Olivine crystal segregations in lunar glass with the composition of pyroxene

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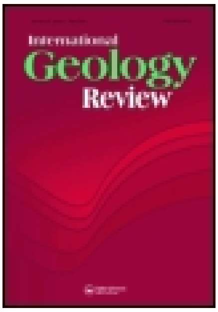

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INHOMOGENEITY IN NATURAL GLASSES

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INHOMOGENEITY IN NATURAL GLASSES

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Translated from "O neodnorodnosti prirodnykh stekol," *Izvestiya AN SSSR, seriya geologicheskaya,* 1988, No. 4, pp. 39-46. The authors are with the Institute of the Geology, Petrography, Mineralogy, and Geochemistry [IGEM], USSR Academy of Sciences, Moscow. They have studied volcanic and impact-generated glasses from both the Earth and the Moon and find inhomogeneities that reflect processes occurring in the superliquidus melts rather than during cooling.

Usually, natural melts are equated with solutions. Processes in magmas, particularly crystallization, are thus considered from the viewpoint of true solutions, but this concept conflicts with recent data on the structure of glasses and melts. Artificial glasses are inhomogeneous. Such techniques as small-angle scattering, IR spectroscopy, and Mössbauer spectroscopy, have shown submicroscopic heterogeneity of both phase and fluctuation types in glasses [14]. Liquids in artificial glass-forming systems also show such submicroscopic inhomogeneity, and inhomogeneity has also been found in some natural glasses [1, 5, 7-9].

We have made systematic studies on various natural glasses by transmission and scanning electron microscopy; the transmission microscope was a Tesla BS-500 (made in Czechoslovakia), or a JEM-100C (Japan), the SEM was a Philips 501-B (Netherlands) or a Hitachi S-450 (Japan). Various electron-microscope methods were used to obtain fuller information on the structure and homogeneity: suspensions, replicas, and replicas with extraction. The inclusions were identified by microdiffraction and microprobe analysis with a Kevex 5100 attachment to the scanning electron microscope. The compositions were calculated for the individual particles with a resolution of 200-300 Å by means of the Vostok package [6] with a Canon CX-1 microcomputer. The following glasses were examined.

Terrestrial Glasses

Mafic eruptive glasses

Glasses from pillow basalts from the Hess depression, Pacific. These Cenozoic glasses have been dredged from a depth of 4600 m. They form thin (0.5 cm) crusts around the pillows, which are up to 0.5 m across. There is a gradual transition from the rim to the relatively crystallized basalt in the central zone. The glasses are hard and black with a blue tinge. They are formed by the quenching at depth of basalt lava disintegrating into spheres during submarine eruption.

Alkali-basalt glasses from the Shavaryyn Tsaram ash cone, Mongolia. The glasses are composed of bomb fragments up to 30-50 cm in diameter. The glasses contain lherzolite xenoliths and occasional megacrysts of clinopyroxene, sanidine, garnet, and apatite. The glasses are hard and black and evidently formed by rapid cooling in the atmosphere of small portions of magma ejected in volcanic eruptions.

Subalkaline basalt glasses from the Tolbachik volcanic eruption 1975-1976. These glasses were collected on the southern cone, where we examined glasses from slag fragments in the tephra (size 1-3 cm, black and highly porous slags) and glasses from the lava flow crusts. The lava temperature at the sources of the lava streams in this eruption was 1050-1070°.

Acid eruptive glasses

Obsidian from the Arteni deposit, Armenia. The glasses constitute layers and zones in lava flows [7]; they include hard gray and dark brown fluidal varieties. It is assumed that the lava eruption temperatures were relatively low: 600-700°.

Cryptoexplosive glasses

Glasses from the Zhamanshin explosion structure, Kazakhstan. These have silica contents from 53 to 79 wt. %; there are also blocks of lechatelierite here. The glasses occur as individual particles and block bombs or fragments thereof. The glasses are white, yellow, black, blue, or green, and usually fluidal, and are characteristically porous. It is clear that the glasses were formed by cooling of small batches of coexisting liquids having various compositions. There is some evidence that the liquids were not highly superheated [12].

Lechatelierite from Meteor Center, Arizona, USA (Bogatikov's collection). This was collected from the central extensively transformed zone. The lechatelierite is derived from the Coconino Sandstone. It is white porous pumice. The Arizona crater is considered a type example of an impact crater. High temperatures (2000-3000°) and pressures of several megabar were produced by the meteorite fall, which melted the rocks. The subsequent quenching produced glasses.

Tektites. We examined an indochinite from Vietnam and a moldavite from Czechoslovakia (collections by Florenskiy and Cilek). The indochinite is dark green, hard, with a few large pores. The moldavite is pale green and dense. Most researchers consider tektites as frozen fragments of liquids formed by the fall of giant meteorites [10].

Lunar Glass

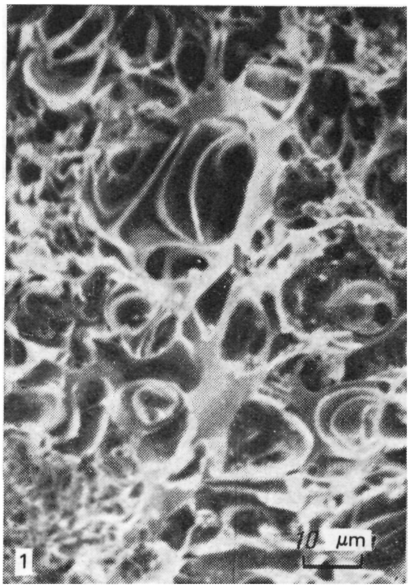
The lunar glasses include eruptive and impact ones; the impact glasses show inhomogeneities due to relicts of fused minerals. They also have high porosity. The eruptive glasses appear more homogeneous. We examined various lunar eruptive glasses: black glass from *Luna-24* [13], green glass [2], as well as endogenous spheres [3].

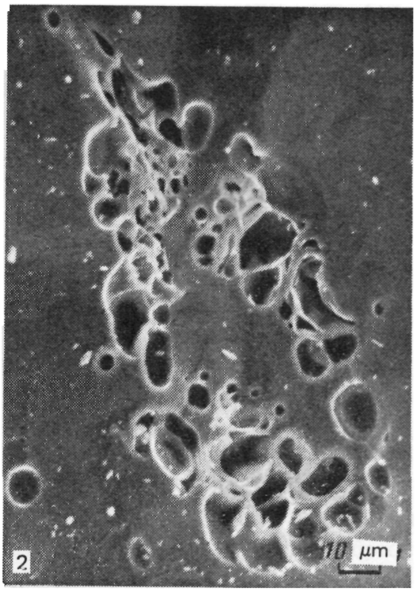
All the glasses appeared isotropic in the optical microscope. Fragments 0.1-2 mm in size were taken for electron microscopy. As the specimens were formed from liquids with various compositions and thermal histories, they clearly record various levels of ordering in the magmas.

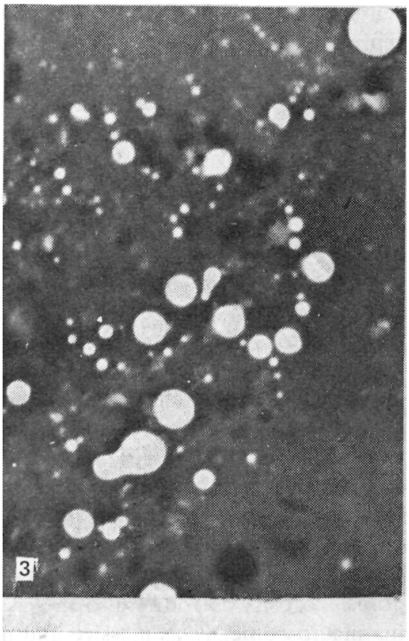
Electron Microscopy

We give some details of the microstructures characterizing the inhomogeneity.

Porosity. All the natural glasses were porous at the submicron level, although to various extents; the highest porosity was found in the tephra, glasses formed from a magma in contact with the atmosphere. The pores in these vary in size and occur in variable numbers even in parts of centimeter size. In part, they form linear systems of linked cavities. High porosity occurs also in the melted glasses, particularly lechatelierite from the Arizona crater (Fig. 1). The pores here vary in size, with ones similar in size often associated in patches up to several mm in size. The smallest pores have sizes of a fraction of a micron. Even more irregular distributions occur in the Zhamanshin glasses, where there are very porous patches with rounded or oval pores from a few tens of microns up to fractions of a millimeter (Fig. 2). These patches sometimes consist entirely of micron-size pores. However, in some cases the central part of such an area consists of monolithic glass. Around these porous areas, the glass contains isolated pores. Porosity is found also in glasses appearing macroscopically as massive and monolithic. In particular, single irregularly distributed pores up to 0.1 mm in size occur in glasses from oceanic basalts [15]. The dense glass from a Mongolian alkali basalt also contained pores, in which the surfaces were uneven. Pores occur also in obsidian and in the glasses from the Tolbachik lava flow crust, as well as in tektites. Porosity occurs to various extents also in the lunar glasses. As a rule, any glass showing signs of impact origin has relatively high porosity







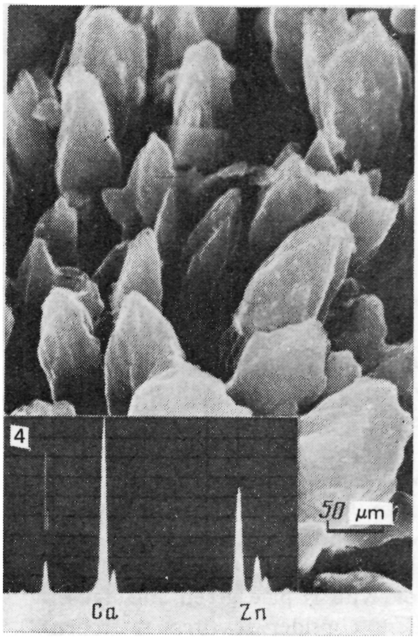
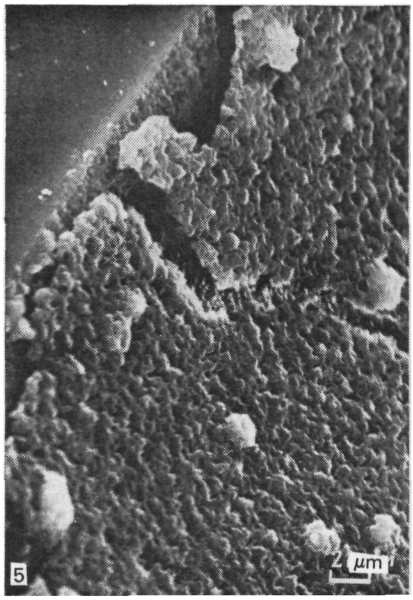


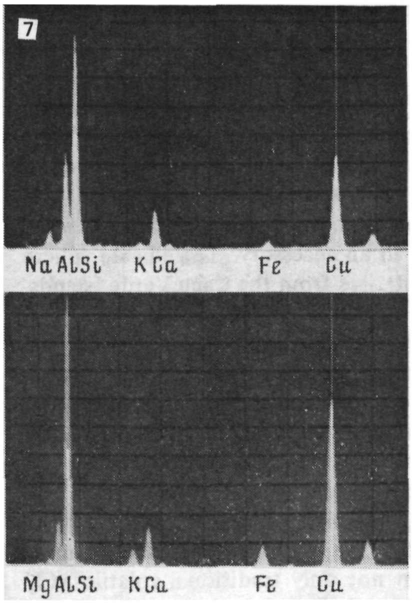
FIGURE 1. Contact between large-pore and small-pore areas in lechatelierite from Meteor Crater, Arizona (SEM, ×1000).

FIGURE 2. Porous area of oval shape, in zhamanshinite (SEM, ×700).

FIGURE 3. Porosity in lunar impact glass; light-colored droplets are meteorite material (SEM "compo" picture, ×720).

FIGURE 4. Pyramidal aragonite crystals on walls of pores in zhamanshinite (SEM, ×200). The inset shows the composition (elements lighter than Na not recorded).





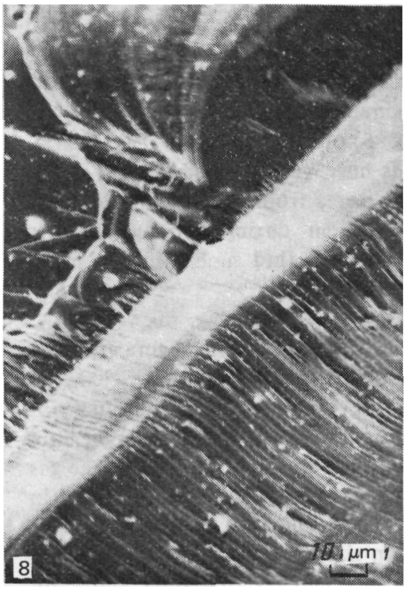


FIGURE 5. Silicate condensation rim in zhamanshinite pore. Pale droplet segregations enriched in sulfur are seen (SEM, ×3500).

FIGURE 6. Spectra showing compositions for two adjacent microareas in green Zhamanshin glass, *Kevex* 5100 microprobe attachment.

FIGURE 7. Microscopic inhomogeneities as rounded segregations in blue Zhamanshin glass (TEM, ×56,000).

FIGURE 8. Regular submicroscopic banding in obsidian (SEM, ×600).

(Fig. 3). Rare pores occur in the black and green lunar volcanic glasses.

Usually, porosity is found in all these natural glasses; one gets high porosity in tuffs, with the pores often having minerals containing volatiles. Cavities of various sizes have been described in spheres from rhyolites [10]. We have observed micropores in an accessory glass silicate sphere from carbonatites from the Cape Verde Islands.

The porosity depends on the presence of volatiles, and this in some cases is confirmed by some of the pores containing minerals bearing such components. In particular, eruptive and cryptoexplosive terrestrial glasses have been found to contain amphibole, layer silicates, sylvine, halite, basanite, anhydrite, sulfides, and Ca carbonates (Fig. 4) [5]. Most of these minerals contain not only traditional volatiles (Cl, H_2O, and CO_2) but also elements not assigned to that category (Ca, Si, Fe, Na, and K). In other cases, the pores contain not only minerals bearing volatiles but also condensation deposits of a silicate material similar in composition to the glass (Fig. 5) or more siliceous [8]. Clearly, these minerals have been formed from material present in the pores from the start rather than diffusing into them during cooling, because otherwise we would find such minerals in all pores.

The volatile elements in the magma are combined with nonvolatile rock-forming elements to give volatile assemblages; such combinations give rise to submicroscopic and then larger bubbles as the solubility in the magma falls. The gas phase condenses to give the minerals found in the bubbles. The bubbles are unevenly distributed and vary in size even in small volumes (for such volumes, one can say that the P and T are the same at any point during glass formation) which shows that the volatile contents vary even at the micron level.

Submicroscopic silicate inhomogeneities. All these natural glasses were inhomogeneous at the micron level; adjacent parts examined by means of attachments to the microscopes showed spectra varying in intensity for many elements (usually Si, Na, and K, but also for Mg, Al, Ca, and Fe, Fig. 6). The inhomogeneity is usually of fluctuation type, since the glasses retain phase homogeneity. Table 1 gives the largest composition variations for some of these glasses.

Auger spectroscopy has shown that the Tolbachik and lunar glasses have microregions 15-40 μm in size in which the oxidized forms of metals are also accompanied by reduced ones, even the native metal [4]. The overall compositions in such regions correspond to that of the associated glass, while the regions have a mosaic distribution. There are also amorphous segregations (size 0.3-3 μm), which have been observed in the Zhamanshin glasses and in tektites, as well as in pillow basalt and alkali-basalt glasses (Fig. 7). These segregations are nearly spheroidal and have compositions differing somewhat from the surrounding glass. Table 1 gives analyses for the surrounding glass and inclusions (which may contain some surrounding glass) for the moldavites and zhamanshinites. Such segregations in the zhamanshinites contain amorphous silica inclusions. Similar inclusions have been described for obsidians [1].

Inhomogeneities also occur in the lunar black glasses [14], particularly as seen in "compo" images, where the linear orientations for the inclusions differed.

Submicroscopic mineral phases. These segregations occurred in most of the glasses and resembled the other inhomogeneities in having sizes of 0.3 μm. The phases were mainly ones corresponding to the crysallline analogs for the glasses, in particular pyroxene and plagioclase for the Tolbachik glasses, or quartz and coesite for the Arizona lechatelierite.

There were also submicroscopic phases not inherent in the system compositions. Quartz, for example, has been demonstrated by electron microdiffraction in alkali-basalt, pillow-basalt, and Tolbachik basalt glasses. The Tolbachik tephra also contained potash feldspar and layer silicates (elemental composition Na, Al, Si, Ca, and Fe); obsidians contained layer silicates (elemental composition Mg, Al, Si, and Fe), clinochlore, and pyroxene. The forms of the latter are often rounded, and they occur richly in certain layers [7]. The pillow-lava

TABLE 1. Compositions of Terrestrial Glasses

	1	2	3	4	5	6	7	8	9	10*	11
SiO_2	100	100	79.04	74.95	75.97	66.79	73.35	68.13	74.25	72.64	82.51
TiO_2	–	–	0.42	0.41	0.47	0.77	1.01	0.98	0.68	0.70	0.28
Al_2O_3	–	–	9.70	2.69	9.93	17.64	17.07	15.94	11.61	13.15	9.25
FeO	–	–	3.15	4.79	4.08	5.89	4.07	6.06	5.23	5.21	1.46
MnO	–	–	0.22	–	–	0.34	0.07	0.06	–	0.14	–
MgO	–	–	0.47	1.22	1.42	2.46	0.63	1.61	3.09	3.05	1.22
CaO	–	–	3.36	6.33	5.80	0.75	0.58	1.77	1.99	1.72	1.87
Na_2O	–	–	1.30	0.31	0.72	1.11	1.29	0.96	1.03	1.01	–
K_2O	–	–	2.34	1.74	1.59	3.98	0.58	4.44	2.13	2.18	3.41
P_2O_5	–	–	–	–	–	–	–	–	–	–	–
SO_3	–	–	–	–	–	–	–	–	–	–	–
Cl	–	–	–	–	–	–	–	–	–	–	–

	12*	13	14	15	16*	17	18*	19	20*	21	22*
SiO_2	79.46	71.42	9.10	78.81	69.82	50.73	49.48	52.06	49.73	53.90	53.85
TiO_2	0.25	0.93	–	0.14	0.02	1.80	1.97	1.87	2.37	2.50	1.55
Al_2O_3	9.77	17.50	0.69	12.76	11.96	13.38	15.10	16.20	16.80	15.40	13.31
FeO	1.47	1.96	0.77	0.89	0.37	10.71	10.64	7.81	9.33	11.00	10.33
MnO	0.08	0.18	–	0.08	0.06	0.09	0.17	0.27	0.14	0.22	0.23
MgO	1.77	1.07	2.28	–	–	7.16	7.73	3.88	3.02	3.95	5.74
CaO	2.59	0.75	11.59	–	0.57	11.31	11.25	5.35	5.58	7.02	8.80
Na_2O	0.03	1.54	25.54	2.77	2.12	4.43	3.11	7.71	6.59	3.44	4.37
K_2O	3.55	1.89	6.45	4.55	4.39	0.29	0.17	4.85	5.49	2.88	1.83
P_2O_5	–	0.05	5.14	–	–	–	–	–	–	–	–
SO_3	–	0.03	3.98	–	–	–	–	–	–	–	–
Cl	–	0.04	28.40	–	–	–	–	–	–	–	–

Analyses: 1 and 2) lechatelierite: 1) from Arizona crater, 2) from Zhamanshin crater; 3-5) blue glass from Zhamanshin crater: 3 and 4) massive glass, 5) sphere containing a little of surrounding glass; 6 and 7) yellow glass from Zhamanshin crater; 8) green glass from Zhamanshin crater; 9-14) tektites: 9 and 10*) indochinites, 11, 12* and 13) massive moldavites, 14) sphere containing a little of the matrix; 15 and 16*) obsidian (Armenia); 17 and 18*) pillow basalt (Hess depression); 19 and 20*) alkali basalt (Mongolia); 21) basalt tephra (Tolbachik volcano); 22*) run in lava (Tolbachik volcano).

*Microprobe analyses (analyst G. N. Muravitskaya, this institute), others in electron microscope with *Kevex* attachment, –) not observed.

glasses contain a K-bearing mica and α-Fe. Such phases do not occur in the crystalline analogs of these glasses, and they are evidently metastable, so they represent an early superliquidus stage.

Fabric inhomogeneity. There is also submicroscopic banding in obsidians, tektites, zhamanshinites, and black lunar glasses [13]. This banding is regular and has a step of 1-10 μm (Fig. 8), the configuration resembling parting in minerals. It evidently reflects ordering.

Discussion

Natural glasses with various compositions and histories show submicroscopic inhomogeneities of fluctuation and phase types, which are indicated by various features and by the contents and distributions of rock-forming components, volatiles, and submicroscopic phases. These inhomogeneities probably were not produced at the time of glass formation but at an earlier superliquidus stage. Laboratory experiments and observations on volcanic eruptions show that the viscosity increases rapidly on cooling, which retards diffusion and gives rise to inhomogeneities. Also, it is impossible for metastable phases to form in the subsolidus region.

These microscopic inhomogeneities show that the magmas are polydisperse systems; the dispersion gives rise to multicomponent compositions with rapid cooling. It is currently thought that interactions in the early cooling stages bind the particles into relatively stable submicroscopic groups (clusters, minals, cybotaxes, etc.). The structures and the cation compositions do not remain constant; they approach in composition and structure to the phases (crystalline and liquid) corresponding to the given system. The liquid may evolve, with the compositions and sizes of the groups changing, together with the structural ordering, or aggregation may occur. A cooling magma is likely to show aggregation as the path favored by energy, because the total energy is reduced on account of the change in surface energy. Inhomogeneities thus arise, manifested as dispersed segregations of various sizes.

The composition and structure in the liquid change as the atomic groups change, and the features approach those of the stable phases, which is evidently related to the presence in the glasses of metastable phases, such as quartz in tholeiitic pillow basalt or alkali basalt glasses, or layer silicates in obsidians and basalts, or pyroxenes in obsidians. The glasses contain frozen groups and assemblages arising at relatively early stages.

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