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CRYSTAL-GROWTH TEXTURES IN MAGNETITE FLOWS AND FEEDER DYKES, EL LACO, CHILE

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ABSTRACT

The unique Quaternary iron oxide melts at El Laco, Chile, crystallized in intrusive and extrusive environments. These differ in morphological, structural and textural details that reflect the different environments of crystallization. Massive magnetite predominates in the intrusive bodies, whereas spherulitic, dendritic and idiomorphic magnetite, now locally oxidized, characterizes the oxide flows and feeder dykes. A transition from spherulite fibre to platy dendrite to euhedral crystal may occur within a few centimetres as an open space is approached. The spectacular rapid-growth features are attributed to sudden supersaturation due to degassing of the oxide melts. Much slower growth of idiomorphic magnetite or hematite (primary) then occurred directly from the gas phase; which of these oxides formed probably depended on the ability of hydrogen to diffuse rapidly out of certain gas pockets.

SOMMAIRE

Nous décrivons les manifestations intrusives et extrusives d'un magmatisme unique à oxydes de fer d'âge quaternaire à El Laco (Chili). Ces deux groupes de roches se distinguent par leurs formes, leurs structures et leurs textures qui diffèrent suivant les conditions différentes de cristallisation. La magnétite est massive dans les roches intrusives, mais elle est sphérolitique, dendritique ou idiomorphe (et localement oxydée) dans les coulées et les dykes alimenteurs. On peut voir, sur quelques centimètres, une fibre de sphérolite passer à une dendrite en plaques, et celle-ci à un cristal idiomorphe à l'approche d'une cavité. Nous attribuons ces faciès spectaculaires à croissance rapide à une sursaturation soudaine due au dégazage des bains d'oxydes de fer. Une croissance beaucoup plus lente de magnétite idiomorphe ou d'hématite primaire suivit, directement de la phase gazeuse; lequel des deux oxydes a cristallisé dépend probablement de la quantité d'hydrogène qui s'est échappée des poches de gaz par diffusion.

INTRODUCTION

The crystal-growth textures to be described here occur in the unusual El Laco iron ore deposits, located in the second Chilean region, Lat. 23° 48′S, Long. 67° 30′W, in the Andean Cordillera, 450 km east of Antofagasta. The deposits occur as extrusive and intrusive bodies

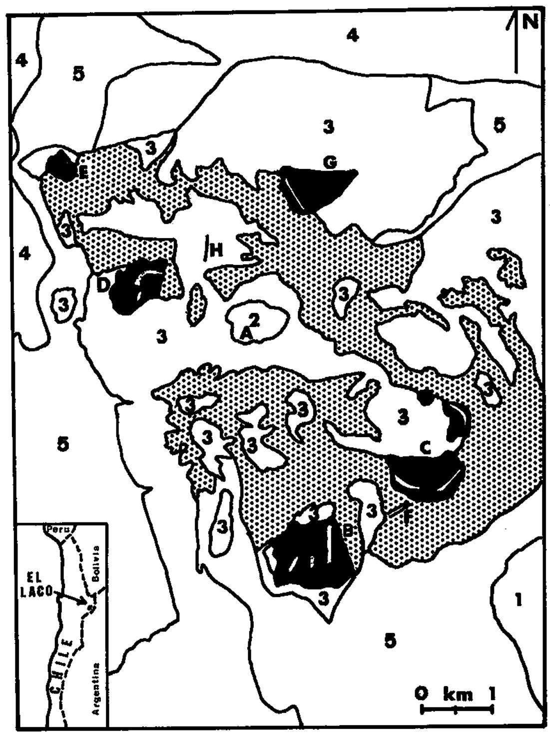

FIG. 1. Geological sketch map showing lithological units (1 to 5) and different localities (A to H) mentioned in the text. 1. Altos de Pica Formation (Tertiary ignimbrites); 2. El Laco rhyodacite dome; 3. El Laco andesitic flows; 4. Plio-Pleistocene andesitic flows; 5. Quaternary morainic and alluvial deposits. Hydrothermal alteration zones, thermal spring deposits and contact aureoles are shown in a stippled pattern. In black are the iron oxide flows, dykes (left white), domes and boulder fields. Localities: (A) Pico El Laco, (B) Laco Sur, (C) Laco Norte, (D) San Vicente Alto, (E) San Vicente Bajo, (F) Laquito, (G) Rodados Negros and (H) Cristales Grandes.

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on the flanks of a Quaternary rhyodacite–andesite cone over an area of 7 × 5 km, at an altitude ranging from 4300 to 5470 m.

The emplacement of the El Laco complex of andesites, rhyodacites and intrusive and extrusive magnetite-rich bodies clearly postdates the regional outpouring of ash-flow tuffs that constitute the ignimbrites of the Tertiary Altos de Pica Formation. Its emplacement seems more or less contemporaneous with that of Plio–Pleistocene andesites in the area. The complex defines a single volcanic edifice topped by a central crater, Pico El Laco, that is partly occupied by a young rhyodacitic volcanic dome (Fig. 1). Iron ore deposits, evidently of magmatic origin, occur in the periphery of Pico El Laco (A, Fig. 1), where they are interlayered with andesitic and rhyodacitic flows. The entire complex shows signs of important and pervasive hydrothermal alteration: small-scale contact aureoles in andesitic wall rocks enclosing feeders to the iron-oxide flows show superimposed broader-scale intense hydrothermal alteration that is still going on. Fumarolic activity is now concentrated at half a dozen centres on El Laco.

Four medium- to large-sized high-grade ore deposits have been mapped: Laco Sur, Laco Norte, San Vicente Alto and San Vicente Bajo; there are also three smaller deposits: Laquito, Rodados Negros and Cristales Grandes. These are shown as localities B to H, Figure 1. At least one additional deposit, inferred by magnetic survey and sought by drilling, remains to be discovered.

Park (1961) emphasized the unusual nature of these iron ore deposits when he called them "magnetite flows". Sanchez (in Ruiz *et al.* 1965) gave a short description of El Laco and produced the first geological map of the complex. Rogers (1969) briefly described the different types of ores, and Haggerty (1970a) provided the first careful description of their mineralogy. Magnetite is certainly the more important primary iron oxide; primary hematite is scarce. Secondary hematite and maghemite develop extensively as oxidation products of primary magnetite. Unusual Ca–Fe and Fe phosphates (Haggerty 1970b) have been identified.

Extrusive and Intrusive Deposits

A two-fold classification of the deposits into extrusive and intrusive types was first proposed by Henriquez & MacLean (1975). They considered Laco Sur, Laco Norte, San Vicente Alto and part of Rodados Negros (Fig. 1) to be extrusive. Evidence for an effusive origin is morphological, structural and textural, and includes: development of blocky flows (Fig. 2), locally highly vesicular (Fig. 3); iron oxide-rich pyroclastic material, largely lapilli and ash; slag-like material in flows characterized locally by contorted flow layering, ropy upper surfaces and gas escape tubes (Figs. 4, 5). Many of the field characteristics are those expceted of a low-

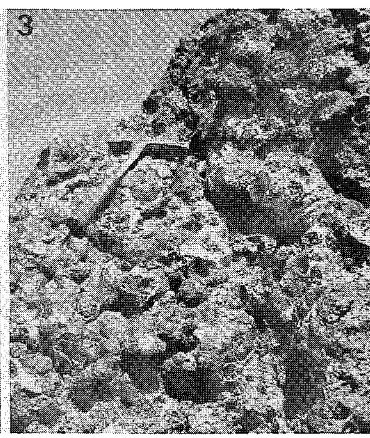

FIG. 2. Blocky flow of massive magnetite at Laco Norte (C, Fig. 1). A part of the arcuate feeder dyke can be seen in the background.

FIG. 3. Top part of a feeder dyke at Laco Norte. Note the highly vesicular, bulbous aspect of the exposure. Typically, an individual bulbous mass has an interior open space lined with idiomorphic magnetite. The white patches correspond to hydrothermal minerals that coat the walls of the large vesicles.

FIG. 4. Inclined gas escape tubes in one of the feeders at Laco Norte.

viscosity basaltic flow (*e.g.*, Macdonald 1967).

The effusive iron-oxide liquids have reached the surface *via* fissures, but these are not uniform in their orientation. At Laco Sur, the fissures belong to sub-parallel swarms, but at Laco Norte and San Vicente Alto, the flows occupy arcuate fractures that may reflect local collapse owing to evacuation of magma reservoirs. A drilling program in the central block between the arcuate fissures at Laco Norte failed to locate a central feeder dyke or any sign of an inwardly dipping cone sheet. Whether the fissures are straight or arcuate naturally leads to differences in outcrop patterns of the deposits and also in the disposition of gas escape tubes: parallel and vertical in the first, inclined and semi-radiating in the second (Fig. 4).

Evidence from surface exposures and from drill cores document the following typical stratigraphic succession: the first product to be expelled from the fissure was a magnetite-rich pyroclastic material deposited on El Laco andesitic lavas. In one hole, this unit is 30 m thick. Magnetite in this unit has been thoroughly oxidized to hematite. On this apron of pyroclastic material flowed a lava that crystallized largely to magnetite. The flows total 50 m in the same drill hole. Apatite is an accessory phase disseminated as fine grains. Where in contact with andesitic lavas, a narrow (1 m) contact aureole of actinolite + scapolite + quartz is developed. The outpouring of "oxide" lava was followed by another pyroclastic unit that contains fragments of massive ore, presumably detached from the feeders during a final explosive event. It measures 20 m in the section studied.

Shallow intrusive bodies have also been recognized: Laquito, Rodados Negros, Cristales Grandes and San Vicente Bajo (Fig. 1). They contrast with the extrusive type by their form, total absence of any signs of effusive activity, abundance of large, well-developed apatite crystals in amygdules and predominance of massive-textured magnetite. In all these deposits, the size and relative abundance of apatite increase towards the top.

The San Vicente Bajo intrusive body is dome-like, having the form of a bowler hat, 500×300 m. The other intrusive bodies are dyke-like, and attain 3 to 15 m thickness. The emplacement of these has led to a contact aureole of actinolite + scapolite + quartz in the host rocks within 2 m of the contact. Amygdules up to 15 cm across in massive magnetite near these contacts are commonly filled with actinolite + quartz + apatite; these must represent gas-rich bubbles such as observed in the extrusive deposits, but here the gas phase has evidently been unable to escape.

Crystal-Growth Textures

From the previous discussion, we infer that at El Laco, gas-charged iron-oxide-rich liquids reached near-surface environments to form intrusive domes and dyke systems; locally, some dykes also served as feeders for thin flows. We now turn to the signs of such an origin in the habits of crystals that grew from these unusual melts or from associated gases.

Crystal habits in intrusive bodies

The intrusive bodies consist essentially of massive magnetite, locally oxidized incipiently to hematite. In texture, the magnetite resembles the occurrences of massive magnetite already described by Park (1972) and Bookstrom (1977) from a number of iron ore deposits along the Chilean Coast Range. These deposits have been variously interpreted to be hydrothermal, contact-metasomatic or magmatic (Geijer 1931, Ruiz *et al.* 1965, Park 1972, Bookstrom 1977). An important difference with these, however, is the occurrence of spectacular apatite prisms that attain 7 cm in length in amygdules in

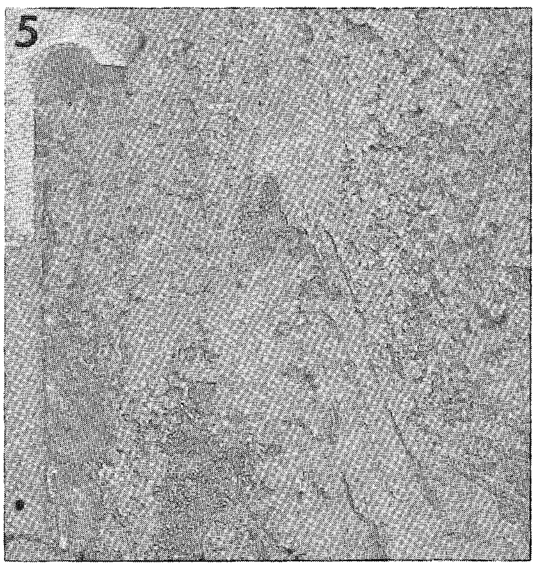

FIG. 5. Close-up view of one of the feeders at Laco Sur (B, Fig. 1). To the left, more or less parallel to the hammer, a vertical gas-escape tube can be seen with euhedral magnetite developed along its walls. At the centre a contorted magnetite layer shows the upward direction of movement.

massive magnetite, typically near the roof zones of intrusive bodies.

Even though these deposits undoubtedly formed from melts, we believe that crystallization was too slow to give rise to distinctive crystal habits.

Crystal habits in extrusive bodies

Our aim here is to document the spectacular and unique growth forms developed in the magnetite flows and feeder dykes of the Laco Sur, Laco Norte and San Vicente Alto occurrences. Dendritic magnetite is the most common expression of rapid growth. The dendrites form *in situ* at high levels in the feeders, particularly in walls adjacent to gas escape tubes and vesicles, and near the uppermost parts of slabs of magnetite flows (Fig. 6). These dendrites appear in uniform arrays of columnar "prismatic" magnetite; each prism typically measures 2 cm across by 10 cm in length, though miniature columnar individuals 1 × 4 mm also are found. The columns are generally distributed approximately perpendicular to a nearby open space (*e.g.*, vesicle, gas escape tube, top of flow) and attain a strikingly parallel disposition (Figs. 6, 7). The branching forms in the dendrites may look like fibres in two dimensions, but are really thin plates in parallel arrays, consistently 0.01 to 0.03 mm apart. The angle between plate and dendrite axis is 45° (Fig. 8), suggesting that the plates define the (111) growth face of magnetite; the complementary set of plates gives the appearance of an arrow pointing towards the open space (Fig. 7). No evidence of twinning has been found.

FIG. 6. Part of single magnetite flow now highly oxidized to hematite, showing the general disposition of "prismatic" dendrites. Laco Norte (C, Fig. 1).

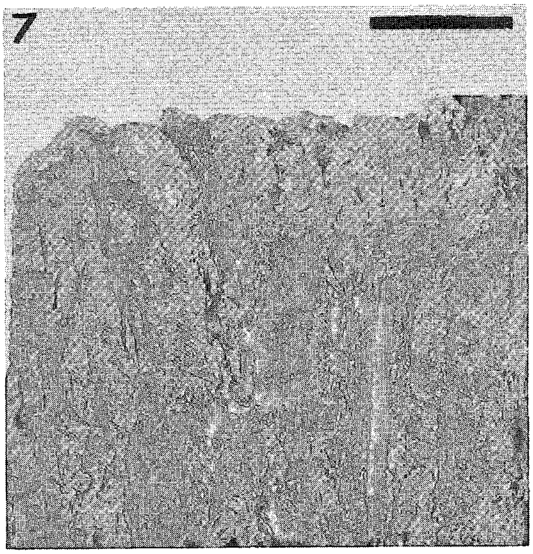

FIG. 7. Detail of an array of parallel platy dendrites pointing upward as arrows towards an open space. The dendrite to the left of centre shows a transition from abundant to few branching plates, then to a euhedral crystal at the top. Laco Sur (B, Fig. 1). Bar = 1 cm.

The number of plates decreases and their individual thickness increases as the crystal approaches the open space. This decrease is gradual, occurring over 2 to 3 cm in some of the larger dendritic individuals. The columnar dendrites are typically terminated by well-developed octahedral faces (111), $(\bar{1}11)$, and $(\bar{1}\bar{1}1)$. (Fig. 7). Depending on the proximity of these clusters of columns to circulating post-magmatic fluids, the dendritic magnetite may or may not show appreciable oxidation to hematite.

Spherulitic textures occur *in situ* at even higher levels in the feeders than dendrites. They are generally found near large vesicles, and smaller open spaces occur between individual spherulites (Fig. 9). In outcrop, the resulting rock looks knobbly and pitted (Fig. 4). The spherulites vary from truly spherical arrays of fibres to fan spherulites, in the usage of Lofgren (1974). They range from 1.8 to 4.5 cm across, and thus could also be called macrospherulites owing to their size (Donaldson *et al.* 1973). The individual fibre varies from 0.05 to 0.21 cm across; approximately 50 such fibres

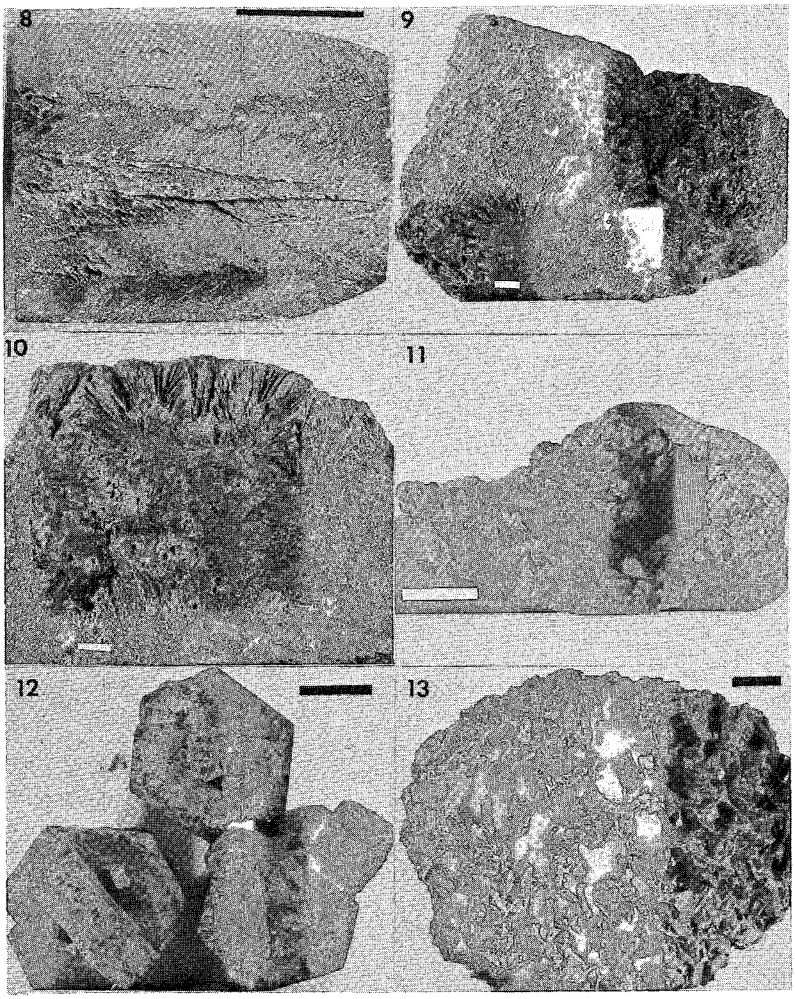

FIG. 8. Platy dendrite showing a 45° angle between plates and dendrite axis. Section from the same hand specimen as in Figure 7. Bar = 1 cm.

FIG. 9. Fan spherulite with small vesicle between individual spherulites. Euhedral hydrothermal minerals can be seen among the clusters of euhedral magnetite crystals in the vesicle. Laco Sur (B, Fig. 1). Bar = 1 cm.

FIG. 10. Fan and almost spherical spherulites. Upper surface corresponds to an open space. Dendrites develop from spherulite fibres in this specimen. The pits here represent gas cavities and axial holes in fibres. Laco Sur (B, Fig. 1). Bar = 1 cm.

FIG. 11. Octahedral magnetite developed along wall of a gas-escape tube. Laco Norte (C, Fig. 1). Bar = 1 cm.

FIG. 12. Primary hematite crystals ("nuts") with prominent {0006} and $\{11\bar{2}0\}$ faces and less common and smaller $\{10\bar{1}0\}$ faces. The crystals interpenetrate but are not twinned. Laco Sur (B, Fig. 1). Bar = 1 cm.

FIG. 13. Primary hematite plates grouped into a rosette pattern. Note the crystal of alunite (right of centre) deposited from late hydrothermal fluids. Laco Norte (C, Fig. 1). Bar = 1 cm.

can be counted in a typical array in two dimensions (Figs. 9, 10). Fibres of adjacent fan spherulites come in contact at angles attaining 100° (Fig. 10). Individual fibres may be partly hollow or filled with accessory co-precipitated phases, thus illustrating the intrafasciculate texture of Drever *et al.* (1972).

The larger spherulite fibres commonly show a transition over a short distance to a typical dendritic growth habit wherever the fibres are oriented towards an open space. There, they are invariably terminated by octahedral faces. Within centimetres, therefore, a fibre in a spherulitic array thickens and widens into a platy dendrite, then this growth form gives way to well-formed, idiomorphic magnetite crystals. Later transformation to hematite is sporadic, increasing near the open space.

The idiomorphic magnetite crystals occur along walls of gas-escape tubes, open spaces formed by the contortions of flow layering, and vesicles up to 70 x 20 x 15 cm (Figs. 3, 5, 7, 11). The crystals have an octahedral habit, with faces that are pitted and not truly planar; a single crystal may reach 6 cm across, but most are much smaller, lining the walls like quartz in a geode. Set among these clusters are occasional euhedral single crystals of minerals deposited from late circulating aqueous fluids (Fig. 9). Detailed X-ray diffraction studies (to be described separately) have led to the identification of intergrown iron-bearing sanidine and plagioclase. Rutile and quartz have also been identified by microprobe analyses. The fluids from which these feldspar crystals precipitated may have been responsible for the mild etching and oxidation of the magnetites.

Primary hematite crystals are very scarce, and have not been found *in situ*. These "nut"-looking tabular crystals attain 3.2 x 0.7 cm (Fig. 12); they have prominent {0006} and {11$\bar{2}$0} faces, and less common and much smaller {10$\bar{1}$1} faces. Interpenetrating crystals give a first impression of being twinned, with their {0006} faces roughly perpendicular (Fig. 12). In fact, the crystals are not related by a twin law. Also found are thinner plates of hematite grouped into a rosette pattern (Fig. 13). Low-temperature quartz, rutile and alunite have been identified as accessories deposited hydrothermally in open spaces between plates.

Discussion

So far as is known, the occurrence of dendritic and spherulitic magnetite in quenched iron-oxide liquids at El Laco is unique. A careful search of the geological and metallurgical literature on rapid-crystal-growth phenomena has provided only one pertinent reference (Hirano & Somiya 1976), a description of dendritic magnetite though from a hydrothermal, not a magmatic environment. Nor has any mention been made of the transition from spherulite fibre to dendrite to euhedral crystal in one single crystal. Comments that follow on interpretation of the textural developments are based on analogous habit modifications in different mineral groups and in metallurgical systems.

Tiller (1964, 1977) and Jackson *et al.* (1967) have described hopper dendrites of bismuth that resemble the El Laco magnetite dendrites; Billig (1955) has studied lamellar dendrites in Ge crystals, and Donaldson (1974, 1976) discussed plate dendrites in spinifex olivine crystals. The plate dendrites of El Laco are stacked like a deck of cards as are olivine plates in spinifex textures, but differ in the absence of other plates that cut across the main set of plates. This may be due in part to be high symmetry of magnetite compared to olivine. Furthermore, the interplate spaces at El Laco are empty.

The habit of a crystal is strongly dependent on such interrelated variables as diffusion rate (*D*), growth rate (*G*) and temperature. The transition in crystal habits from spherulitic to dendritic to hollow or skeletal to well-formed is related to variation in D/G ratio (Jackson 1958; Lofgren 1971, 1974; Donaldson 1974, Kirkpatrick 1975). Unfortunately, experimental studies are lacking and thermodynamic data of relevance are rare, such that semi-quantitative inferences of diffusion and growth rates here would be hazardous.

We do know the melting point of Fe_3O_4 at 1 atm, 1870 $\pm$ 2 K, and its enthalpy of fusion ΔH°_{m}, 33.0 $\pm$ 2.0 kcal/mole (JANAF tables 1971). The entropy of fusion of Fe_3O_4 we get from these data, $\Delta S^{\circ}_{m} = 17.3$ *e.u.*, is relatively high (Uhlmann 1972) and suggests that the structural reorganization in crystallizing a liquid of composition Fe_3O_4 is rather important. In turn, this implies that nucleation will require high supersaturations and that growth will be anisotropic (Jackson *et al.* 1967, Uhlmann 1972) favoring the development of faceted dendrites. This presumably accounts for the rigorously parallel platelets in Figure 6.

But what is the effect of adding volatiles to the system? Gases (*e.g.*, H_2O, HCl) certainly played a major role in lowering the temperature of the liquidus at El Laco, as no evidence of unusually high temperatures can be found in the contact aureoles. The fluxing effect of gaseous species has been documented already: in the system Fe–C–O, Weidner (1968) found that liquids rich in Fe_3O_4 could be expected at temperatures as low as 815°C at 350 bars; Gibbon & Tuttle (1967) have also described an Fe_3O_4-rich liquid at 1060°C in the system $FeO–Fe_2O_3–SiO_2–H_2O$; Philpotts (1967) found an iron oxide liquid at 1420°C in the "system" apatite–diorite–magnetite at low pressures.

Diffusion rates depend rather importantly on viscosity. No data are available for liquids even close to Fe_3O_4 composition, nor for liquids containing appreciable Fe_2O_3. The closest approach to an answer comes from rare investigations of slag systems, in which the component FeO is by far more important than Fe_2O_3. Extrapolation of viscosity determinations in the system $FeO–SiO_2$ between 1100° and 1450°C (Urbain 1951) and in the system $CaO–FeO–SiO_2$ between 1200° and 1350°C (Röntgen *et al.* 1960) to pure FeO gives a value of the order of 0.2 poises. It could be argued that Fe_2O_3 does not affect viscosity differently than an equivalent amount of FeO (Bottinga & Weill 1972), but the bulk composition effect here would weaken this inference based on silicate systems. The El Laco liquids were undoubtedly more viscous than 0.2 poises, but by how much? Lower temperatures in nature would lead to an increase in viscosity, but complicating the system by adding H_2O, HCl and P would likely induce a compensating effect; Ca and Si impurities will raise viscosity at constant T (Röntgen *et al.* 1960). We tentatively envision fluidities comparable to those of ultrabasic liquids that led to rapid-growth habits of olivine (*e.g.*, Arndt *et al.* 1977).

Degassing of the El Laco Fe_3O_4-rich liquids probably played a crucial role in determining rates of crystal growth. Jackson *et al.* (1967) have shown experimentally that degassing can increase growth rate in certain organic systems; Donaldson (1974) has postulated that spinifex olivine clearly forms in response to sudden supersaturation. Lofgren (1974) and Donaldson (1976) have catalogued the growth habits of plagioclase, pyroxene and olivine as a function of degree of supercooling in isobaric experiments. We contend that the gas escape tubes, the vesicles and the pyroclastic unit confirm the role of degassing, locally violent, in the case of the extrusive flows and related feeder dykes. This is the best mechanism to induce sudden supersaturation. In contrast, the intrusive bodies at El Laco, though very shallow, failed to degas completely or as rapidly. The presence of amygdules with large apatite crystals suggests that volatiles were more efficiently trapped where they did separate; degassing may also have been more gradual.

At El Laco, spherulitic magnetite grew at the top of the feeder dykes, where supersaturation due to degassing or to thermal quenching (or both) might be most extreme. Spherulites develop wherever $G >> D$ (Lofgren 1974). Platy dendrites grew lower down in the feeder dykes where cooling rate might have been slower or where degree of supersaturation due to less complete or rapid gas loss might have been lower. The change in morphology towards a platy dendrite involves a decrease in G/D (Lofgren 1974, Kirkpatrick 1975, Donaldson 1976). With decreasing degree of supercooling, $\triangle T$, in isobaric experiments, Lofgren (1974) found the diameter of spherulite fibres to increase. At El Laco the diameter of these fibres is relatively large compared to those in other mineralogical systems, such that they may have formed at a degree of supersaturation already close to the transition to dendritic growth forms. The decrease in rate of growth with time may result from local buildup of dissolved gaseous species owing to initially very rapid growth of an anhydrous mineral from a liquid that was not totally degassed.

One may argue qualitatively that spherulitic magnetite would not have been possible at El Laco if the melt viscosities were as low as those obtained in the systems $FeO–SiO_2$ and $CaO–FeO–SiO_2$. Higher viscosities would be required to ensure that $G >> D$. From an analysis of the factors that affect the transition spherulite–dendrite, Tiller (1977) proposes that viscosities exceeding 1 poise are necessary to obtain either habit.

Whereas massive, spherulitic and dendritic magnetite undoubtedly grew from melts of variable degrees of supersaturation, we visualize a different mode of formation for the idiomorphic crystals. These grew from a gas phase; the entropy change during this process of crystallization is considerably larger than when Fe_3O_4 crystals grow from oxide melts. Consequently, using the reasoning of Jackson (1958) and Uhlmann (1972), we expect fewer but well-formed and euhedral crystals. A question arises concerning the possible dissociation of water in these high-temperature gas pockets, and subsequent loss of hydrogen. This phenomenon may have caused localized unusually oxidizing conditions in some pockets; this would explain the formation of the "nut"-shaped hematite crystals. Kolb *et al.* (1973) have shown that hematite may form in this way experimentally, confirming Eugster's (1959) prediction. The persistent nucleation and growth of hematite from an aqueous fluid in the experiments of Martin & Piwinskii (1969) with granitic rocks may also find an explanation in the same phenomenon.

Also, Hirano & Somiya (1976) found that hydrogen plays a very important role in producing euhedral magnetite crystals grown from an aqueous fluid, whereas Tiller (1977) emphasizes the importance of hydrogen content of the gas phase in producing euhedral crystals in general.

Another important variable in the growth of these idiomorphic crystals might be the partial pressure of Fe at high subsolidus temperatures. These partial pressures have been shown to be appreciable above 900°C by Darken & Gurry (1946) and Eugster (1959). Characterization of the post-magnetite and -hematite minerals deposited from the gas phase will allow a more complete reconstruction of the evolution of the post-magmatic hydrothermal fluids at El Laco.

We have presented here the striking evidence for growth of magnetite from iron-oxide-rich liquids, and of magnetite, hematite and other minerals from related gas phases. Petrological, mineralogical and geochemical studies, now in progress, will hopefully resolve the meaty issue of the ultimate derivation of these unusual, gas-charged liquids.

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