an increase of SiO_2 content and a decrease in bulk density the content of lechatelierite and of bubbles in moldavites generally decreases.

The moldavites containing a relatively lower amount of strongly melted lechatelierite or no lechatelierite at all and only a few bubbles originated from more heated material than the moldavites with a relatively higher amount of lechatelierite in grains of angular shape typical of original quartz and with more bubbles.

Combining these conclusions with an assumption that moldavites (and probably also other tektites) originated by a remelting of solid silicate substance to glass, then the bulk density of glass should decrease with an increased melting temperature. In such glasses obtained by a direct remelting of the remaining silicate material the SiO_2 content should increase simultaneously with an increase in temperature. The moldavites showing the features of relatively more heated glass from localities in Bohemia generally display this tendency. However, the experimental work whose results are presented in the following paragraphs has disclosed that this tendency is inversed in the silicate melt. The theoretical presumption of the origin of moldavites and probably of other tektites by sudden remelting of a chemically suitable rock directly into glass is not applicable. It is of interest to note that this presumption has not been based on any experimental data and has only been deduced from the simple idea that glass must be produced by a direct melting of a solid substance as can be seen, e.g., in glassworks.

Experimental part

Silicate material studied. A green-grey sandy-silty clay with sparse psephitic fragments of the Upper Miocene age from Besednice in southern Bohemia was selected for the study of fugacity of major oxides in silicate material at high temperatures. It contains about 35 % quartz, 32 % muscovite, 17 % kaolinite, 7 % montmorillonite, 6 % feldspars, and accessory fragments of rocks and heavy minerals (Störr and Konta, 1964). The sediment is mined for the production of bricks. The fact that this material represents a weathering crust of the most common rocks, i.e. gneisses and granitic rocks, which probably covered a large area in central Europe in the period of the fall of moldavites, make it particularly suitable for the problem studied. The weathered rock was transported over a small distance and was rapidly deposited in the south Bohemian lake basin.

Melting. The temperatures during the thermal shock that accounted for the origin of tektites were distinctly higher than the temperatures that could be produced in our laboratory. The easiest way to produce the volatility of oxides in the silicate melt at low temperatures was to increase melting time. The finely powdered and carefully homogenized sediment, weighed in 10 g charges, was put into a shallow molybdenum crucible with a diameter of 50 mm. The melting was performed in the range 1600 to 2100 °C and separate charges were prepared for each experiment, planned to reach maximum temperatures spaced at 100 °C. The crucible was placed in the centre of the graphite winding of the furnace. The furnace was closed with a graphite cover and air was expelled by a stream of argon (5 l/min for 20 min). As soon as the heating of the furnace began, the flow of argon was reduced to 3 l/min. In about 45 to 70 minutes the desired temperature was reached and maintained for 20 minutes. Electric current was then switched off, and the furnace was allowed to cool. The flow of argon was kept at 3 l/min during cooling.

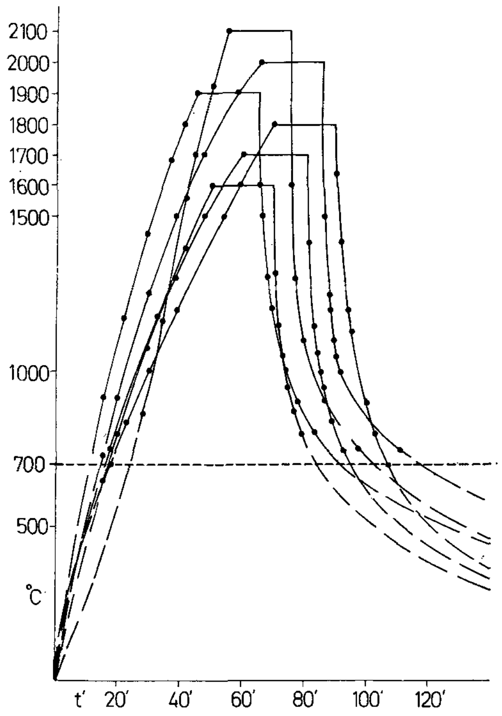


Fig. 1. The heating (and melting) course in minutes (t') of six samples of the sandy-silty clay from Besednice and the attained maxima between 1600 and 2100 °C.

The temperature of the side of the molybdenum crucible was measured by an optical pyrometer every 5 to 10 minutes during heating. Within the 20-minute period of the maximum temperature at least five readings were taken. The points on temperature curves in fig. 1 show the frequency of temperature measurements. Judging from experience with the apparatus used, the temperatures measured have to be regarded as the minimum values; the actual temperatures were probably several tens of degrees higher. The apparatus did not permit the temperature to be held constant with time and, consequently, all further experimental results should be evaluated with regard to temperature as well as to the time of heating (fig. 1). Thus in figs. 2 and 3 a thermal function *A* is plotted instead of temperature; *A* is defined for each experiment as proportional to the area under the temperature curve of fig. 1 multiplied by the maximum temperature reached, and thus has the dimensions T^2t. In the original drawing of fig. 1, 10 °C and 1 minute are each represented by 1 mm, so that 1 mm^2 represents 10 (°C min); *A* was taken, for figs. 2 and 3, in units of 10^7 (°C)^2 min. For the six experimental runs:

T_{max}	1600 °C	1700	1800	1900	2000	2100
T-t area	3693 mm^2	4857	5847	5343	6960	5706
A	5·91 units	8·26	10·52	10·15	13·92	11·98

Chemical analyses. Table I contains chemical analyses of both the sandy-silty clay from Besednice and of the silicate glasses obtained by melting of this sediment using different values of the above thermal function. In the lower part of the table the

TABLE I. *Chemical analyses of natural sediment, of silicate glasses prepared from it by melting using different thermal functions (A), and of evaporated silicate material condensed on the lid of the furnace*

	Natural sediment	$A = 5·91$ at 1600 °C	$A = 8·26$ at 1700 °C	$A = 10·15$ at 1900 °C	$A = 10·52$ at 1800 °C	$A = 11·98$ at 2100 °C	$A = 13·92$ at 2000 °C
SiO_2	73·23	77·04	76·78	75·95	73·68	72·63	69·35
TiO_2	0·58	0·65	0·66	0·68	0·78	0·86	0·95
Al_2O_3	14·09	14·78	15·18	15·89	18·14	18·83	21·88
Fe_2O_3	1·87	0·17	0·19	0·15	0·04	0·04	0·01
FeO	0·32	1·86	1·83	1·73	1·59	1·96	1·76
MnO	0·01	0·01	0·01	0·01	0·01	0·02	0·02
MgO	0·44	0·48	0·43	0·46	0·61	0·59	0·67
CaO	0·32	0·34	0·35	0·37	0·39	0·42	0·48
Na_2O	0·82	0·90	0·90	0·86	0·82	0·76	0·82
K_2O	3·36	3·65	3·69	3·76	3·82	3·82	3·97
H_2O^+	3·16	—	—	—	—	—	—
H_2O^-	1·84	0·20	0·20	0·23	0·13	0·18	0·13
P_2O_5	0·01	0·02	0·01	0·01	0·01	0·00	0·00
Total	100·05	100·10	100·23	100·10	100·02	100·11	100·04
Total Fe_2O_3	2·23	2·23	2·22	2·07	1·81	2·22	1·97
Recalculated to 100 % of water-free material:							
SiO_2	77·04	77·12	76·76	76·05	73·77	72·68	69·41
TiO_2	0·61	0·65	0·66	0·68	0·78	0·86	0·95
Al_2O_3	14·82	14·80	15·17	15·92	18·16	18·85	21·91
Fe_2O_3	1·98	0·17	0·19	0·15	0·04	0·04	0·01
FeO	0·34	1·86	1·83	1·73	1·59	1·96	1·76
MnO	0·01	0·01	0·01	0·01	0·01	0·02	0·02
MgO	0·46	0·48	0·43	0·46	0·61	0·59	0·67
CaO	0·34	0·34	0·35	0·37	0·39	0·42	0·48
Na_2O	0·86	0·90	0·90	0·86	0·82	0·76	0·82
K_2O	3·53	3·65	3·69	3·76	3·82	3·82	3·97
H_2O^+, H_2O^-	—	—	—	—	—	—	—
P_2O_5	0·01	0·02	0·01	0·01	0·01	0·00	0·00
Total	100·00	100·00	100·00	100·00	100·00	100·00	100·00
Total Fe_2O_3	2·36	2·23	2·22	2·07	1·81	2·22	1·97
Partial analyses of evaporated material:							
SiO_2	—	—	*	—	90·60	*	86·42
Al_2O_3	—	—	*	—	3·85	*	6·89
Fe_2O_3†	—	—	*	—	4·10	*	5·15
MgO	—	—	*	—	nil	*	nil
CaO	—	—	*	—	nil	*	nil
Na_2O	—	—	0·21	—	0·52	0·33	0·61
K_2O	—	—	1·16	—	1·16	0·66	1·19
TiO_2	—	—	*	—	0·24	*	0·17

* Not determined. † Total iron as Fe_2O_3.

analyses are recalculated to 100 % of water-free material. Reducing conditions prevailed during the melting and this led to a substantial change in the ratio Fe_2O_3:FeO; therefore the value of total Fe_2O_3 is given for every analysis. The resulting values of major components recalculated to 100 % are plotted in fig. 2 against the thermal function A, and in fig. 3 the percentage of SiO_2 is expressed relating to the bulk density, which nearly coincides with the true density in these almost bubble-free glasses.

In four melts dark green-grey fibrous 'glass wadding' was found on the lid of the furnace. It is a material of the silicate melt evaporated and condensed on the cooler lid. Its quantity was sufficient for a partial chemical analysis only (Table I); the results contribute to our knowledge on the behaviour of major oxides in silicate melt under high temperatures.

Bulk density. The bulk density of each glass was determined by weighing in air and in distilled water at 20 °C of five selected fragments in which no bubbles had been found under a magnifying glass. Because some microscopic bubbles occur in some fragments, the values obtained should be taken for bulk density (fig. 3) rather than for density, although both values are nearly identical. The density of a dried natural sandy-silty clay from Besednice determined pycnometrically is 2·70.

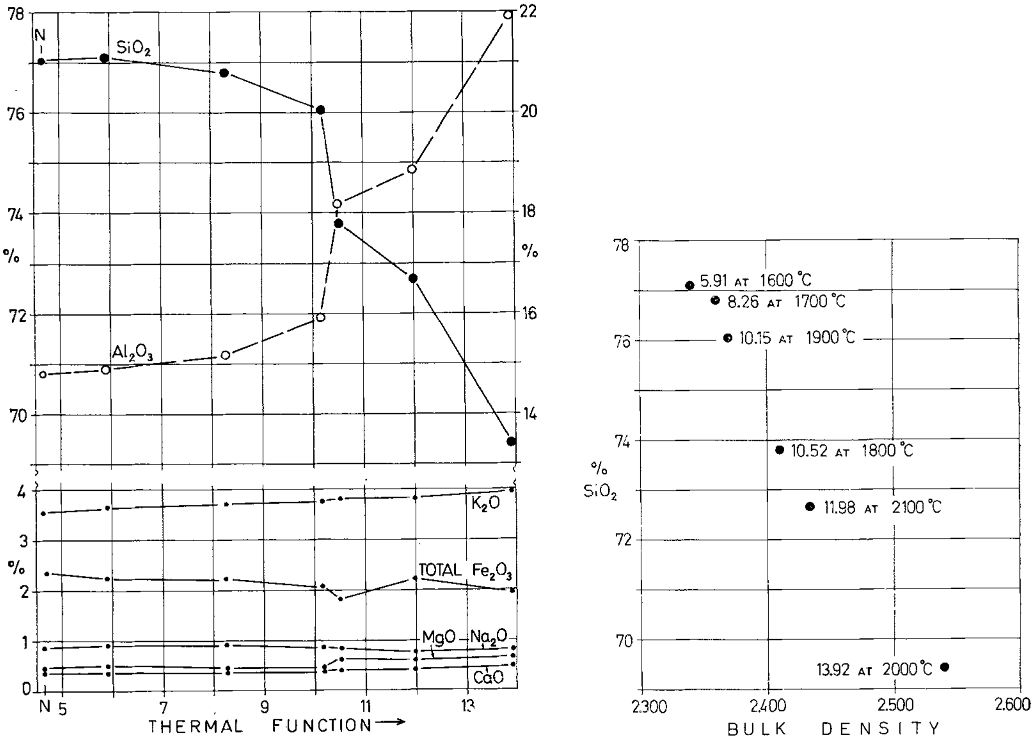
Discussion and conclusions

The data obtained are particularly valuable for recognition of the differences in the volatility of major oxides from the silicate melt under high temperatures, and for hypotheses concerning the origin of tektites, including the problem of their source.

Fig. 2 shows that with the consumption of a higher amount of energy an increasing amount of SiO_2 evaporates from the silicate melt; Na_2O and total Fe_2O_3 behave similarly. On the other hand, the volatility of Al_2O_3 is substantially slower, so that during the higher input of thermal energy the melt enriches relatively in alumina; K_2O, CaO, and MgO behave similarly. The differences established in the volatility of oxides under high temperatures help us to explain why more-heated moldavites (Barnes, 1968, 1969; Konta and Mráz, 1969; Konta, 1971a) from the localities in Moravia show relatively higher ratios of $(SiO_2+Fe_2O_3+Na_2O)/(K_2O+CaO+MgO)$ than the less-heated moldavites from localities in Bohemia. The differences in the contents of Al_2O_3 observed in the Bohemian and Moravian moldavites are not, however, very distinct. Philpotts and Pinson (1966) reported that the Moravian moldavites have lower and relatively constant average contents of MgO and CaO, whereas Bohemian tektites have high and variable alkali-earth contents. They also explained the variations in terms of fractional volatilization that occurred during the brief thermal event.

Konta and Mráz (1969) demonstrated that in the moldavites from Bohemian localities the bulk density (and also true density) decreases with increasing SiO_2 content and with the features of higher heating. Deviations from a linear increase of SiO_2 with decreasing density are caused by variation in the ratios of other light (Al, Mg, Na) and heavy (Fe, K, Ca) elements in moldavites. Simultaneously, these authors have shown that the more-heated Bohemian moldavites contain generally a higher amount of SiO_2 than those less heated.

However, the ratios mentioned are inverse in glasses prepared by a direct melting of the sandy-silty clay. With an increasing thermal function their SiO_2 content decreases, the Al_2O_3 content increases (fig. 2) and, simultaneously, bulk density (fig. 3) rises. Judging from these conspicuous differences between the glasses prepared by direct melting of a solid silicate substance using different thermal functions and the differently heated moldavites, the widely held opinion that tektite material originated by direct melting of a silicate rock to glass should be thoroughly revised. The silicate glasses prepared under high temperatures by direct melting of solid material and solidified from the melt have gradually a lower SiO_2 content and higher values of bulk density with an increasing thermal function; taking into account their thermal history, the situation in moldavites is reversed.



FIGS. 2 and 3: FIG. 2 (left). Weight percentage of the major components in glasses obtained from the sandy-silty clay, Besednice, Czechoslovakia, in dependence on values of the thermal function, *A*. FIG. 3 (right). Weight percentage of SiO_2 in glasses obtained by melting of the sandy-silty clay from Besednice plotted against their bulk densities; the thermal function and temperature are shown for each point.

These conclusions are also supported by partial chemical analyses of the silicate material that evaporated during the melting of sandy-silty clay and condensed on the cooler lid of the furnace. Data in Table I demonstrate that the condensed material is substantially enriched in SiO_2 and total Fe_2O_3.

All the data hitherto obtained indicate that in moldavites the evaporated and condensed material prevails over the material that passed directly from a solid state to a liquid one. The present experimental evidence requires that the chemical composition of moldavites and other tektites should not be compared directly with the chemical composition of their probable terrestrial source rocks. Therefore, very few conclusions can be drawn on the basis of compositional similarities as to what rocks were the source material (cf. Adams *et al.*, 1959; Heide, 1961; Pinson and Schnetzler, 1962; Schnetzler and Pinson, 1963; Taylor and Sachs, 1964; Greenland and Lovering, 1965; O'Keefe, 1966; Taylor, 1966; Bouška, 1968). The evaporation of the silicate material changed the chemical composition of the remaining impact glass, which consequently differs from that of the source rocks and that of the tektites formed dominantly by condensation of evaporated material strongly enriched in the sum SiO_2+total Fe_2O_3+Na_2O. The discrepancy emphasized by some authors between the chemical composition of the impact glasses and tektites assumed to be genetically related to them can now be better understood. The enrichment of silicate vapours in silica, total Fe_2O_3, and Na_2O and the impoverishment in calcium oxide, MgO, K_2O, and Al_2O_3 taking place during thermal shock can attain such a degree that the chemical composition of the glass condensed from the hot silicate vapour differs substantially from the chemical composition of the original material and from the glass generated from it by direct melting. Judging from the sizes and depths of the well-known impact craters (cf. Ries crater) regarded as a source area of some tektites it can be assumed that a complex of several types of rocks was the source material rather than only one rock type, and these source rocks were substantially poorer in SiO_2+total Fe_2O_3+Na_2O and richer in K_2O+CaO+MgO+Al_2O_3 than the tektites formed from them.

The conclusions presented in this paper can be applied to the study of other tektites and impact glasses found on the Earth and of the glasses produced by direct remelting or by evaporation and condensation of silicate material on the Moon.

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