

## Formation of the hour-glass structure in augite

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**SUMMARY.** A study of augite in over three hundred thin sections of mainly alkalic rocks permits the distinction of two main types of hour-glass structure. The common 'swallow-tailed', sometimes skeletal augite crystals are found in the fine-grained groundmass of many rock types, and it is suggested that rapid crystallization alone accounts for their formation. Hence, this type of hour-glass structure has been called 'quench hour-glass'. The hour-glass structures of larger augite crystals of porphyritic and coarse-grained rocks are commonly described as hour-glass 'zoning', as they result primarily from compositional differences between the different sectors. These were formed under conditions of relatively slower cooling than the 'quench hour-glass', and thus cannot be explained in the same way. They are thought to have formed by a process involving adsorption of impurities on a particular crystal face so as to impede growth of these faces, producing an initial skeleton of hour-glass form, which is completed by later crystallization of augite richer in FeO, Na<sub>2</sub>O, TiO<sub>2</sub>, and Al<sub>2</sub>O<sub>3</sub>. This hypothesis also explains the patchy zoning of other augite crystals, casting doubt on some petrogenetic interpretations of such zones as core zones.

THE present study developed from a more general investigation of volcanic rocks of the Comores Archipelago, rocks that are generally rich in clinopyroxene showing numerous striking examples of the hour-glass structure. Although the geological literature contains no generally satisfactory explanation of how this feature forms, numerous studies of its formation under ideal experimental conditions have been described. These are not readily applicable to geological phenomena, however, and it thus seemed desirable to investigate their possible application to crystals of augite formed in natural systems of great physico-chemical complexity.

Although a true hour-glass may be thought of as two cones that intersect at their apices, the 'hour-glass' in augite is formed by the intersection of pyramids, each one identifiable by the crystal face that would form its base, whether this face is developed or not. Two-dimensional views of these pyramids, as in thin sections, may be triangular, rectangular, etc., and are commonly called sectors. As the sequence of development of pyramids is important in discussing the origin of the hour-glass, a principle axis, H, will be designated as connecting the set of pyramids thought to have been last-formed. The crystallographic orientation of H can then be used for comparison of the hour-glass structure in different crystals.

### *Previous work on the augite hour-glass structure*

Rosenbusch (1905) concluded that the augite hour-glass structure resulted from growth of an initial hour-glass skeleton, which was later completed by infilling of the hollow (100) sectors, i.e., with H parallel to the *a*-axis. Scott (1914) studied optical properties of the different sectors and accepted Gaubert's (1911) explanation of the structure as resulting from selective adsorption and incorporation of impurities on particular faces of the crystal during its growth. Although Scott thought this a

rejection of Rosenbusch's explanation, the two are not incompatible, and compositions determined from Scott's data (using the curves given by Deer *et al.*, 1963, fig. 41, p. 132) suggest that the (100) sectors were formed last, i.e. that H is parallel to the *a*-axis. Wilkinson's (1957) optical data, though for different sectors of the hour-glass, are not related to crystallographic orientation. However, taking the long axis of his crystals as the *c*-axis, and using data given in his table 1 (p. 124) to obtain compositions of the different sectors, it is suggested that H is parallel to the *c*-axis. Farquhar (1960) summarized ten possible hypotheses to explain formation of the hour-glass structure, and these will be discussed below. Preston (1966), in his study of 'An unusual hourglass structure in augite', showed that this structure was made up of parallel triangular prisms, rather than intersecting pyramids, and differences in composition between sectors suggested that the (010) sectors were last-formed, H being parallel to the *b*-axis.

The studies reviewed above thus show that the hour-glass structure in augite has no consistent crystallographic orientation, i.e. that the compositional trends show no consistent crystallographic preference. Furthermore, Buckley (1951) reviewed studies showing that the hour-glass could be formed in a variety of synthetic crystals by adsorption of dyes on particular crystal faces, both by their incorporation into the growing crystal and by interference with growth at the point of adsorption. And because different dyes are adsorbed on to different faces, different orientations of the hour-glass can be produced in otherwise similar crystals.

#### *Present study*

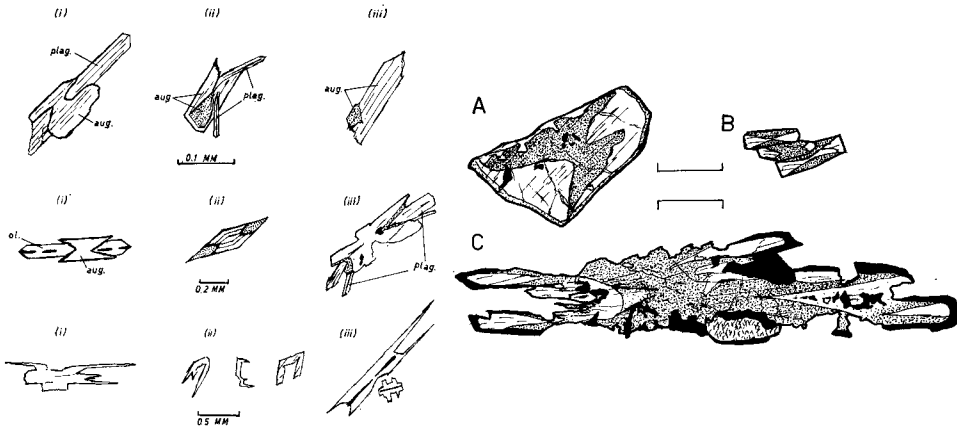
Approximately three hundred thin sections were studied to try and ascertain the possible effects that chemical environment or conditions of crystallization might have on the augite hour-glass structure. A reconnaissance study was made of thin sections in the teaching collection of the Grant Institute of Geology, Edinburgh, including both silica-saturated and undersaturated intrusive and extrusive rocks from Hawaii, Mauritius, Reunion, Japan, the Cascades, and the Shiant and Whin sills, as well as a collection from the Nuanetsi province and the author's collection from the Comores Islands. Although rare hour-glass structures were observed in ground-mass augite of the saturated rocks, they were found to be far more common in undersaturated types. The study consequently became confined to alkalic rocks, several examples of which were selected from the Comores Islands for detailed study of the hour-glass structure with the electron microprobe and on the universal stage.

The microprobe analyses were carried out at Durham University using a Cambridge Geoscan instrument, under the direction of Dr. C. H. Emeleus. Standards similar in composition to the unknowns were used, with correction being made for background and instrument drift. The error in this method is considered to be  $\pm 2$  % of the values obtained for CaO, MgO, and FeO,  $\pm 5$  % of TiO<sub>2</sub> values, and  $\pm 10$  % of the Na<sub>2</sub>O and Al<sub>2</sub>O<sub>3</sub> values (Emeleus, pers. comm.).

#### *Results*

The petrographic studies showed consistent differences between the hour-glass structures in groundmass augite of the rapidly cooled rocks and those seen in augite

phenocrysts and augites of more slowly cooled rocks. The groundmass augites, as in alkali olivine basalts (fig. 1A, B) or glassy tholeiites (fig. 1C), are less than a millimetre long and elongated parallel to the *c*-axis. The ends of these crystals are 'swallow-tailed' and show striking re-entrants commonly filled with groundmass glass or microlites (fig. 1A (ii), 1C), but may be overgrown with single crystals of olivine (fig. 1B (i)), plagioclase (fig. 1A (i)), or later augite (fig. 1A (ii, iii), B (ii)). When overgrown



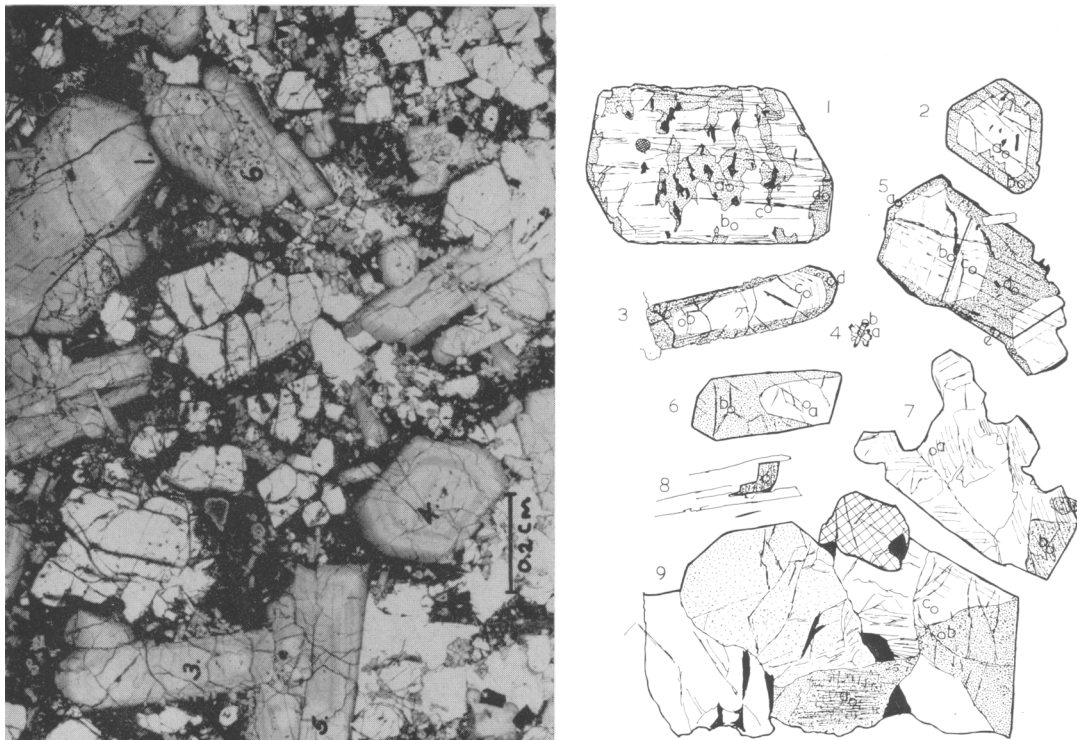
FIGS. 1 and 2: Fig. 1 (left). Quench hour-glass structures. A (top) (i), plagioclase infilling; A (ii), augite and plagioclase infilling; A (iii), patch of later augite filling gap on the side of an augite crystal; B (middle) (i), skeletal olivine overgrowth; B (ii) later augite overgrowth; B (iii), overgrowth of later augite with subsequent infilling with plagioclase crystals; C (bottom) (i) and (iii), longitudinal (parallel to *c*-axis) sections of skeletal augite with hollows filled with glass; C (ii) cross-section of similar crystals. A and B are from olivine basalts of the Comores Islands, and C is from glassy tholeiite of the Nuanetsi province. Stippled areas are the darker, and presumably  $\text{TiO}_2$ -rich parts of the crystal. Fig. 2 (right). Hour-glass structures in microphenocrysts of augite from olivine basalt (A and B), and in a large titanite crystal from ijolite (C), all from the Comores islands. Note incipient hour-glass development on the 'limbs' of the hour-glass in crystal C, also shown in fig. 5. Upper scale-bar 0.5 mm. lower 2 mm.

by the later augite, these re-entrants are similar to hour-glass structures of the larger crystals, except that boundaries between sectors are more abrupt and irregular (fig. 1B (ii, iii)). It is this similarity that led the author to consider all such 'hollow-ended' crystals a form of hour-glass structure, and these have certainly been described as such by many petrographers in the past. Thus, the re-entrants make up the (001) sectors, which are the last-formed, and this gives an orientation of H parallel to the *c*-axis. Because rapid crystallization is considered essential for their formation I have termed these 'quench hour-glass' structures.

Unlike the quench hour-glasses, however, those in augite of porphyritic and coarse-grained rocks do not display such irregular discontinuities at sector boundaries. The different sectors appear to result from compositional differences only, as reflected in differences in optical properties and electron microprobe analyses. These larger augite crystals are also typically elongated parallel to the *c*-axis, but less so than the groundmass augites. All such hour-glass structures observed in this study were found to consist of the pyramidal units as described by Scott (1914) rather than the prisms

described by Preston (1966), although the sector boundaries in these may be straight (fig. 2A) or curved (fig. 2B, C).

Study with the universal stage showed that large crystals display the hour-glass form best in sections more or less parallel to the  $c$ -axis (fig. 3, crystals 1 and 5). In



FIGS. 3 and 4: Fig. 3 (left). Different views of the hour-glass structure in augite phenocrysts in ankaramite from the Comores Islands. Crystals 1 and 5 are (010) sections, crystal 3 is cut approximately parallel to (110), crystals 4 and 6 are (001) sections. Fig. 4 (right). Crystals analysed with the electron microprobe. Numbered circles show the areas analysed, with corresponding data given in table I. Crystals 1 and 2 are from alkali picrite-basalt (GC-31); 3-5 from alkalic ankaramite (Mo. 30); 6 to 9 from plagioclase-olivine-augite phyric basalt (GC-30). All rocks from the Comores Islands.

sections widely oblique to  $c$ , only one sector of the hour-glass can be seen (fig. 4, crystal 6). Sections perpendicular to  $c$  show what must be the base of the major hour-glass pyramids, as the hour-glass form is not seen at all (fig. 3, crystals 4 and 6).

The (001) sectors of these crystals are consistently lighter-coloured than the (010) and (100) sectors, except for an outer rim that envelops the whole crystal. The dark (purple-brown) colour of the (010) and (100) sectors is similar to that of the crystal rims and groundmass augites, as well as the patchy zones of other crystals.

Results of the microprobe analyses are shown in table I, in which the number of each analysis corresponds to the spots similarly numbered on the crystals shown in fig. 4. These results show that the darker parts of the crystal, whether in the

hour-glass sectors, in patchy zones, or in groundmass augite, are generally enriched in FeO, Na<sub>2</sub>O, TiO<sub>2</sub>, and Al<sub>2</sub>O<sub>3</sub> and depleted in MgO, with CaO remaining essentially constant.

### Discussion

The experimental studies reviewed by Buckley (1951) were concerned with crystallization at low temperatures in simple aqueous systems, and are thus not readily applicable to the augite hour-glass structure. Of the ten hypotheses summarized by

TABLE I. *Electron microprobe analyses of augite crystals shown in fig. 4*

	Al <sub>2</sub> O <sub>3</sub>	FeO	CaO	MgO	Na <sub>2</sub> O	TiO <sub>2</sub>		Al <sub>2</sub> O <sub>3</sub>	FeO	CaO	MgO	Na <sub>2</sub> O	TiO <sub>2</sub>
GC. 31							Mo. 30						
1a	3.1	5.2	22.5	16.1	2.6	1.6	4b	5.0	7.9	22.3	12.8	3.0	2.9
1b	3.4	4.9	22.5	16.7	0.9	0.8	5a	5.8	6.4	22.3	12.6	3.0	3.1
1c	—	4.5	—	17.3	1.0	0.9	5b	—	6.5	22.2	13.4	1.6	—
1d	—	6.2	—	15.9	2.0	1.6	5c	—	6.2	22.4	15.1	1.6	—
2a	3.3	4.5	22.2	17.7	0.8	0.7	5d	5.5	6.2	22.1	13.9	1.9	2.5
2b	4.2	6.0	21.9	16.1	1.4	1.8	5e	—	6.3	22.4	14.8	1.6	—
gm*	3.9	5.4	21.7	17.1	1.1	1.3	gm*	7.1	6.4	22.3	14.2	3.3	2.5
gm*	—	5.4	—	13.5	0.1	1.0	GC. 30						
Mo. 30							6a	—	7.8	22.0	12.7	—	3.1
3a	—	5.8	—	14.4	—	—	6b	—	7.5	21.7	13.8	—	2.4
3b	—	5.8	—	14.8	—	—	7a	—	5.8	21.8	14.0	—	1.6
3c	—	6.0	—	14.8	—	—	7b	—	7.6	19.5	13.2	—	2.0
3d	—	7.6	—	12.9	—	—	8	—	8.0	21.7	12.2	—	3.5
4a	4.8	6.2	—	14.1	—	2.1	9a	—	7.1	18.2	11.9	—	1.6
							9b	—	7.4	21.2	12.0	—	2.7
							9c	—	7.4	21.5	13.8	—	1.8

\* gm = groundmass crystal.

Farquhar (1960), several are little more than rephrasing of the same process (thus, 'difference in molecular attraction', 'selective ionic adsorption', 'precipitation of different chemical substances', and [selective] 'staining' are essentially the same thing), some fail to account for compositional differences between sectors, and some are really descriptive statements rather than explanatory hypotheses. There are actually only three distinct hypotheses to consider, namely Farquhar's *a*, *c*, and *d*.

The question of resorption of crystals (Farquhar's hypothesis *c*) has been of more general interest than the hour-glass structure to petrographers, and has received detailed study by several workers. In a study of skeletal olivine crystals, Drever and Johnston (1957) showed that many of the features previously attributed to resorption could be produced by rapid crystallization or quenching. However, Blackerby (1968) has shown that similar features ('embayments') in plagioclase crystals can also be produced during slow crystallization. He interprets these embayments as being caused by adsorption of viscous liquid to parts of the crystal surface. This liquid, by acting as a 'semipermeable membrane' slows the rate of diffusion to and subsequent growth

at the points of adsorption. Although the continuation of zoning around these embayments is convincing evidence for their being produced during growth of the crystal (as opposed to resorption), the present author hesitates to accept viscous liquid as the adsorbed material. As discussed by Mullin (1961), the heat of crystallization causes the temperature at a crystal face to be slightly higher than that of the surrounding liquid, and this should cause liquid at the surface to have slightly lower viscosity rather than higher. Some other adsorbed material might thus be responsible for these features.

Similar 'embayments' have been observed in augite crystals in the present study. These have compositional zoning that continues around the 'embayments', which in many cases are filled with later augite (fig. 2C, fig. 4 crystal 1, and fig. 5). It can be seen that such 'embayments' are nothing other than a form of the hour-glass structure (figs. 2C and 5) or patchy zoning (fig. 4, crystal 1).

Following Blackerby's reasoning, resorption is not likely to have produced the hour-glass structures or patchy zoning, and we are left with Farquhar's hypotheses *a* and *d*, essentially those discussed by Scott (1914).



FIG. 5. Large augite crystal in ijolite from the Comores Islands.  $\times 5$ . To be compared with the sketch in fig. 3C.

*Type 1. Quench hour-glass.* Skeletal crystals are an established result of rapid crystallization from the magma (e.g. Wyllie *et al.*, 1962; Drever and Johnston, 1957), and there is no doubt that the shapes of groundmass augite shown in fig. 1 can be explained in this way. Ideally, therefore, such crystals should merely be called skeletal augites. However, when these skeletal augites are overgrown and infilled with later augite (e.g. fig. 1, crystals A (ii, iii) and B (ii, iii), and fig. 2, crystal A), they must be described as having an hour-glass structure, and have thus been named quench hour-glass in this study.

*Type 2. Hour-glass structures of slowly formed augites.* In contrast to the quench hour-glass structures, those in larger crystals cannot be accounted for by rapid crystallization, and Farquhar's hypotheses *a* and *d* may be considered. The microprobe analyses (table I) help to establish hypothesis *a* as follows.

Because the chemical differences between sectors encompass more than one element, one cannot assume that the hour-glass results from adsorption of only one ion on a particular face (e.g. titanium ions, Heinrich, 1965, p. 216). Indeed, the chemical differences are just what one would expect to find between two pyroxenes formed at different times in a crystallization series. For two such pyroxenes to have formed simultaneously requires that a miscibility gap exists. This is unlikely, however, because

the 'gap' does not occur at the same compositions in different crystals, and it is not supported by any other natural data (Deer *et al.*, 1963) or experimental data (see, e.g., Yagi, 1967). One must therefore accept that different sectors formed at different times, i.e. that Farquhar's hypothesis *a* is correct. As the enrichment in FeO, Na<sub>2</sub>O, and TiO<sub>2</sub> has taken place in the later stages of crystallization, i.e. phenocryst rims and groundmass augite, the (010) and (100) sectors of these hour-glass structures presumably also formed during the later stages. Consequently, H lies in the plane (001), probably parallel to the *a* or *b* crystal axes, and the initial hour-glass skeleton consisted of the (001) sectors. Thus, two more orientations of H may be added to those of previous workers.

Preston (1966) postulated that, because increase in Ca content enlarges the *a*-axis but has little effect on the *b*-axis, the Ca may 'prefer' an ordered orientation on a growing face whose spacing it more readily determines, and is thus enriched over Fe in the *a*-direction. This implies not only that all sectors grew simultaneously, a possibility dismissed above, but that such a process should operate under all conditions of formation, since the effects of Ca on the lattice do not change. Yet the compositional differences observed by Scott, Wilkinson, Preston, and the present author show no consistent crystallographic preference, making Preston's explanation unacceptable.

If we accept that the hour-glass sectors formed at different times, we must explain why the initial skeleton forms, without invoking rate of crystallization or control by the structure of the crystal. The only other reasonable variable to be considered is the chemical environment, i.e. an external influence as opposed to controls within the crystal itself. This leads to the theories of adsorption reviewed by Buckley, and the present author suggests that impurities in the magma produce the hour-glass structure by adsorption on to the crystal surface and interfering with growth at the points of adsorption, which would have been on the (010) faces of Preston's crystals, the (100) faces of Scott's, possibly the (001) faces of those described by Wilkinson, and on both the (010) and (100) faces of the large crystals of the present study. It is not necessary that this adsorbed material be that presently in the sectors, nor that it is the same material in all cases. From the available experimental evidence (Buckley, 1951) one would expect that different materials might be responsible for the different orientations. Probably a variety of natural substances, from other mineral grains to particular ions, can have this effect. As the process of adsorption is sensitive to physico-chemical conditions, slight changes in the conditions of crystallization (e.g. movement of the crystal through inhomogeneous magma) would cause the adsorbed material to leave the surface, permitting filling of the sectors and further peripheral growth. Such fluctuations in the effective adsorption could thus explain the crossing of sector boundaries by compositional zoning (e.g. fig. 4, crystal 5).

Similar reasoning (i.e. adsorption of impurities to parts of the crystal surface) suggest that the patchy zones enriched in FeO, Na<sub>2</sub>O, and TiO<sub>2</sub> were also formed in the later stages of crystallization by filling of 'embayments', which were caused by adsorption of foreign material. These patchy zones must not therefore be mistaken for 'core zones' (e.g. see Frisch and Schminke, 1968; Wilkinson, 1957), an error that would produce misleading theories of petrogenesis.

*Conclusion*

The variety of crystallographic orientations and chemical trends in the hour-glass structures of augite, observed by previous workers and the present author, strongly suggest that their formation is not controlled by the crystal structure as suggested by Preston (1966). The same may be said about the patchy zoning of augite crystals. Eight of the ten hypotheses listed by Farquhar (1960) to explain formation of the hour-glass structure have been dismissed as a result of this study. The two hypotheses accepted, essentially those discussed by Scott (1914), have been combined to suggest that such features result from adsorption of unidentified impurities on particular crystal faces, impeding the growth of these faces, and producing an hour-glass-shaped skeleton, which is completed by later crystallization of augite of different composition. This conclusion is similar to those from experimental studies on other chemical compounds, and may apply equally well to other natural crystals.

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