CRYSTALLIZATION OF NEPHELITE FROM INDUSTRIAL GLASSES

By

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On melting a mixture of glass constituents of adequate composition, no crystallization of nephelite can occur. However, nephelite can crystallize from lime-soda glasses when alumina sporadically accumulates. The formation of nephelite stones is due to the action of a substance, of special microscopic structure in furnace linings, which delivers corundum crystals to molten glass. Crystals of corundum are, however, not present in refractories as original constituents, they develop as contact products in the surface layer of furnace linings facing the molten glass. Nephelite ($NaAlSiO_4$) appears in hexagonal pyramidal crystals (C_6^6). Hemimorphy was not observed on crystals which crystallized from molten industrial glasses. Fig. 1. shows crystals of symmetrical development.

Nephelite crystals become columnar. Only the faces (1010) and (0001) are clearly observable. Nephelite crystals forming stones often appear as an agglomeration of particles, i. e. of crystals of uniform orientation (Fig. 16). A resemblance to a pinetree branch often occurs, probably due to the stepwise growth of faces. The details of the faces of the crystal shown in Fig. 2 seem to have been developed, as a rule, quite independent from one another. Moreover, also negative faces (referring to the whole crystal) formed. Thus, in the direction of the principal axis, the crystal has a coat-like, spiral shape (Fig. 2).

An alternated growth of faces can also be observed in the crystal shown by Fig. 3. The stepwise development of the basal face is visible on the inside of the crystal as well, due to occlusions located, during the growth of crystals, parallel to the actually developed face portions (Fig. 3).

Crystal shapes resembling pinetree branches have developed stepwise, rhytmically repeated and parallel shifted faces. The stepwise, incomplete development does not occur along the line connecting opposite peaks. Thus the substance of crystals forms ribs along this line. In the case of occlusions, this area remains free from them (Figs. 4 and 5).

The occurrence of occlusions is characteristic of nephelite crystals which, therefore, sometimes show quite striking colours. Whereas the crystals shown in Fig. 1 are white, due to minute colourless occlusions, those in Figs. 2 and 3 macroscopically show an ochre yellow, and those in Fig. 4 a black colour.

The refraction indexes of nephelite $\omega = 1,537$, $\varepsilon = 1,533$ in general exceed, those of lime-soda glasses usually produced. Nephelite has a negative optical character. Under identical genetical conditions, also crystals of carnegieite may form stones besides nephelite, or independently of this latter.

Carnegieite is of an optically negative character, forms triclinic crystals with refraction indexes $a = 1,509$, $\gamma = 1,514$. It occurs in stones as fibre-like agglomerations sometimes showing twin formations. At the borders of agglomerations, crystals resembling feathers are to be found (Fig. 6).

Carnegieite often occurs with nephelite in the same stone. They can be distinguished from one another, on the basis that by an oblique illumination, they show different refractions, the nephelite of stronger refraction becomes shadowed and presents plastic shapes,
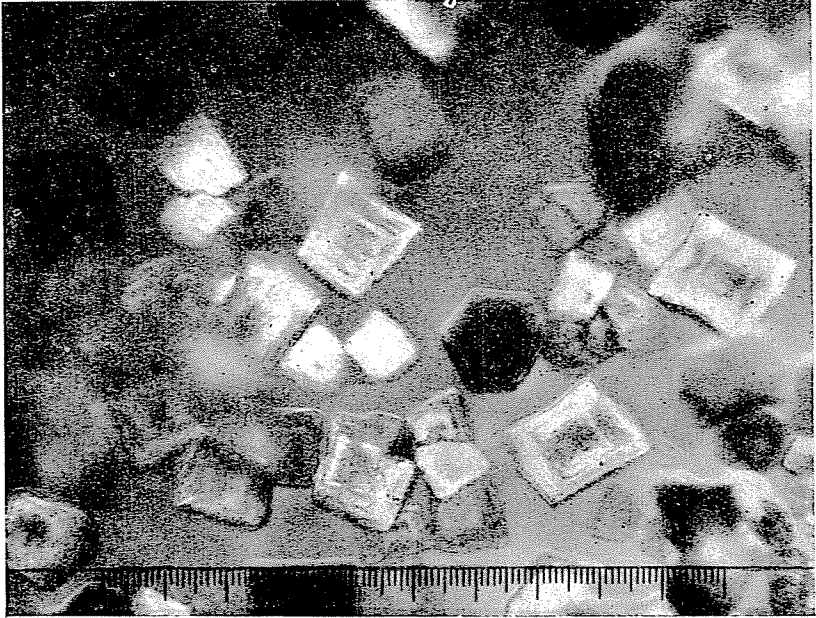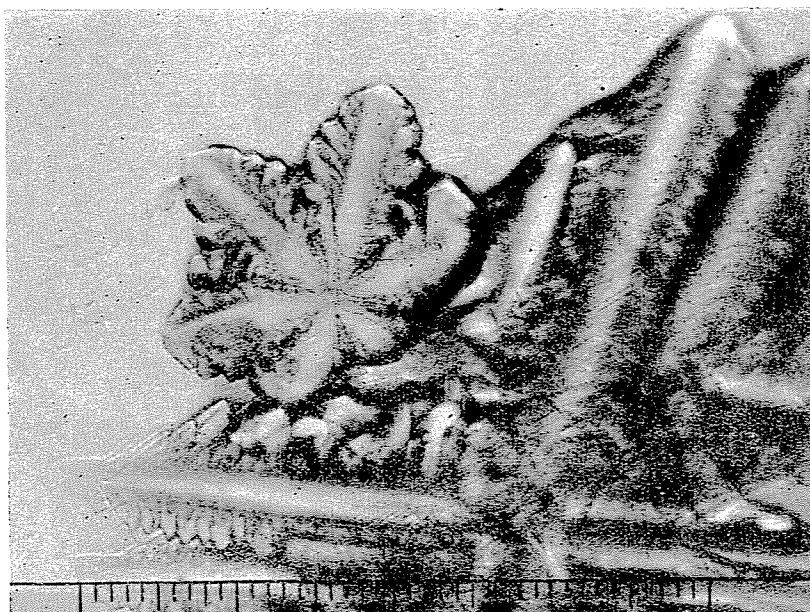
Fig. 1. Magnification 100×. Scale : 10 microns. Crystals of nephelite crystallized from the sediment in the recuperator of a tank furnace
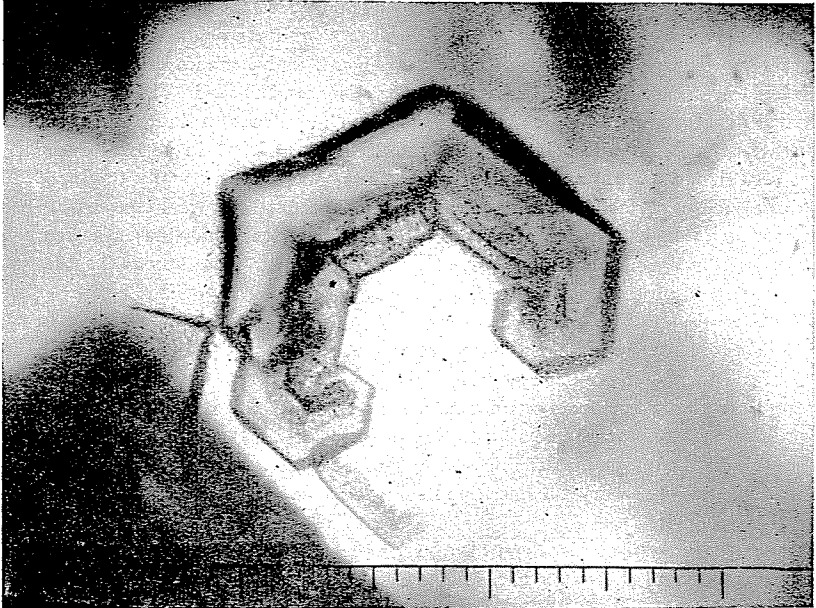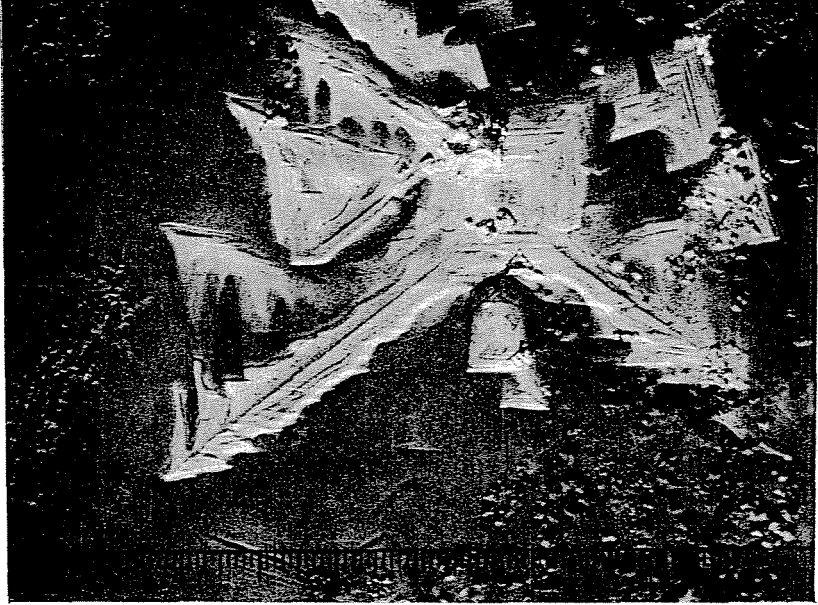

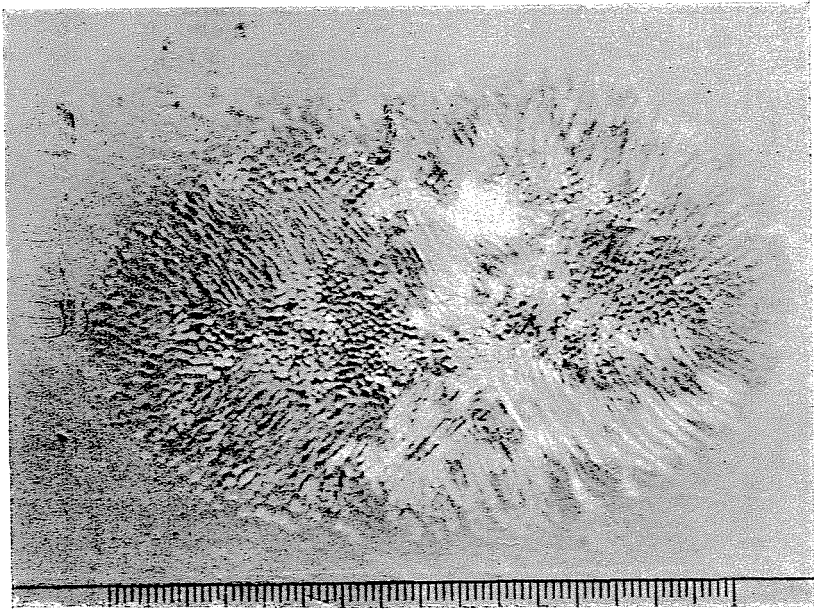*Fig. 2.* Magnification 300×. Scale 10 microns. Nephelite crystal of spiral development
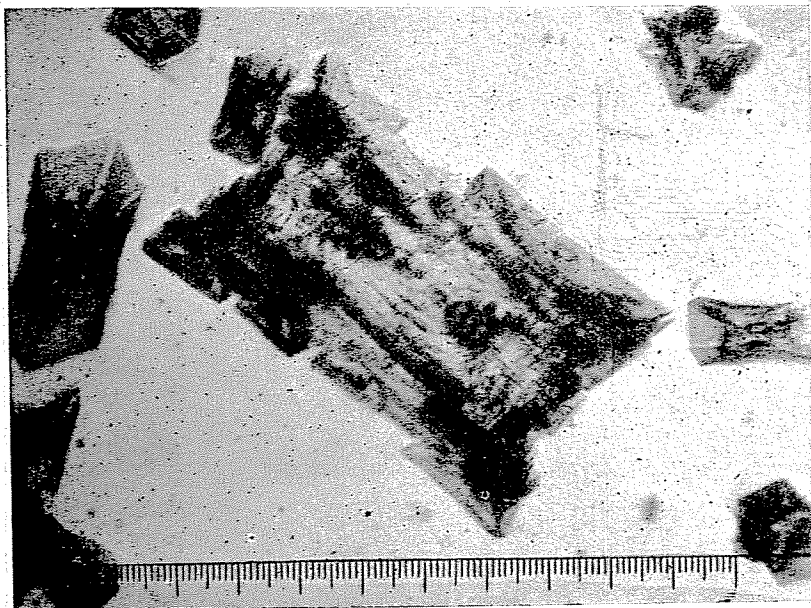
Fig. 3. Magnification 70×. Scale 10 microns. Nephelite crystal with faces developed stepwise, from precipitate of a recuperator from a tank furnace
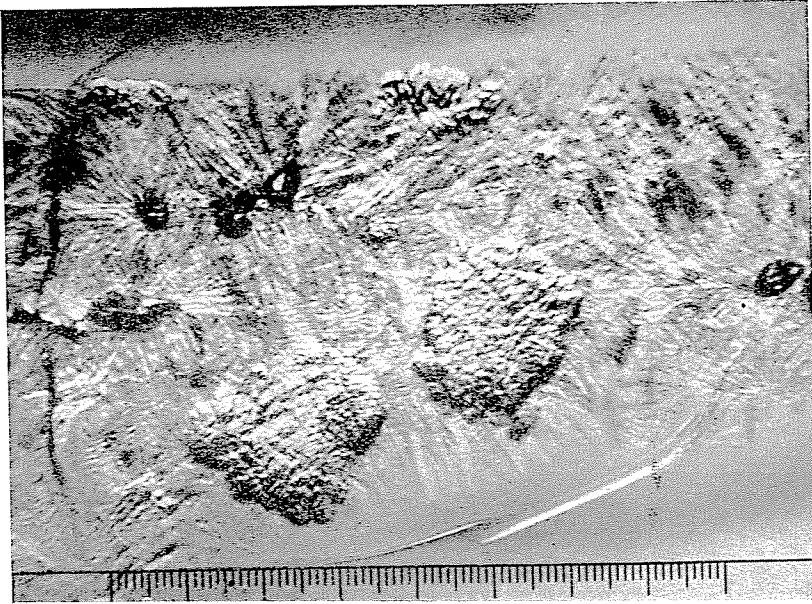

Fig. 4. Magnification 200×. Scale 10 microns. Nephelite crystals resembling pinetree branches. The principal axis of the upper crystal is perpendicular to the plane of the figure, whereas the lower crystal has its principal axis in the plane of the figure. The crystals developed in the contact surface of re-fractory and glass
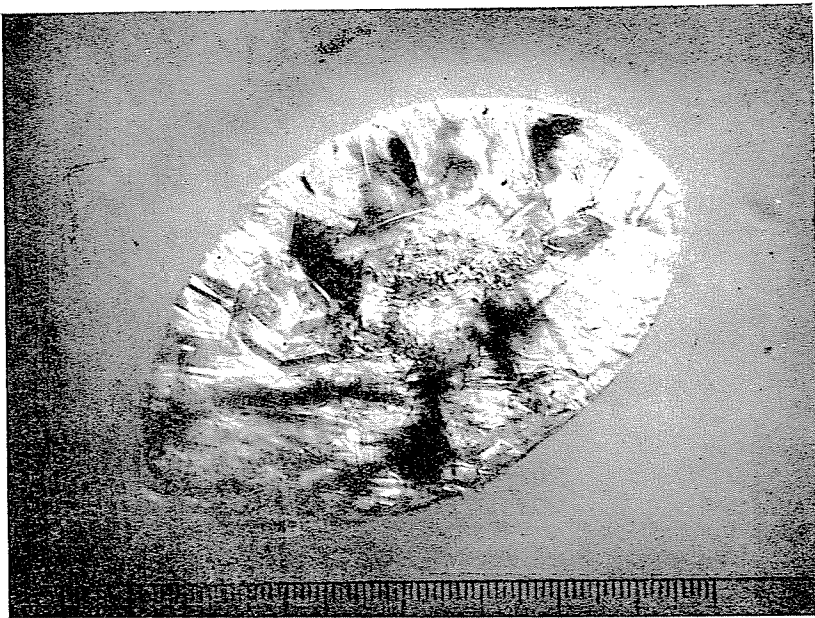
Fig. 5. Magnification 200×. Scale 10 microns. The light coloured crystal is nephelite, the small crystal agglomerations, of darker colour in turn, are sodalites. The principal axis of nephelite crystals lies in the plane of the figure. Nephelite crystals are poorly developed, only crystal substances appear, rather in the ribs, along the line connecting opposite peaks

Fig. 6. Magnification 500×. Scale 2 microns. Agglomeration of carnegieite crystals from lime-soda glass

Fig. 7. Magnification 100×. Scale 10 microns. Stone from lime-soda glass. In the centre, two larger agglomerations of grainy nephelite. The less shadowed fibrous agglomeration almost filling up the whole area, surrounded, mainly in the lower centre, by feather-like crystals, consists of carnegieite. The dark particles of 4—7 microns are grains of corundum rounded by dissolution

Fig. 8. Magnification 100×. Scale 10 microns. Nephelite stone, with mullite crystals in the centre

whereas carnegieite is almost indiscernible. This difference, however, disappears in polarized light, under crossed nicols, due to the contrast effect of extinction (Fig. 7).

Owing to the absence of adequate concentrations of alumina, nephelite or carnegieite do not crystallize from lime-soda glasses. The prerequisite for the crystallization of nephelite is the addition of a substance containing alumina, in such a form which causes, at least local accumulations of alumina in the molten glass mass. Thus, no nephelite can develop of adequate composition and particle size from a molten glass mass. Stones consisting of nephelite and carnegieite almost always include particles of corundum, rarely also those of mullite. Nephelite stones in lime-soda glasses are secondary precipitates, the formation of which is induced by a grain of corundum or of another substance rich in alumina. When this grain is dissolved more or less imperfectly, the adjacent glass mass is enriched in alumina. Nephelite or more rarely carnegieite crystallizes in the enriched layer. When the mass is further cooled, often also sodalite, hauyn may appear. Fig. 8 shows the microphotograph of a stone from nephelite, with dissolved crystals of mullite in the centre. The mullite crystals are presented by Fig. 9 in a higher magnification.

Rather well-developed nephelite crystals are shown by Fig. 10, including corundum platelets of a coarse surface (due to partial dissolution).

Refractories in contact with molten glass may lead to changes in the structure of the contact layer. Microscopic investigations of the changed structure may furnish information as to whether the different types of refractories deliver certain constituents into the molten glass, or not.

When in contact with molten glass fire-clays show two types of behaviour: while acid fire-clays deliver to the contact glass layer needle-like crystals of mullite and thus show the structural elements mullite + glass, in alkaline fire-clays (with higher alumina contents) these structural elements consist of mullite + glass + corundum. Fig. 11 shows the contact layer of acid fire-clays. Needle

crystals of mullite, which rarely intrude into the glass mass, were not present during melting, as they develop only during cooling. Single mullite crystals embedded in extremely viscous glass cannot get into the glass mass. The contact layer of substance of this structure slowly disappears, due to a simple dissolution.

Fig. 12 presents the glass contact layer of a first class fire-clay rich in alumina where mullite crystals attained a relatively larger size than in the previous figures. Crystals of corundum coaxed by glass appear in the contact layer. Corundum crystals can develop to extremely large sizes. As they are scattered at relatively great distances in the glass layer, they may pass into molten glass where they swim about as stones, and finally are found as nephelite stones in the produced glass.

Compared to the alkaline types of first class fire-clays, differences of mullite gall-stones appear fused in electric furnaces, under otherwise identical structural constituents, in the size and relative quantities of these structural constituents, in that the mullite crystals are of a larger size, and the quantity of glass filling up the interspaces is negligible, in the original substance. The quantity of interspatial glass, however, increases in the contact layer, due the partial intrusion of glass mass. In consequence, mullite dissolves in glass and corundum crystallizes in the meantime, until only corundum crystals embedded in glass remain in the portion of the contact layer facing the glass mass. As these crystals lie close to each other, they slowly disappear, due to dissolution and in this way, corundum crystals may get into the molten glass mass. Fig. 13 shows the contact layer of a KORHART gallstone. From the centre to the right a mullite crystal, representing an original constituent of the stone appears. Adjacent corundum crystals are separated from one another by glass layers, and from the mullite crystals. With the disappearance of mullite crystals, those of corundum increase. As the quantity of alumina required for building up the corundum lattice is made from mullite crystals, the layer in contact with the glass, in the sites consisting of the constituents corundum + glass,
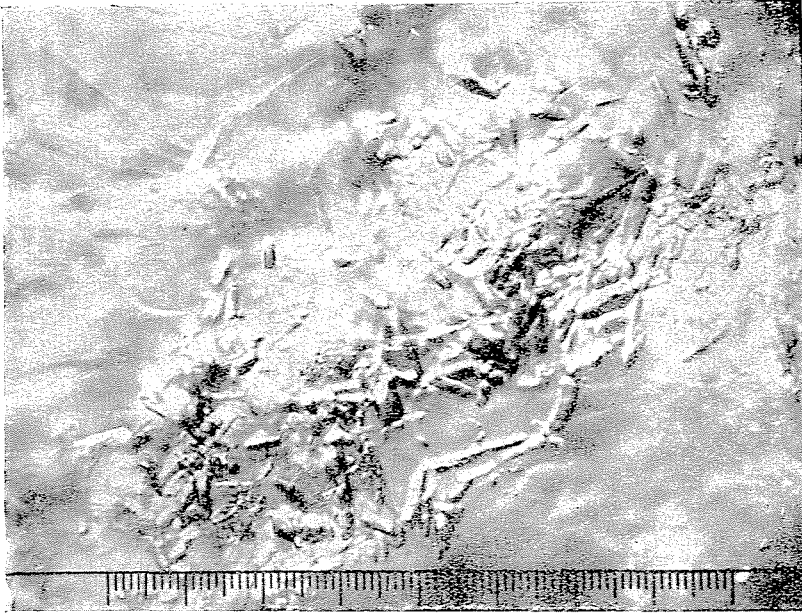
Fig. 9. Magnification 100×. Scale 1 micron. Mullite crystal agglomeration of Fig. 8

Fig. 10. Magnification 300×. Scale 10 microns. Nephelite gall-stone with dissolved corundum platelets of a coarse surface

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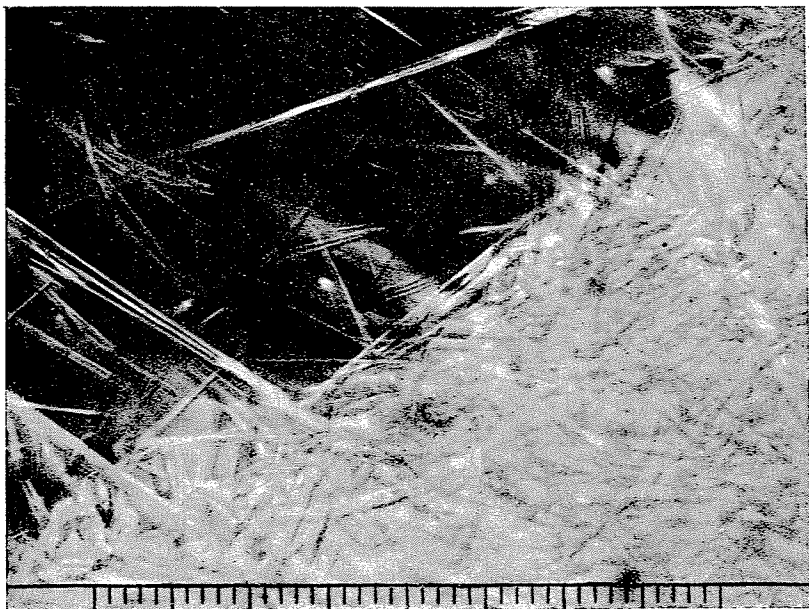
Fig. 11. Magnification 200×. Scale 10 microns. Contact layer of an acid beaker mass. The dark portions consist of glass, whereas the light ones are agglomerations of mullite crystals
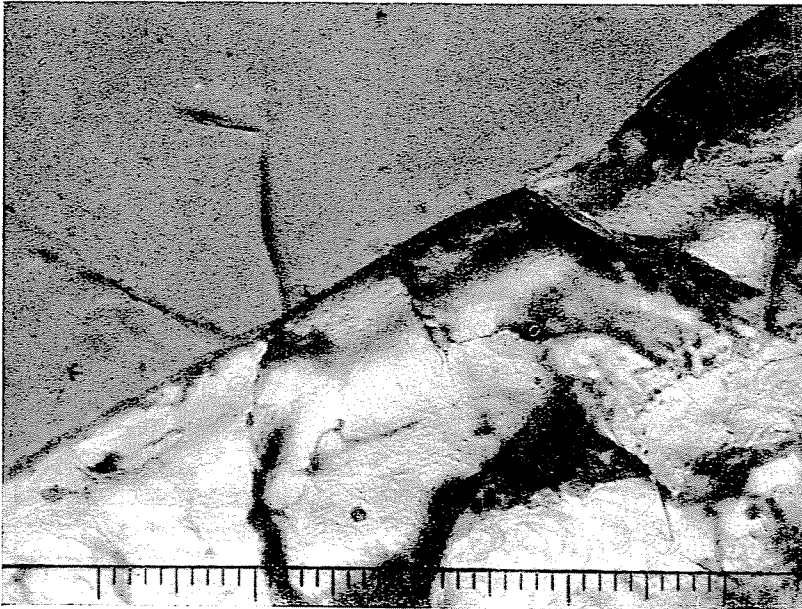

Fig. 12. Magnification 200×. Scale 10 microns. Layer of stone precipitates from first class fire-clay tub lining in contact with glass. The large crystals are corundum platelets, whereas the smaller crystals on the right represent mullite agglomerations embedded in the glass
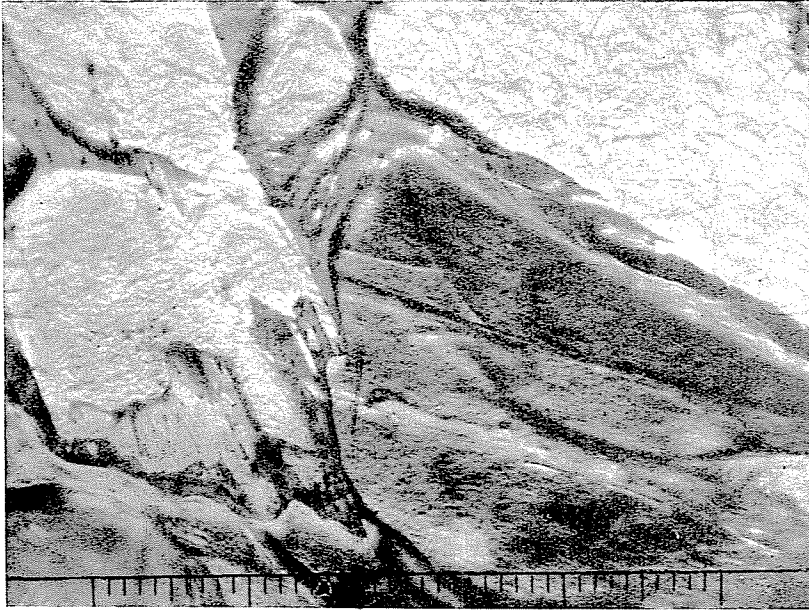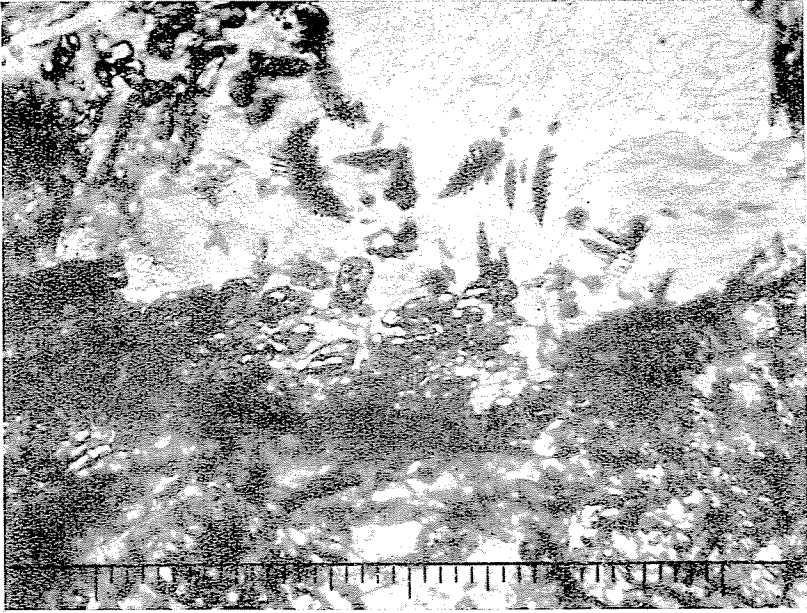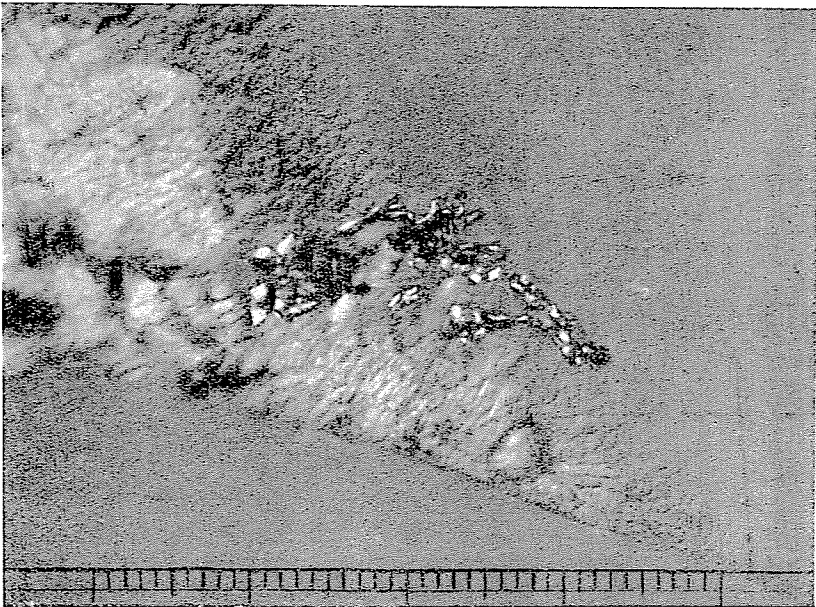
Fig. 13. Magnification 200×. Scale 10 microns. Portion of the contact layer of a disintegrated stone. Right, a dark mullite crystal, the other crystals being corundum, interspaces filled up with glass. The minute needles on the surface of the corundum crystals are rutile crystals

Fig. 14. Magnification 200×. Scale 10 microns. Contact of Korvisit stone with glass. The upper greyish portion is glass with two cracks in it. Corundum crystals appear in the lower part

Fig. 15. Magnification 200×. Scale 10 microns. Contact layer of ZAC tub stone with glass. The dark spots are crystals, the light portion the glass mass

Fig. 16. Magnification 200×. Scale 10 microns. Nephelite stone. In the centre crystals containing zirconium

an not have the compaction of the original mullite substance.

Molten glass does not intrude into the texture of corundum tank blocks cast in a fused state. These latter, mainly the pure types, have no perfectly compact, packed structure. However, molten glass cannot pass the leaks since it becomes highly viscous, due to uptake of alumina, and this film of extreme viscosity prevents the intrusion of glass into the substance of the structure. The contact layer is lacking. In the layer in contact with the glass in general no change of substance is perceivable. The mass of stones decreases, due to dissolution. No single corundum crystals get into the molten glass. Fig. 14 shows the contact layer of a pure corundum gall-stone and glass.

The structure of zirconium-containing substances cast in the molten state, also include amorphous glassy particles, and this fact is satisfactory to facilitate the intrusion of glass into the contact layer. Molten glass seems to loosen the texture of the substance which is recrystallized in the contact layer, although no corundum crystals develop. Crystal agglomerations of relatively high zirconium content can get into the molten glass. The contact layer of a stone of this type facing molten glass is presented in Fig. 15. The light portion is the glass mass in which dark crystal agglomerations are embedded.

Crystalline particles of this substance may be liberated and so pass into the molten glass. Fig. 16 shows a portion of a nephelite stone. In the centre, rounded crystals containing zirconium appear among the nephelite agglomerations.

As the specific gravity of nephelite (2,6) exceeds that of soda-lime glass, nephelite does not settle in molten glass, it may even float on the surface when, as it often occurs, occlusions are present. Corundum crystals of higher density sink much faster. This is the predominant reason why nephelite stones reach the processing zone.

LITERATURE

As regards nephelite : any mineralogical textbook.

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