Fluid-inclusion Evidence on the Environment of Formation of Mineral Deposits of the Southern Appalachian Valley

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Abstract

Approximately 1,330 fluid inclusions were studied in samples of ore and gangue minerals from both massive ore and late-stage vugs from a series of Appalachian deposits and five active mines in the East Tennessee zinc districts. Most primary inclusions in sphalerite, fluorite, dolomite, and quartz from East Tennessee homogenized at 82°–149° C. Most primary inclusions in fluorite, barite, and sphalerite from the Central Kentucky, Central Tennessee, and Sweetwater barite districts, ranged from 72°–132° C. Pressure corrections to be added are probably less than 10° C.

Freezing data were obtained as a crude measure of the salinity of the inclusion fluids. With few exceptions, all primary inclusions contained very strongly saline brines (mostly >20 weight percent salts), with appreciable amounts of at least some salts other than NaCl, and some contained immiscible globules of oil. The exceptions include several quartz, fluorite, calcite, and barite samples that contained only moderately saline brines (12–16 percent), and some inclusions containing essentially fresh water, in vug calcite. Secondary inclusions had lower salinities than adjacent primaries. Eight primary inclusions in the differently colored outer millimeter of one Central Kentucky fluorite had low salinity (4–6 percent).

The data indicate that all these deposits formed from hot, saline brines, with sphalerite forming from slightly hotter and more saline fluids than later gangue minerals. This places severe restrictions on possible modes of origin and makes deeply circulating connate brines the most probable ore fluids. The data give no information on the cause or direction of fluid circulation, which would be controlled by topography, salinity, and temperature during ore deposition, and they permit wide latitude in the construction of possible models. Other observations, on "colloform" textures, seem to indicate the admixture of at least small amounts of surface waters with the brines.

Introduction

The zinc and barite deposits of the Southern Appalachian Valley constitute a rather large mineral district, spread over a considerable geographic area. Despite much detailed field work, there remains a long-standing disagreement on the origin of these deposits. This work has revealed many valuable facts that must, of course, be considered in any suggested mode of origin. These many field studies are adequately covered in the other papers (and their bibliographies) in this symposium number and hence are not repeated here.

In this paper I will attempt to show only the nature and validity of the evidence as to the environment of formation of these mineral deposits that can be obtained from a study of the fluid inclusions in them. Most of the work deals with the deposits of East Tennessee, but a large number of apparently similar deposits and prospects throughout the Appalachian Valley have been examined more briefly.

Materials Studied

Samples of both ore and gangue minerals were obtained from many different mines and prospects, as described in the Appendix. One or more very carefully prepared doubly polished plates was cut from each sample. These varied in thickness from about 0.1 mm (dark sphalerite) to 2 mm (transparent fluorite and light sphalerite). Although numerous samples of ordinary ore were examined, including material showing replacement textures, many of the samples were of special materials that appeared to have better chances of providing usable fluid inclusions. For this reason, the atypical coarser, late-stage vug mineralization is more heavily represented among the samples (Figs. 1–11). Even in these late stage materials a complex history is frequently evident in the form of fracturing and later overgrowth, color zoning, etc. As indicated later, no significant difference could be recognized between the fluid inclusions in these various types of samples from any given locality, although the range of data obtained from any given deposit probably represents real variations in the ore fluids with

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Photomicrographs

(All taken of polished plates in transmitted plain white light)

Fig. 1. Sphalerite, Coy Mine, Mascot-Jefferson City district, East Tennessee. Very large primary inclusions from the termination of a rosette group (sphalerite "egg") of radial yellow crystals several cm in diameter protruding into vug. Freezing temperature $-10.3^\circ$ C. Homogenization temperature not determined, as these leaked on freezing, but similar inclusions in same sample ran 150-178$^\circ$ C; ER 65-111.

Fig. 2. Fluorite, Coy Mine, Mascot-Jefferson City district, East Tennessee. Pale purple, 3-mm cube from vug, enclosing many small crystals of quartz and carbonate, some of which serve to outline the growth stages along with planes of primary fluid inclusions. The insert is an enlargement of one of these planes. Freezing temperatures range from $-11.2^\circ$ in the center to $-8.8^\circ$ C in the outer rim. Homogenization temperatures range from 138$^\circ$ to 170$^\circ$ C, with no recognizable pattern; ER 65-114.
stage of paragenesis that could not be resolved at this sampling frequency.

Much of the material examined proved to be relatively unsuited for fluid inclusion studies for one reason or another. The most common reason was that the inclusions were too small and rare. Numerous specimens that showed no suitable inclusions are not listed in the Appendix. In spite of these limitations, however, approximately 1,350 usable fluid inclusions were found and determinations made of either freezing or homogenization temperature or both. Many of these inclusions could be characterized as primary, with little ambiguity. A few fairly obviously secondary or pseudo-secondary planes of inclusions were studied (e.g., Fig. 3), and these gave results which were compatible with the history of the sample as recorded in its primary fluid inclusions.

Several samples were examined on the crushing stage (Roedder, 1970), to check for the presence of noncondensable gases under pressure. All inclusions seem to contain at least some gas, under pressures of several atmospheres, and some, such as ER 69-36b from the Central Tennessee district, contain large volumes of gas. This gas is probably mainly methane.

Barite in particular was difficult to work with, as most of the inclusions in it are apparently secondary. Many of the barite samples are very rich in fluid inclusions, and some are almost porous, containing an estimated 20 percent by volume of inclusions. Even a casual examination of such material shows, however, that most of these inclusions are now along healed cleavage fractures, and also that there are wide differences in the degree of filling of individual adjacent inclusions, indicating that considerable necking down and leakage has occurred. These barite crystals have been fractured numerous times since they first crystallized, and hence any larger, possibly primary fluid inclusions in them may actually contain later fluids. Most data on the Sweetwater barite district thus had to be obtained on the fluorite that is intimately associated with the barite, but it, too, is not easy to work with (Fig. 4).

Most sphalerite samples were disappointingly free of inclusions although some contained small areas with numerous very large ones. Figure 1 is an excellent example. This one small crystal, protruding from the surface of a sphalerite "egg" from the Coy mine, had three large primary inclusions in it.

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**Fig. 3.** Fluorite, Coy Mine, Mascot-Jefferson City district, East Tennessee. Planes of presumably secondary inclusions along former cleavage fractures. Individual planes each had specific freezing and homogenization temperatures, but all were in the same range as those found for primary inclusions in the same sample. ER 65-114.

**Fig. 4.** Fluorite, Ballard mine, Sweetwater barite district, East Tennessee. Both fluorite and barite from this district are thick with fluid inclusions, mostly of unknown origin, and some crystals of white barite may contain twenty percent inclusions by volume. Note that many of the inclusions are full of liquid with no bubble (a), some have small bubbles (b), some have large bubbles (c), and some are dark and filled with gas (d, out of plane of focus). This variation may be a result of leakage, necking down, or many generations of fracturing and rehealing. ER 65-106.

**Fig. 5.** Fluorite, Ballard mine, Sweetwater barite district, East Tennessee. Small, probably primary inclusion containing brine, vapor bubble, and a cluster of grains of brownish material, in part birefringent, that is presumed to be organic. Homogenization temperature unknown, as inclusions leaked during heating. ER 65-103.

**Fig. 6.** Fluorite, Moore or East Prospect mine, Central Kentucky district. Colorless fluorite covering scalenohedral calcite. This is typical of the clean primary inclusions found in such samples. Freezing temperature 15.9°C; homogenization temperature 107°C. ER 62-24.

**Fig. 7.** Fluorite, Faircloth mine, Central Kentucky district. Colorless cubes from vein. Photographed with partially crossed polarizers. Although this appears to be two separate inclusions, the birefringent daughter crystals (x) go to extinction simultaneously and may be connected by a thin red. Presumably after trapping, the daughter crystal started growing in an area that was necking down, and this seed crystal was hence available for further growth in each of the two resulting inclusions. ER 63-177.

**Fig. 8.** Sphalerite, Flat Gap mine, Copper Ridge district, East Tennessee. This inclusion, presumably primary, is in a coarse clear yellow late sphalerite that has completely replaced a single crystal bleb of galena (see Fig. 9, from same sample). The sawtooth interface between the red-brown "schalenblende" (black in photo) and the sphalerite is a relic from the series of euhedral facets on the galena bleb, formed during its growth. Freezing temperature 27.0°C, homogenization temperature 90°C. ER 65-42.

**Fig. 9.** Sphalerite, Flat Gap mine, Copper Ridge district. East Tennessee. This "colloform" or "schalenblende" sample of banded yellow to red-brown sphalerite contains many blebs of galena (black), elongated in the direction of growth, that have grown simultaneously with the surrounding finely banded crystalline druse of sphalerite. Some of these (x) that happened to be oriented with (100) parallel to the bands, grew out over the sphalerite in skelletal, cuplike forms (from prematurely edge growth). Renewed nucleation of an additional band of pale yellow sphalerite then covered the galena crystals (Roedder, 1968b, Fig. 23 II, p. 464). These galena crystals were then in part replaced with coarse, clear yellow and in part purple sphalerite (Fig. 8), particularly along the interface with the schalenblende (bright spots in the lower right). Other galena blebs are replaced with a combination of calcite, ridden with inclusions, and yellow sphalerite. ER 65-42.

**Fig. 10.** Sphalerite, Faircloth mine, Central Kentucky district. Many single crystals show such zones of very finely striated pale to dark purple gray color. With increasing magnification, up to the limit of the light microscope, more and more striae become visible (see inset strip at top, from this same field, taken with a long exposure at an initial magnification of 1,700 X). These striae are parallel to the crystal growth surfaces, and presumably represent oscillations in growth conditions. The crystal grew from left to right. ER 63-179.

**Fig. 11.** Sphalerite, Flat Gap mine, Copper Ridge district, East Tennessee. "Colloform" crust of sphalerite ranging in color from colorless through yellow and red to dark brown in doubly polished plate 0.15 mm thick. At high magnification individual bands present a zig zag appearance, outlining former crystal growth terminations in the many tiny elongated crystals that comprise this fine druse (Roedder, 1968b). Growth was from the bottom upward. ER 65-44.
However, all of the other crystals sectioned from this same “egg” were almost optically clear. The bulk of the sphalerite examined from the Flat Gap deposit is the early generation schalenblende variety. Inclusions in this material were exceedingly difficult to work with, but although they gave results of poorer quality due to the larger limits of error, the results were always compatible with the data from the larger inclusions in later generations of vug minerals.

Fluorite crystals were particularly suitable for inclusion work, in that they frequently contained large primary fluid inclusions with good optics (Figs. 2, 5 and 6), but it should be remembered that fluorite is a late-stage mineral in many of these deposits. Similarly, quartz and calcite are generally late-stage minerals. Most of the dolomite examined is cogenetic with late sphalerite.

With few exceptions, as indicated in the Appendix, the inclusions consisted of a brine with a very small bubble (generally 2–5 volume percent). The salinity of the brine is evidenced by its reluctance to freeze at $-78^\circ$ C and by its low coefficient of thermal expansion on heating, as well as by the freezing temperature determinations described later. The exceptions consist mainly of accidentally trapped liquid and solid yellow to brown organic matter, which was found along with brine in the Central Kentucky, Central Tennessee, and several of the East Tennessee barite districts (Fig. 5). Some inclusions in Central Kentucky also contained a very small highly birefringent daughter crystal, probably a carbonate (Fig. 7), and a very few inclusions in quartz from the Coy mine had daughter crystals of NaCl that dissolved at $+78^\circ$ C.

Strongly fetid barite was examined from a series of localities in East Tennessee (e.g., Laurence, 1938), in the hopes that liquid H$_2$S might be recognized. Some inclusions from the Cox prospect, Fall Branch district, Tennessee (ER 63-80) showed two immiscible fluids, but they were too small in size to distinguish between oil and possible liquid H$_2$S. On the crushing stage these emitted large volumes of a gas that was very soluble in kerosene, so it is presumed that the second liquid was probably liquefied organic gas. Similar evolution of a kerosene-soluble gas, and a strong petrolierous odor, was found in barite from the Nuns Cove, Tennessee barite prospect (ER 63-82).

One group of liquid CO$_2$ inclusions was found, but these were obviously secondary inclusions in a healed fracture across a detrital, rutile-tailed quartz grain partially replaced by sphalerite in sample ER 63-45 from Flat Gap. These are merely an indicator of the earlier higher temperature-pressure conditions under which these detrital quartz crystals originated.

**Ore-Fluid Salinities from Freezing Temperatures**

After selection, the vapor bubble in each inclusion was checked for sensitivity and direction of movement in a thermal gradient as an indication of compositional variations in the fluid (Roeder, 1963). This test showed that there were variations in the composition of the fluid in the various inclusions, as some bubbles moved up a thermal gradient, some moved down, and some failed to move at all in a thermal gradient, even though they moved freely by gravity.

Each of the specimens containing suitable inclusions was then examined on the freezing stage (Roeder, 1962) to estimate the salinity of the inclusion fluid. As is common in fluid inclusions from Mississippi Valley-type deposits, many of these remained super-cooled, without freezing, for considerable periods of time. The freezing temperature results on 865 inclusions, all primary, are summarized in Figure 12. For any one mineral and locality, the data represent the summation of one or many sample plates, which may have come from one or more locations in the mine (see Appendix). In addition to these 865 primary inclusions, numerous planes of secondary inclusions checked during the runs showed freezing temperatures that were either comparable to the data from the adjacent primaries, indicating possibly that the planes of inclusions were pseudosecondary rather than secondary, or they showed much lower salinities, presumably indicating formation at some later time from fluids more nearly approaching fresh water. Only very few primary inclusions showed freezing temperatures near 0° C, indicating essentially fresh water. All of these were in calcite, and hence may indicate that this calcite grew at a late stage from ground water. Not all calcite is in this category, however, as 26 inclusions in a calcite crystal from a vug in the Young mine showed freezing temperatures of approximately $-10^\circ$ C, corresponding to about 13 weight percent salts in solution—certainly not a ground water. A series of eight primary inclusions in the outer millimeter of one crystal of fluorspar (ER 63-178) from the Central Kentucky deposits (Fig. 12) provided the only sample of moderately saline fluids found. These inclusions may indicate an overgrowth at a later time, as this outer millimeter is purple, although the rest of the crystal is colorless.

Some inclusions are represented with arrows off...
Fig. 12. Frequency distribution diagram of freezing temperatures of the 865 primary inclusions measured. C-calcite; S-sphalerite (solid bars); F-fluorite; Q-quartz; D-dolomite. The bars show the range of data for the number of inclusions indicated, with the mode shown by a vertical line. See text for other details.

to the right of Figure 12. The actual freezing temperatures on these varied over a wide range, but as they were the melting temperatures of the last crystal of NaCl·2H₂O instead of ice, these solutions must be much more concentrated than those fluids yielding ice as a last crystalline phase at the liquidus temperature. Although the freezing temperatures of inclusions in the gangue minerals such as fluorite and quartz are frequently about −10° C, corresponding to only 13 weight percent salts in solution, the inclusions in sphalerite (all deposits) average closer to −20° C, corresponding to approximately 25 weight percent salts in solution.

Many of these inclusions, both those with high and low salinities, showed “superheated ice” at high negative pressures (Roedder, 1967b), although the
temperatures were below 0° C for the saline ones. As these temperatures represent metastable equilibria, they are not plotted in Figure 12, but they were compatible with the data shown there.

Actually, the true salinities for these various solutions can only be estimated, because different salts depress the freezing point differently. Since NaCl·2H₂O has an eutectic with ice at -21° C, all inclusions showing freezing temperatures below -21° C must also have salts other than sodium chloride. First melting temperatures of -28° C and even colder, as commonly observed in these samples, would indicate that calcium chloride is present in these solutions to an appreciable degree, although no quantitative analyses have been made. The type of sample material available at present does not look very favorable for chemical (or isotopic) analyses of inclusion fluids, unless some new techniques can be developed.

Many of these inclusions were so nearly filled with liquid that fracturing might be expected on freezing. As a result these were run while immersed in oil in a miniature double-windowed cell inside the regular cell of the cooling stage. This technique prevents the entry of the circulating fluid (acetone), and hence permits reasonably accurate freezing temperature determinations even on inclusions that do fracture on freezing. The thermal mass and conductivity of this inner cell results, of course, in a much slower approach to thermal equilibrium.

Ore-Fluid Temperatures from Homogenization Temperatures

After the freezing runs, homogenization temperatures were determined for some of these same inclusions (Fig. 13), using a Leitz model 350 heating stage. The pressure correction to be added to these data to obtain trapping temperatures (except for oil inclusions—see below) is probably less than 10° C. In those samples in which cracking was expected due to the freezing, similar inclusions from other parts of the same sample were used for the heating runs. The effects of temperature gradients in the stage were minimized by essentially leveling off the temperature after each increment, and by the use of melting-point standards sealed in capillary tubes placed on the stage with the sample chips. The major problem (in addition to sample selection), however, was not calibration but the optical quality of the material itself. The rather low aperture afforded by the heating stage design makes it impossible to get light into many inclusions in sphalerite that appear perfectly transparent with a high power condenser and no diaphragms. Strong oblique illumination from above or below the stage, using a flexible fiber optics illuminator, helped to reveal the bubble in some cases, but most of the homogenization data of Figure 13 probably has an observational uncertainty of ±2° or 3° C.

Following the homogenization determinations, each inclusion was checked for leakage either by a repeat run, or by remeasuring the size of the bubble at room temperature, or both. The former is a necessary but not sufficient criterion. A few inclusions with initially very small bubbles failed to regain their bubbles upon cooling, following the homogenization run.

The homogenization temperatures of the sphalerites in Figure 13, with the exception of five inclusions in sphalerite from the Central Kentucky district, and one from the Young mine, were all over 100° C, whereas the gangue minerals mostly homogenized at approximately 100° or slightly under. In addition to the 784 inclusions shown on Figure 13, many planes of secondary inclusions were also measured. Most of these showed homogenization temperatures comparable to those of the primary inclusions, and hence are presumed to be pseudosecondary inclusions. A few showed homogenization temperatures that were considerably lower, and some planes of secondary inclusions appeared to be full of liquid at room temperature, indicating they were formed at near surface temperatures. Many such full inclusions were found in the Sweetwater barite samples.

Temperature measurements have already been reported on fluid inclusions from several of the districts investigated in the present study. None of these previously published data are given in Figure 13, except for preliminary reports of some of these same data in earlier papers (Roedder, 1967c; Jolly and Heyl, 1968). The present work on fluorite samples from the Young mine yielded homogenization temperatures considerably below those reported by Miller (1967, 1969). A similar difference occurs between the present results on fluorite from the Sweetwater barite district and those reported by Nadeau (1967). The cause for these differences is not yet agreed upon, and will be discussed in a later note.

Hoagland et al. (1965) suggest that the difference in color and iron content of the two main stages of

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Fig. 13. Frequency distribution chart of homogenization temperatures of the 784 inclusions measured (mainly primary). C-calcite; S-sphalerite; F-fluorite; Q-quartz; D-dolomite; B-barite. Each small square represents one determination; an arrow and a number at the top indicate larger numbers of inclusions in a given interval than could be plotted conveniently. Hollow squares represent determinations on oil inclusions. See text for other details.
sphalerite mineralization at the Flat Gap mine (early high iron sphalerite, 4.75 percent, and late yellow sphalerite, <1 percent) reflects a decrease in temperature. This early sphalerite is particularly free of usable fluid inclusions, but the few that were found were in the same homogenization temperature range as those in the later, low-iron sphalerite, an observation completely compatible with the phase relations in the Fe-Zn-S system (Barton and Toumin, 1966).

Fluid-inclusion measurements (incorrectly attributed in part to Roedder) are stated to "suggest a fairly high-temperature origin" for the Friedensville zinc deposits (Heyl et al. in U. S. Geological Survey, 1962, p. A3). The "fairly high temperature" quoted here was used in a relative sense, compared to other, rather low-temperature Mississippi Valley type deposits (A. V. Heyl, oral communication, 1969).

D. M. Pinckney (U. S. Geological Survey, 1962, p. A4*) reported fluid-inclusion temperatures of 185° to 207° C for late quartz from the Young mine, and 115° to 125° for fluorite (Heyl et al., 1966 and oral communication, 1969), but further details have not been published. Newhouse (1933) reported a range of homogenization temperatures from 70° to 95° C for sphalerite from the Boston-Kennedy mine, Lockport, Henry County, Ky. This is part of the Central Kentucky district and may well have been another mine on the Gratz vein (sample ER 63-174).

A series of samples of calcite and fluorite from seven near-surface veins in the Central Tennessee district (Jewell, 1947) gave results consistently lower than those from East Tennessee, and showed a consistent difference (discussed below) between the homogenization temperatures for oil inclusions and for cogenetic water inclusions. Most of the brine inclusions from these veins, and from cores from the major orebodies lying deep beneath these surface veins, homogenized in the range 90°-120° C, significantly below the temperature determinations for the East Tennessee districts.

**Microscopic Banding of the Sphalerite**

When doubly polished plates are cut from the "colloform" or "schalenblende" sphalerites from the Flat Gap mine, a peculiar periodic color variation is noticed that is believed to represent annual "varves" (Roedder, 1968b). Generally this manifests itself as an alternating dark and light color in transmitted light, visible at moderate to high magnification. In reflected light it is not usually visible. The range of colors between adjacent bands may vary from small differences in slightly olive brown colors to the difference between dark brown and practically white. Detailed examination of these sections shows that these color bands are perpendicular to the growth direction and result from some change in the conditions of the growth of the crystals making up these fine druses. When examined at very high magnification, it is seen that most of the changes from earlier light color to later dark color are more or less gradual or transitional, whereas many of the color changes from an earlier dark to a later light band are rather abrupt. Although there are many exceptions, I believe the conclusion is statistically valid that, whatever is the cause for the variation in color of the deposited sphalerite, the change or oscillation in conditions was generally gradual in one direction and abrupt in the other.

In many of the deposits, for example the Central Kentucky sphalerite, no "schalenblende" was examined—only coarser crystals. When these individual single sphalerite crystals are examined in detail, however, it is seen that many of them have a banding. There may be large differences between the thicknesses of adjacent bands, but this banding, as shown in Figure 10, is very minute. Some band pairs (one dark and one light) have a total width of as little as 0.5 μm. The cause of the dark color in the dark bands is not known, although the color is very intense (Roedder and Dwornik, 1968). Some evidence indicates this coloration is controlled in part by the specific growing crystal face involved, as a few of the exceedingly dark zones in some crystals have almost perfectly transparent bands running through them at an angle to the individual growth layers. These appear to be crystallographic twin bands, presumably representing portions of the crystal formed by growth of a different crystal face, that formed a lesser number of color centers.

**Origin of the Deposits, in the Light of Fluid-Inclusion Evidence**

As is commonly the case in geology, the data presented here are not adequate to determine the origin of these deposits. However, these data present some limitations on the possible theories of origin that can be proposed, and any theory of origin that is not compatible with the fluid inclusion evidence must also include an explanation of why the fluid inclusion evidence is considered to be invalid. A general discussion of the validity of fluid inclusions as samples of the ore fluids has been given earlier (Roedder, 1967a, c) and will not be reviewed here.

Entirely apart from the theories of origin of the paleoaquifer system in East Tennessee, and of the breccia, it appears inescapable that these deposits formed from hot, saline brines. Meteoric water, if
present, could only be a minor diluent for the saline brines. Thus, even if it can be shown that fresh meteoric water was responsible for the formation of the paleoquifer system (and this itself is not universally accepted), it merely provided the plumbing for a later invasion of saline brines from some other source.

Obviously the topography over the deposits was of vital importance in establishing the hydraulic gradients in the plumbing system at the time of ore deposition. There is a large uncertainty in the evaluation of this variable, ranging from essentially surface conditions (as at the time of karst development) to thick piles of thrust plates stacked on each other. The minimum depth can be determined from the vapor pressure of aqueous salt solutions at the inclusion homogenization temperatures. At a homogenization temperature of 150° C, pure water has a vapor pressure of 4.7 bars and hence would require a minimum of 47 meters of hydrostatic head of cold water (at 1.0 g/cm²) to keep it from boiling. Gases such as methane in solution would of course raise this minimum. If the water over it were heated, it would be lighter and this minimum head would have to be higher; if the solution were saline, its vapor pressure would be slightly lower, requiring a lower head. The inclusion evidence places no upper limit on the pressure at the time. However, if another, independent (and correct) thermometer were available, the difference between this temperature and the homogenization temperature would be a true “pressure correction” and would permit a valid estimate of the pressure. Such an independent thermometer would have to be very accurate. Thus the compressibility of 20 percent NaCl solution, at 150° C, is so small that even 3,300 meters of hydrostatic head on it would only cause a pressure correction of 25° C (Klevtsov and Lemmléin, 1959).

An appreciable depth of burial at the time of mineralization is indicated by the homogenization data from the near-surface veins in the Central Tennessee district. As these veins occur in rocks as young as Mississippian, lie far above the new discoveries of major stratiform ore deposits in the Knox Dolomite, and have inclusions with relatively low homogenization temperatures, it might be expected that they would represent deposition under relatively near-surface conditions. However, the 162 homogenization determinations on these veins were made on planar groups of inclusions, some of which are primary growth planes, others of which are as obviously secondary planes. Within each plane some of the cogenetic inclusions contained oil, some contained water, and some contained both fluids. (On Figure 13 these latter have been plotted on the basis of the major component.) The important point is that the oil inclusions have homogenization temperatures consistently about 10 degrees lower than the cogenetic water inclusions trapped in the same plane. These differences in homogenization temperature almost certainly are caused by differences in the pressure corrections for these two types of fluid, from gross differences in compressibility (Roedder, 1963, p. 175–177). As the pressure correction on the water solutions will be smaller, the homogenization temperatures for water inclusions are closer to the trapping temperatures. But the difference in apparent pressure correction requires an unknown but appreciable pressure at the time of mineralization, even for these deposits, formed under possibly the most shallow conditions. Once calibrated, such mixed groups of inclusions may provide us with a relatively precise measure of both pressure and temperature.

The density of the fluid or fluids involved is the single most important variable for the ore-forming fluid that must be considered in all discussions of fluid circulation, once the paleoquifer plumbing system has been established. With very few exceptions, the fluids found in the fluid inclusions all had densities of greater than 1.0 g/cm³ at the temperature of trapping. Thus pure water in equilibrium with vapor at 150° C has a density of only 0.917, but a solution of approximately 12 weight percent NaCl will have a density of 1.0 gm/cm³, and 20 percent will be up to 1.06 gm/cm³, or 1.14 at 100° C (Unterberg, 1966). The fluid in the highest temperature inclusions (170° C, from the Coy mine) had a freezing temperature corresponding to a salinity of about 13 weight percent salts. Assuming NaCl, this would yield a density of just 1.00 g/cm³ at the temperature of trapping.

The importance of these high densities in any plumbing system full of cold ground water at density 1.0 is obvious; once such saline fluids are injected, they would flow down hill, even in a full, stagnant, system, readily displacing the preexisting cold water. Mixing with the cold ground water would be minimal in laminar flow, except by diffusion at the interface and by the complex involutions of this interface implicit in the passage of such an effectively two-phase system through tortuous passages in brecciated rock. Rising gas bubbles, from below or evolved from the hot fluid, would cause local mixing, and any solution slumping of the walls could cause gross turbulence and mixing. As shown earlier (Roedder, 1967c, p. 352–353) the sudden collapse

*The notable exceptions are a group of 10 oil inclusions with very low temperatures from the Macey vein, and a group of 11 water inclusions with low temperatures from the Alcorn mine. Both of these groups were in separate planes, independent of the other oil and water inclusions plotted for these samples.*
of a cavity during ore deposition could easily yield the fast flow indicated by the loose, abraded sphalerite crystals in some vugs from East Tennessee, even though the general depositional conditions were probably very quiet, as evidenced by the frequent and gross supercooling, as well as the persistence of metastable superheated ice under negative pressures. The rather wide range of salinities found, and the minute banding in some of the ores are both believed to be a result of mixing of brines with various amounts of fresh ground water, whose composition or proportion varied over the annual cycle of the seasons at the surface.

Further verification of the generally slow rate of movement of the ore-forming fluids is found in the very thin, graded laminae of the varved breccia matrix (Kendall, 1960), and in the common asymmetry of the vein fillings throughout the East Tennessee districts that has been described by many (e.g., Callahan, 1967, p. 16), in which coarse sphalerite coats mainly the top surfaces of the breccia fragments and lines only the bottom surfaces of any but almost vertical veinslets. This would be expected if the first tiny sphalerite crystal nuclei were free to settle by gravity in a nearly static fluid. Even an asymmetric "schalenblende" could easily form this way, if the initial nucleation of sphalerite resulted in the settling of a high concentration of nuclei per unit area. The coarse ores, however, in which individual, well-developed sphalerite crystals occur at intervals along each fissure, would seem to require an exceedingly slow approach to nucleation conditions and exceedingly slow growth thereafter. The slowest approach to the precipitation of ZnS from 100° C fluids in the laboratory generally results in a milky solution, containing vast numbers of nuclei, and coarser crystals can be grown only at much higher temperatures, and very slowly, with very careful control.

Sometimes the concept of very slow flow rates is rejected on the assumption that the ore fluids would have to be very rich in metal to form an ore deposit under such conditions. However, even very slow flow rates, with very dilute fluids, can deposit large orebodies. For convenience, let us assume a flow of 1 μm/sec (8 cm/day), perpendicular to the bedding of rocks with 20 percent porosity, for 10,000 years, with only 1 ppm zinc precipitated from each unit of fluid. This would form a layer of 10 weight-percent zinc ore one meter thick in this period of time (see also Roedder, 1960, Fig. 6). The same result would be obtained in 1 million years from flow rates of only 0.8 mm/day.

The origin of the ore fluids themselves is even more speculative than the circulation patterns at the site of deposition. Igneous rocks are generally ab-
Ordovician St. George carbonate rocks in Newfoundland was examined. The inclusions, in pale yellowish sphalerite from the Newfoundland zinc mine (see Appendix, "Miscellaneous Valley zinc deposits and prospects," sample ER 69-67), were too small to run, but visual estimates of the relative bubble size, made under oil immersion, indicate that both the sphalerite and the dolomite grew at temperatures comparable to those found for East Tennessee.

The geographic and stratigraphic range, and the similarities between these many occurrences as well as the other Mississippi Valley-type deposits throughout the world are perhaps best explained by the movement of heated saline connate waters. These must, in general, be moving, however slowly, in response to hydraulic gradients established by surface topography and precipitation, if not by temperature or orogenesis. Such brines may dissolve and carry significant amounts of heavy metals (Lebedev, 1967), and may travel great distances laterally (Davidson, 1966; Roedder, 1968a). They would, of course, follow any faults, paleoquarriets, or other permeable zones they found, and slow dilution with surface waters would be expected to occur. Either this dilution or other chemical changes could actually cause the slow precipitation of the ore minerals, in large part in limited favorable horizons, and as minor mineralization throughout the stratigraphic column through which they were moving. Perhaps the major chemical problem is that of explaining the almost quantitative separation of lead from zinc, so that some deposits such as the Southeast Missouri and Laisvall, Sweden are all lead, whereas other deposits such as Friedensville and Mascot-Jefferson City (and many of the other Appalachian deposits) are all zinc.

Although the scope of the present work does not provide any details of possible changes in the nature of the ore fluids with paragenetic stage, other than the generalization that the sphalerite has formed from more saline, slightly hotter fluids than the later vug minerals, there may well have been many such changes, within the ranges of data given. These could only be delineated by much more detailed sampling and study than the available material permitted. In spite of such variations, however, the ore fluids for this entire group of deposits fell in a remarkably narrow range of temperature, and all were strongly saline—much more saline than sea water. Furthermore these ranges of salinity and temperature are fairly consistent with the data obtained from numerous other Mississippi Valley-type deposits all over the world and probably signify similarity in the processes involved in their origin.

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Mascot-Jefferson City Zinc District, Jefferson City, Tenn.

Jefferson City mine (The New Jersey Zinc Co.)
ER 65-38 Fluorite, as slightly purple cubes (plus tetrahedra) up to 5 mm, on vuggy dolomite. Collected by Heyl, June 60. Numerous inclusions show superheated ice, at temperatures as low as 9.2°C.
ER 62-26 Sphalerite, coarse crystals with dolomite lining vug. Collected by Heyl, 9/30/60, no. JC-H1-1.

Mascot no. 2 mine (American Zinc Co.)
ER 65-122 Calcite crystals from vug. Many birefringent crystal inclusions as ghosts. First visible melting at about -0.5°C. Some superheated ice. Greene no. M-1.

Young mine (American Zinc Co.)
ER 63-2 Sphalerite radial rosette ("egg") under late dolomite vug crust. Gift of Richard Douglas, 9/18/62. Shows concentric color zoning, with pale yellow euchedral crystals on surface. Some carbonate crystals completely surrounded by sphalerite. Many primary inclusions did not freeze even after overnight at -78°C.
ER 65-48 Sphalerite, 412 level south of fault ("GSA stope"). Discrete yellow crystals on lower surface of each narrow fracture opening in crackage breccia, even though steeply inclined. Individual crystals slightly zoned, with colorless core and yellowish eucleral rim, against carbonate vein filling. A few patches and bands of purple.
ER 65-115 Fluorite, 2-cm crystal from vug. Pale purple cube, with rounded, leached (?) faces. Some small sulfide crystals embedded just under surface; [and] one inclusion of black (organic?) material. Superheated ice frequently observed. Greene no. Y-1.

ER 65-117 Calcite crystal from vug. Inclusions all full of liquid, with no bubble. Most showed superheated ice in freezing runs. Greene no. Y-3.
ER 65-118 Quartz crystals from vug, some with ghosts consisting of carbonate rhombs. One shows minute cubes and octahedra (fluorite?) attached to the surface. Liquid inclusions all have strongly birefringent, in part acicular daughter crystals. Some superheated ice. Greene no. Y-4.
ER 65-119 Calcite crystal from vug, with inner colorless steep scalenohedron and outer whitish, more equant form. Some indication of possible decline in salinity and temperature between inside and outside. Green no. Y-5.

New Market mine (New Market Zinc Co.)
ER 65-39 and 39a. Sphalerite crystals in dolomite. In polished plate sphalerite is colorless, purple, orange and yellow. Some purple zones are minutely striated. Collected by Wedow.

Coy mine (American Zinc Co.)

Appendix-Sample Descriptions and Miscellaneous Inclusion Data
Note: Only those individual samples that showed some usable fluid inclusions are included here. Many other samples given me were mostly unusable. The freezing and homogenization data plotted on Figures 12 and 13 are not repeated here because of space. Only those other incidental observations on the inclusions are listed that are in some way pertinent to this paper, but generally not given in text. Samples collected personally except as indicated; major sample donors, identified below by last name only, are as follows: Helmuth Wedow, Robert A. Laurence, Allan V. Heyl and Janice L. Jolly (U. S. Geological Survey); Robert Greene (American Zinc Co.).
ENVIRONMENT OF FORMATION OF MINERAL DEPOSITS

ER 65-110 Quartz crystals from vug. Large numbers of carbonate rhombs occur in growth zones in quartz, but no usable fluid inclusions. Greene no. C-2.

ER 65-111 Sphalerite “egg” from vug, with several large primary inclusions in one clear yellow termination (Fig. 1). Greene no. C-3.

ER 65-112 Quartz crystals from vug. Some inclusions have an NaCl-2H₂O liquidus; others show superheated ice, even at -18°C. Several secondary inclusions have NaCl daughter crystals that dissolve at 78°C. Greene no. C-4.

ER 65-113 Calcite crystals from vug. Many inclusions show superheated ice. Greene no. C-5.

ER 65-114 Fluorite crystals from vug. Pale purple cubes up to 5 mm, with zones of embedded quartz and carbonate crystals outlining growth zones. Some superheated ice, particularly in inclusions from outer zones of the crystals. Greene no. C-6.

North Friends mine (American Zinc Co.)

ER 65-121 Dolomite crystals, pearly white, lining vug. Greene no. NF-2.

Copper Ridge District, North of Morristown, Tenn.

Flat Gap mine (The New Jersey Zinc Co.)


ER 65-42 Sphalerite, color banded “colloform” or “schalenblende” crust, with elongated blebs of galena (see Fig. 9). Wedow no. T 353.

ER 65-43 Sphalerite, similar. Good example of gradual change from early light to later dark, then abrupt change to light again. Wedow no. T 358.

ER 65-44 Sphalerite, similar. White, yellow, brown, and red bands all show “sawteeth” representing former euhedral druse surfaces. Wedow no. T 359.

ER 65-45 Sphalerite similar. Some calcite replacing galena blebs is thick with inclusions. One grain of rutile-filled quartz (residual from limestone) shows plane of secondary liquid CO₂ inclusions, inherited from its former metamorphic (?) environment. Wedow no. T 360.

ER 65-46 Sphalerite, coarse, relatively nonbanded yellow crystals, on very fine grained banded sphalerite, almost opaque at a thickness of 388 µm. Coarse crystals show some variation in intensity of yellow color, and a few patches of purple, banded sphalerite. Wedow no. T 361.

ER 65-47 Sphalerite, similar coarse crystals. Wedow no. T 363.

ER 69-8 Sphalerite, zoned dark olive to pale schalenblende, grading into coarser sphalerite. Inclusions poor but usable.

ER 69-8a Sphalerite and pink dolomite, same locality as above. From vugs in vein of dolomite 2–3 cm wide, lined with sphalerite crystals on lower side only. Sphalerite crystals have inclusions too poor to run, but optically similar to those in ER 69-8 (i.e., small bubbles, estimated to homogenize at 100°–150°C).

Sweetwater Barite District, East Tennessee

Ballard mine

ER 65-103 Early blue fluorite, covered with barite and green fluorite as rosettes, coated with pyrite, broken and then recemented with fine silica, limonite and colorless to purple fluorite. Fine pyrite crystals within barite have been limonitized, indicating considerable late brecciation. Solid brownish matter in some inclusions (see Fig. 5). Wedow no. T 409.

ER 65-106 Similar but with fluorite cubes in vug. Some birefringent daughter minerals present. Wedow no. T 412.

ER 65-108 Similar; bedding vein of barite and late purple fluorite filling in between barite blades. Many tiny birefringent mineral inclusions in fluorite. Some opaque wire-like inclusions in fluorite are probably co-genetic sulfides. Wedow no. T 414.

Other Barite Occurrences in Tennessee

ER 63-80 Cox prospect, near Fall Branch, Tenn. Collected by Laurence, from residuum. One of many similar prospects in area yielding fetid, dark barite. (Secrist, 1924, p. 101.) Very large evolution of gas under pressure on crushing stage; gas very soluble in kerosene but gives no visible reaction with Cd₃, solution hence is not high in H₂S. Wide range of degree of fill; some planes of secondaries show about 10 percent by volume of a second, transparent liquid.

ER 63-81 Barite prospect near Click Creek, Hawkins County, Tenn. Greenish, clear barite occurs with scalenohedral calcite but no sulfides. No odor on crushing. Secondary inclusions with small bubbles show superheated ice at +4.15 and +4.35°C; others have freezing temperatures of -0.15 ± 0.03°C. Collected by Laurence.

ER 63-82 Nuns Cove barite prospect, Sevier County, Tenn. Gray massive cleavable barite giving strong odor of petroleum and large evolution gas under pressure on crushing; gas exceedingly soluble in kerosene. Many secondary inclusions of yellow-brown petroleum(?). Some such single-phase liquid inclusions do not have vapor bubble at room temperature until after being cooled to -78°C. Collected by Laurence.

ER 69-9a Last Creek barite mine, East Tennessee. Sphalerite, with barite, pyrite, and some greenockite stains. Deep red and orange zoned sphalerite as disseminated crystals and as radial crusts (black or brown in hand specimen), including many rhombs of carbonate, and with prominent sector zoning and some “varves.”

ER 69-9b Barite from same specimen. Barite forms crusts up to 4 cm thick on breccia. Many inclusions, mostly full of liquid, but some with small bubbles; all too small to run.
ER 69-10 Fall Branch, Tenn. Barite, black, fetid. Crystals up to 10 cm, but inclusions too small to run. Some planes of secondaries are filled with liquid, others have small bubbles regularly. Obtained from W. M. Johnson, 3/29/69.


Central Tennessee District—Surface Veins

ER 69-36 Alocorn fluorite mine, Buffalo Valley, Tenn. Fluorite float. Many excellent secondary oil and water inclusions. Shows odd parallel curved "fibers" of purple color comprising purple surface zone. Many inclusions have small birefringent grain in water solution which is apparently unaffected by homogenization runs. Yellow oil melts in range -6° to +10° C; its coloration lightens above 50° C. Jolly-Heyl sample Alocorn 64-2, 6/15/64.

ER 69-39 Boatman fluorite mine, 10 miles east northeast of Lebanon, Tenn. Calcite, 10-cm crystal. Primary inclusions, outlining a ghost in crystal, contain water and dark brown oil. Jolly-Heyl sample Boat 64-203, 6/17/64.

ER 69-40 Boatman fluorite mine (see above). Fluorite, purple banded. Primary inclusions containing dark brown oil contain black and dark translucent birefringent spots of higher index of refraction. These appear to melt and diminish somewhat in size at about 70° C but never dissolve completely. Jolly-Heyl samples Boat 64-220 and Boat 64-225, 6/17/64.


ER 69-43 Oakley mine, north of Lebanon, Tenn. Sphalerite, 1 cm crystals. Many sets of parallel tubular (?) inclusions in geometric array, particularly adjacent to cracks. Some of the larger seem to be tubes open to the crack, and hence may be the result of leaching. Jolly-Heyl sample Oakley 64-114.

ER 69-44a South Oakley vein, southwest shaft, north of Lebanon, Tenn. Sphalerite, disseminated 2-mm crystals. No fluid inclusions but many isotropic cubic inclusions (flourite?). Jolly-Heyl sample Oakley 64-1, 6/17/64.

ER 69-44b Fluorite, 2-cm colorless crystal from same sample as above.


ER 69-47 Dillard vein (see above). Fluorite. Oil and water inclusions; birefringent daughter minerals in oil inclusions dissolved at about 50° C and some recrystallized on cooling. Jolly-Heyl sample Dil. 65-125.


ER 69-51 Holt vein, west of Nolensville, Tenn. Fluorite. Many large primary oil inclusions, with liquid oil of an index of refraction equal or very slightly greater than that of the fluorite (n 1.434). Some visible only from presence of bubble and yellowish color. Those with n above that of the fluorite disappeared at about 50° C (i.e., the index then matched the fluorite). One primary homogenized at 118° but was leaking (temperature too high). Jolly-Heyl sample Holt 110.

ER 69-52 Holt vein (see above). Fluorite. Two primary oil inclusions found. Jolly-Heyl sample Holt 65-203.

Central Tennessee District—Deep Ore

ER 69-65 Knox Dolomite at 1,213 feet, near Carthage, Tenn. Sphalerite, from drill cores. Orange-yellow crystals, with quartz, in breccia. Sample from Heyl.

ER 69-66 Kingsport Formation, at 2,091 feet, near Carthage, Tenn. Sphalerite with primary inclusions, from drill core. From Frank G. Snyder.

ER 70-1 Knox Dolomite at 1,363 feet, near Carthage, Tenn. Fluorite with many secondary inclusions and barite with many primaries, some with uniformly small bubbles. From drill core. Sample from Heyl.

ER 70-3 Knox Dolomite at 1,329 feet, near Carthage, Tenn. Fluorite and dolomite, from drill core. Sample from Heyl.

Central Kentucky District

Moore or E. Prospect mine


Gratz vein (across Kentucky river from Twin Valley mine)


ER 63-175 Sphalerite, disseminated zoned crystals (with light centers) in calcite. Cox property as above. Some sphalerite banded purple, yellow and orange. Primary inclusions show about 25 percent liquid at -27.8° C; at -23.7° they show about 3 percent NaCl·2H₂O, 15 percent ice, and the balance solution (no bubble). The dihydrate crystals disappear before the stable freezing point (ice-solution-vapor changing to solution-vapor) at -16.5 ± 0.2° C. Collected 1962, Heyl no. Cox 113.

Faircloth mine near Lexington, Ky.

ER 62-31 Sphalerite, red and brown, plus fluorite. One yellow-brown oil inclusion in fluorite. Most sphalerite inclusions tetrahedral in shape and very

ER 63-177 Fluorite, clear colorless cubes, from within vein. Many secondary inclusions show a highly birefringent daughter crystal (see Fig. 7). Many show and NaCl2H2O liquidus. Collected 1960, Heyl no. F-19.

ER 63-178 Fluorite, similar cubes up to 2 cm, but with a thin crust of purple color. Some superheated ice. Heyl no. Ck-311.

ER 63-179 Sphalerite, dark disseminated crystals, many with zones of finely striated purple-gray color (Fig. 10) but no usable inclusions. Heyl no. Ck-328.

Hanna Morgan mine (central part of district)

ER 63-181 Fluorite, 5 cm colorless cubes. Most inclusions show NaCl2H2O crystals in range −24° C to −22° C, but have a normal ice liquidus. Fairly regular progression from −17.5° C in core of crystal to about −16° C at rim. One yellow oil inclusion. Collected 1943 by W. R. Thurston, U.S.G.S.; Heyl no. TL 43A.

Purdy vein, north of Millersburg, Ky.

ER 63-182 Sphalerite, euhedral crystals with segmented cores (yellowish and blue-gray striated) and red exteriors. High in germanium (0.1%). Some inclusions have (accidental?) enclosures of highly birefringent carbonate (?) crystals.

Timberville, Virginia, Zinc District

ER 62-27 Sphalerite, cleavable, massive, coarse grained, dark brown, with individual crystals up to 5 cm. From massive 5-foot layer of ore, north end, 5th level, Bowers-Campbell mine. Collected January 1959, Heyl no. V-25A.

ER 63-404 Sphalerite crystals from vug with dolomite in massive ore, from near sump in Bowers-Campbell mine, between 7th and 8th levels. Euhedral sphalerite crystals contain good primary inclusions. All bubbles move up a thermal gradient.

ER 63-407 Sphalerite, green, coarsely crystalline high grade ore from ore pocket of Timberville mine. Primary inclusions small but usable. Some bubbles move up a thermal gradient, others move down.

Miscellaneous “Valley Zinc” Deposits and Prospects

Almeda mine, near Bloomsburg, Pa.


Bamford mine, near Lancaster, Pa.

ER 62-33 Dark sphalerite and quartz. Collected by Robert Young; Heyl no. P-22.

Birmingham mine, near Birmingham, east of Altoona, Pa.


Linville, North Carolina (North Cove prospect)


Newfoundland zinc mine, just east of Daniel’s Harbor, Northwest Peninsula, Newfoundland, Canada.

ER 69-67 Sphalerite, pale yellow high grade ore showing sphalerite grains up to 4 mm, birefringence practically zero, with white dolomite in brecciated, dolomitized Lower Ordovician St. George carbonate rocks. Euhedral quartz crystals embedded in sphalerite contain large sphalerite inclusions, and there are sphalerite inclusions in the partially replaced remnants of dolomite crystals. Dolomite crystals contain many tiny primary inclusions with small bubbles, some in fast motion. Some grains of sphalerite have many tiny inclusions, possibly pseudo-secondary. Sample provided through the courtesy of A. C. Freeze, Cominco Ltd.


Friedensville District, Friedensville, Pa.

ER 62-25 Sphalerite, as late yellow crystals, with moderate birefringence and patchy extinction, on dolomite and quartz. Sphalerite crystals enclose many rhombs of carbonate and contain several sets of parallel tubular (?) inclusions in geometric array, many <1 μm in diameter. Collected before 1940 by Heyl, his no. H-1.

ER 61-52 Quartz, crystals from fracture filling, associated with late yellow sphalerite, from northeast end of Ueberoth pit, Friedensville zinc mine.

Quartz Crystals from Geodes

ER 65-5 Quartz crystals, from shoreline of Douglas Lake, Tenn., just south of dam. Presumably weathered out of geodes at contact of Lenoir Lime-stone with Knox Dolomite. Some gaseous and liquid organic inclusions, and slightly saline water inclusions (freezing temperature about −2.5°).

ER 65-37 Quartz crystals from geodes (formerly gypsum-anhydrite nodules?) in Salem Limestone, near Boone Hollow, just off Kentucky highway 1638, about 1 mile west of Muldraugh, Ky., in Meade County (Rock Haven quadrangle). Collected by C. F. Withington. Inclusions poor; one showed metastable superheated ice up to −1.8° C.

ER 65-38 Same locality and description as ER 65-37 above. Four runnably primary inclusions gave freezing temperatures of −19.5°, −19°, −18°, and −0.6° C (superheated ice).

ER 62-23 Same locality and description as ER 65-37 above. Very small (primary or secondary?) inclusions show either zero or about 1.5 volume percent gas, corresponding to homogenization at about 60° C. Many inclusions show superheated ice at +0.5 to +3.5° C; one froze at −1.22° C; and one presumed primary liquid as a phase at −22.82 ± 0.02° C, and NaCl2H2O at about −18° C.