PHOTOATLAS
of Inclusions in Gemstones

with the cooperation of

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Origin of Fluid Inclusions in Gemstones

Most people are surprised to learn that almost all minerals, including gems, contain fluid inclusions. The largest such samples are the special type of hollow agate nodules called “enhydros”, that are partly filled with water and can even make a splashing sound when turned quickly. Visitors to one national gem and mineral show were treated to such splashing sound, highly amplified on the public address system! Liquid inclusions are seldom large enough, however, to be obvious to the untrained eye, and hence transparent specimens containing large liquid inclusions with vapour bubbles that can move, like the bubble in a carpenter’s level, are highly prized by collectors as “bubble crystals”. For every such large inclusion in nature there are untold billions of very small fluid inclusions, however, only visible with a microscope.

It is not too difficult to visualize a process whereby a crystal growing in a rock deep down in the earth could enclose other crystals within itself, but why should a crystal enclose a fluid? The answer lies in two features common to most natural crystal growth: most have actually grown from one or another type of fluid and most are imperfect. With relatively few exceptions, minerals have grown from either a hot watery solution or an even hotter fluid rock melt. Examples of the former type of fluid are not familiar to us, as they can exist only under moderate to high pressures, deep in the earth. Many of the resulting crystals, and particularly gemstones, have grown from such fluids, on the walls of cavities.

Gems are normally crystals containing relatively few imperfections. Most of the imperfections or defects that make the difference between a ruby crystal selling for $5 as a mineral specimen and another, much smaller ruby crystal, of the same colour, worth $50,000 as a gem are inclusions of foreign substances, solid, liquid or vapour. The mineral specimen may have billions of such inclusions in each cubic centimeter, whereas the gem will have perhaps thou- sands or even millions. Solid inclusions are covered in other chapters; here only fluid inclusions are discussed, i.e., those that were fluid at the time they were enclosed. Even if they have formed a hard glass as they cooled, if the captured material was a fluid at the time of trapping, the result is called a fluid inclusion. Most of the scientifically valuable information that can be obtained from fluid inclusions stems from studies of the nature and behaviour of these original hot fluids. Relatively few detailed studies have been made of fluid inclusions specifically in gemstones, but many studies have been made of the inclusions in non-gem-grade crystals of gem minerals from localities that have produced gemstones (e.g., Roedder, 1982). These studies are not always recognizable as being related to gems. Thus, although the term “gems” is not mentioned in most studies of fluid inclusions in the mineral tourmaline, many of the geological occurrences in which this mineral is sufficiently transparent to permit inclusion studies (e.g. Roedder, 1972) are potential or actual sites for gem tourmaline.

Although fluid inclusions might be found in any gemstone, most of the detailed studies of fluid inclusions in gemstones have been on relatively few species, particularly peridot, quartz (citrine, smoky, etc.), tourmaline, topaz, sapphire and emerald (Roedder, 1982). The abundant solid inclusions in diamonds have been extensively studied and provide exceedingly valuable data on the conditions of diamond formation (see Meyer, this volume), but there have been no verified reports of liquid inclusions in diamond containing water, carbon dioxide, or otherwise. These various studies make it obvious that a wealth of useful data can be obtained by simple microscopy. Gemmologists the world over should become aware of what fluid inclusions look like and what they can tell us. If nothing else, knowledge of the fascinating data available from fluid inclusions might help to sell what
would otherwise be considered merely a flawed stone. The crystal imperfections that result in the enclosure of fluid inclusions may come about by too rapid growth. Under such conditions the resulting crystal may develop feathery, dendritic habits; later, slower growth may cover these porous areas with solid, relatively perfect growth, thereby enclosing some of the fluid from which the crystal was growing. Such imperfections are all too common in the commercial growth of synthetic crystals for gems or technical applications. When growth (and hence production) rates are pushed too high, nature was not concerned with maximizing production yield and may well have used vastly less efficient processes of material transfer to the growing crystal over much longer times. Thus, some mineralogical and geological evidence suggests that at least a few and perhaps most of the large crystals seen in mineral collections grew over much longer periods of time, tens of thousands of years or more. Gemstones, because they are even more perfect crystals, may have grown even more slowly. At present, we really have no way of knowing.

A variety of other scenarios leading to the envelopment of fluid inclusions during crystal growth into a free fluid—covering of earlier formed etch pits, sub-parallel growth on adjacent growth spirals, enclosure of some fluid as the host crystal grows around a solid object on the surface, etc.—are all well known in the synthetic crystal industry and have natural counterparts.

Most gem sapphires, rubies and spinels, and some emeralds, have formed in metamorphic (recrystallized) rocks. Hence, instead of growing as free crystals, protruding into the fluid from which they grew, they have grown in an essentially solid medium, by migration of some or all of the constituents to the surface of the crystal nucleus via diffusion through the other crystals, along grain boundaries, or through a fluid film on the grain boundaries. The space for the enlarge-
(page 84). The plane may be curved, if the original fracture was conchoidal, or flat, if it was a smooth cleavage fracture. Most of the planar groups of inclusions in gemstones, described by such terms as finger prints, feathers, etc., represent early stages in this process.

Many gemstones contain both planes of secondary inclusions, outlining former fractures, and, unhealed cracks. One may ask why so many cracks in natural crystals have not healed, as they have all been immersed in ground water beneath the surface of the earth for at least many thousands (or millions) of years before being collected. Healing will take place only if a fluid is present in which the crystal is at least slightly soluble, thus permitting dissolution and redeposition of the mineral along the crack. Gem minerals are essentially insoluble in cold ground-water, but deep in the earth, where temperatures are higher and where various salts are in solution in the water, the solubilities, although perhaps vanishingly small, are still adequate to permit the cracks to heal in the large amounts of time available. As a result, secondary inclusions in gems; though they may have been enclosed at lower temperatures and pressures than were the primary inclusions, will generally not have been captured at surface temperatures. Normally, the fluid that becomes locked in a fluid inclusion is a single homogeneous phase at the time of seclusion. However, in rare situations, the fluid may be heterogeneous, i.e. consist of several phases. Thus it may be a suspension of small solid particles in liquid. More commonly, a mixture of bubbles of gas or vapour in liquid are trapped; in this case each inclusion will catch a random ratio of gas to liquid. Similarly, if globules of oil are present in the water-rich fluid surrounding the crystal, these two immiscible fluids may be retained in the inclusions in random ratio. Still other more unusual types of fluid inclusions are known (e.g., exsolution inclusions), but are beyond the scope of this chapter.

**Evolution of fluid inclusions after trapping**

Most inclusions are not the same now as when they were captured. They have changed in many ways, and to make full use of fluid inclusions as clues to unravel the sequence of geological events that a sample has undergone, it is essential that these changes be recognized and understood. The changes are of two types: phase changes, and physical changes.

Phase changes refer to the formation of new daughter phases, i.e., substances (gases, liquids or solids) that were not present at enclosure. Such phase changes provide most of the data that are known on the pressure, temperature and composition of the fluids from which the host crystals grow, and hence most of our knowledge of the origin of gemstones (and ore deposits).

Most fluid inclusions contain a bubble in the liquid. This gas bubble (i.e. vapour phase) may move under the influence of gravity or a thermal gradient, and if small enough (less than several micrometers), may be in constant motion that is mistakenly called «Brownian» motion. Relatively few inclusions are without this daughter phase, and as this vapour normally is a more or less spherical bubble (unless constricted by the walls), the too common use of the term «bubbles» for fluid inclusions in general is misleading and should be avoided. Although, as mentioned above, a small number of inclusions give evidence of embracement of heterogeneous systems that is of considerable value in understanding the conditions of trapping, most minerals from gem (and ore) deposits commonly show millions of inclusions per cubic centimeter, every one a simple two-phase system of liquid and gas. These two phases result from the fact that we are examining the inclusions at a lower temperature than that at which they were sealed off. As the coefficient of thermal expansion for most minerals is one to three orders of magnitude lower than that for water solutions and CO₂ on cooling from the temperature of entrapping
to room temperature, the container for the inclusion (the hot crystal) shrinks much less than the fluid it contains, yielding a vapour (shrinkage) bubble.

The bubble may be very-low-pressure water vapour, if a water solution of non-volatile solutes was enveloped, or nearly a vacuum, if a silicate melt without volatiles was caught. Any gaseous species present (e.g. CO₂, CH₄) will generally partition strongly into this vapour bubble, so it may be a dense fluid under high pressure (tens or hundreds of atmospheres at room temperature). If the vapour bubble is a dense CO₂-rich fluid, on cooling it also may split again into two fluids, liquid CO₂ and gaseous CO₂.

In 1823 the Scottish physicist Sir David Brewster described a "remarkable new fluid found in the cavities of rocks." By careful microscopy he found its index of refraction to be less than that of water, its thermal coefficient of expansion to be 30 times that of water, and he noted that it did not mix with water or wet the walls of the cavities. Later workers found that some bubbles of this liquid in otherwise water-filled inclusions could be made to sink or float in the water by adjusting the temperature. Identification of the remarkable fluid had to wait 46 years until the discovery that carbon dioxide can be liquefied. Then it became apparent that the properties Brewster had observed were precisely those of liquid carbon dioxide. The material has since been found in many crystals from certain types of geological environment, and is particularly common in inclusions from gemstones.

Considerable pressure is required to liquefy carbon dioxide: at 31 degrees C., the highest temperature at which it can be liquefied at all, the pressure must be 73 atmospheres, or more than 1,000 pounds per square inch. The magnitude of such pressures in carbon dioxide inclusions can be verified, using an adaptation of a method devised by the French mineralogist Georges Deichia. With the help of a vernier micrometer eyepiece on the microscope, the diameter, and therefore the volume, of the bubble of liquid carbon dioxide can be estimated in cubic micrometers (trillionths of a cubic centimeter). Then the sample is slowly crushed between heavy glass plates immersed in oil. During the process it is under continuous observation with the microscope, lighted from below through the plates. When a crack reaches the inclusion, the liquid carbon dioxide vaporizes and expands tremendously—nearly 400 volumes. The resulting bubble is big enough to measure quite accurately. As little as 10⁻¹⁴ (a hundred-trillionth) gram of liquid carbon dioxide will form a gas bubble several micrometers in diameter, readily visible in the microscope.

At high temperatures water and carbon dioxide mix in all proportions, and such mixtures are sometimes trapped as fluid inclusions. At lower temperatures the mutual solubilities are very low, and an inclusion that was originally a homogeneous, mixed fluid separates into two immiscible fluids, one rich in carbon dioxide, the other rich in water. On dropping still further to some temperature below the critical 31 degrees, the carbon dioxide phase separates into a liquid and a gas, giving a total of three phases. Hundreds of inclusions in a healed crack will all be divided in the same proportions, meaning that they must have been trapped as a homogeneous fluid at high temperature. On reheating they all pass in reverse through the phase changes described above. When they freeze, these inclusions develop large, almost invisible crystals of the interesting and improbable-looking compound 8CO₂·46H₂O, in which 46 water molecules form a kind of cage with cells for eight carbon dioxide molecules. Since these crystals are usually stable below about 10 degrees, they probably occur naturally in mixed inclusions of water and carbon dioxide in the rocks of cold climates.

In a few crystals the author has found different ratios of two immiscible fluids in neighbouring
primary or secondary inclusions. This is clear evidence that the two fluids were present as distinct phases while the crystals were growing. For example, there were minute bubbles of highly compressed carbon dioxide in the slightly salty water solution from which a mercury deposit in Alaska formed, and they were trapped in various ratios with the water. Globules of yellow petroleum are common in the strongly saline brines trapped as both primary and secondary fluid inclusions in the Kentucky-Illinois fluorite and zinc ores. Globules of dense organic gas were present in the liquid that gave rise to the well-known and beautifully clear quartz crystals from Herkimer, N.Y. Although generally trapped along with some brine, on occasion some gas stuck to the surface of the growing crystal and was enclosed without brine.

Not all the bubbles that are found in inclusions consist of carbon dioxide. Most of them, in fact, are water vapour. These minute gaseous cavities make possible one of the most intriguing applications of inclusions to geological problems: they serve as recording thermometers, registering ancient temperatures.

In 1858 the English geologist Henry Clifton Sorby suggested that vapour bubbles result from shrinkage of the liquid in originally full inclusions that were trapped at high temperatures and then cooled. A simple inspection of the bubbles in various samples provides qualitative confirmation of the idea. Inclusions in rocks known from independent geological evidence to have formed at relatively low temperatures have small bubbles or none at all; inclusions in rocks formed at high temperatures have large bubbles. Sorby went further than this. He reversed the process of shrinkage by warming inclusions gradually, watching them under the microscope. Eventually the bubbles disappeared. The temperature at which each one vanished—the “filling” temperature—he took to be the temperature at which the inclusion had originally been trapped millions or even billions of years ago. Some minerals have primary inclusions with large bubbles and secondary inclusions with small ones, indicating that the temperature dropped after crystallization; many crystals with distinct growth zones show lower filling temperatures for primary inclusions in the outer, last-formed zones than for those in the core. Crystals of daughter minerals present in inclusions dissolve on heating and thus may also be used as geological thermometers, giving minimum temperatures of trapping.

Sorby’s experiment gives the true temperature of formation only if the liquid was at its boiling point when it was trapped. In that case the slightest shrinkage would have produced some vapour. If, however, the liquid was under more than sufficient pressure to prevent its boiling, it would have had to cool enough to relieve the excess pressure before it could begin to shrink and form vapour. Accordingly the temperature at which the vapour disappears on warming would be lower than the trapping temperature. From other geological evidence it is often possible to estimate the depth at which the rock sample was formed and therefore the pressure on the liquid. Where this is known, the apparent filling temperature can be corrected to give the true value. There is some evidence that occasionally fluid may leak into or out of inclusions, and that the recrystallization of inclusions with the passage of time may sometimes result in anomalous filling temperatures, but generally these situations can be recognized. One particularly useful check on the whole procedure comes out of studies of the inclusions in synthetic crystals, such as quartz, made under known high temperatures and pressures. These inclusions fill at the correct temperatures.

At room temperature the vapour pressure of water is very low and so a vapour bubble in an inclusion is practically a vacuum, like the “bubble” of mercury vapour at the top of a mercury baro-
meter. This was first discovered by Davy, who pierced some large inclusions while the crystals were immersed in oil and found that the oil was drawn in and the bubbles contracted greatly. In many inclusions on which the author has performed the same experiment the bubbles disappeared completely and instantaneously, indicating that they must have contained considerably less than $10^{-14}$ gram of noncondensable gases—less than a billion molecules.

When an inclusion with a bubble is heated, the pressure in it—the vapour pressure of the water solution—increases rapidly, reaching one atmosphere at 100 degrees C., 15 atmospheres at 200 degrees and 218 atmospheres at 374 degrees, the critical temperature above which water cannot exist as a liquid. The build-up of pressure in large inclusions, or in planes of many small ones, can cause a series of little explosions that make the sample decrepitate, or explode. If the more compressible gas bubble disappears before the critical temperature is reached, the pressure increases even faster and decrepitation becomes more probable.

This generally occurs at temperatures below the critical point for pure CO$_2$, 31.1 °C, because deviations from the critical density, and most common impurity gases, such as N$_2$ or CH$_4$, all depress the homogenization temperature.

When several different fluids, such as silicate melt, water solution, liquid CO$_2$, oil or gas, occur in the same inclusion, they usually assume positions of minimum surface energy. The normal sequence in which these fluids are found, which provides some assistance in phase identification, is as follows (from outside of the inclusion): host crystal-melt (glass)-H$_2$O-CO$_2$ or organic-phase gas-bubble. Thus when melt (now glass) was present, it was in contact with the walls, i.e., it wetted them, and any other fluids, such as "vapour" (essentially a vacuum bubble), or liquid water or CO$_2$ occur as bubbles within the glass rather than wetting the walls. Where water solution exists together or with immiscible CO$_2$ fluid (liquid or gaseous), the globule of CO$_2$ always occurs within the water solution, which in turn wets the walls and fills all minor re-entrants in the cavity. When a gas bubble occurs with liquid water and liquid CO$_2$, the bubble is always completely surrounded by the CO$_2$ liquid, an observation first made many years before the true nature of liquid CO$_2$ became known from studies in the laboratory. This simple fact causes considerable difficulty in recognizing the presence of small amounts (<~10 volume percent) of liquid CO$_2$ phase, unless the inclusions are strongly flattened.

The same relationship is shown by vapour bubbles in inclusions containing water and an immiscible organic fluid. In these, the vapour bubble nearly always occurs in the organic phase, which in turn is generally free from the walls. Only rarely (in some fluorites) does an oil phase preferentially wet the walls of an otherwise brine-filled inclusion.

At the time of trapping, the fluid in any inclusion is almost certainly saturated with respect to the host crystal, and hence, upon cooling, should precipitate some of that material. Such precipitation normally just crystallizes on the walls of the inclusion. The solubilities of the host minerals are so small in most geological environments that such crystallization cannot be detected. However, the originally homogeneous fluids sealed off in inclusions may become saturated with respect to solid phases other than the host mineral and nucleate new crystals of these phases, called daughter minerals. By far the most common of these is halite (NaCl), but many other minerals, sometimes rare or even new, have been found as daughter crystals. Most inclusions having daughter minerals contain only a single crystal of each daughter phase, unless it has a low solubility in the liquid phase. As the daughter minerals are part of the composition of the original homogeneous fluid, their identifica-
tion and measurement provides important information on the composition of the fluid that was locked in. The many available methods, mainly based on petrographic measurements or thermal behaviour, are summarized elsewhere (Roedder, 1972, 1984).

In addition to phase changes, most fluid inclusions have undergone physical changes, such as in shape, position, volume or contents. Such changes may not be as obvious as the phase changes, but are of great importance in interpreting inclusion data. Only a few of the major points can be covered here.

Many, perhaps most, inclusions do not now have the shape they had at the instant of imprisonment. As originally trapped, many fluid inclusions have relatively large surface areas, for example, narrow healed cracks, and the long tubes characteristic of the primary inclusions in some minerals. If the host mineral is at all soluble in this fluid, processes of recrystallization, generally termed necking down, immediately start to reduce the high surface energy of the system. Because the distances involved are small, diffusion is not a major rate-limiting step, but apparently the solubility of the host mineral is a major limitation. Re-entrants in the walls become filled with material dissolved from the main part of the walls, so that the whole inclusion may coalesce into a single, more equant cavity. Some elongated inclusions gradually develop bulges separated by thin necks, which eventually become sealed off. The net result of such necking down is the formation of several smaller inclusions that have a smaller total amount of surface energy.

Necking down makes no difference to the total composition of the system, but if any phase change has occurred before the necking down or coalescence, the resulting new inclusions may have grossly different compositions and densities. This possibility is of considerable significance to geological thermometry and barometry based on inclusion-filling temperatures because the single gas bubble may be embraced in one of the new inclusions and the other new inclusions may then nucleate their own gas bubbles with further drop in temperature. If a daughter mineral has formed before or during necking down, it may be left behind, completely surrounded by the host mineral, but with a nearby fluid inclusion that lacks that particular daughter mineral. Quartz crystals from the Colombian emerald mines frequently show sharp cubes of halite (NaCl), completely enclosed in the host quartz. A few such cubes show very thin tubes, sometimes almost at the limit of resolution of the microscope, connecting them with a liquid inclusion many micrometers away.

Regardless of the original shape, if the host mineral has any finite solubility in the fluid of the inclusion under the conditions at which it is held after trapping, recrystallization generally occurs, and hence most inclusions now have different shapes than when originally imprisoned. The final shape assumed by those fluid inclusions in which time and composition have permitted recrystallization may be either smoothly rounded and globular, or faceted negative crystals. The balance between two competing but exceedingly weak forces determines which form will have the lowest free energy, i.e. the smallest amount of surface (spherical), or the lowest energy surfaces (negative crystal faces).

Inclusions may change their position within the host crystal in response to gradients in temperature, stress or other variables, but, for several reasons, such changes are uncommon in gemstones and need not be discussed here. Changes in volume or contents are more pertinent to gemmology. If an inclusion changes in volume, shrinking (or expanding) from external (or internal) pressure, inclusion data on the pressure and temperature of trapping will be erroneous. Minor volume changes in the past may
leaving no visible evidence, but major changes are
plainly evident.

Some of the various geological processes that
gemstones have undergone result in major
changes in external pressure. Thus, a Hawaiian
peridot crystal, formed at 5,000–10,000 atm
pressures far below the volcano, may be
erupted into the surface at one atmosphere ex-
ternal pressure. After eruption, while still hot, the
internal pressure in a CO₂ inclusion in such a per-
dot would still be 5,000–10,000 atm. Under such
a pressure difference, the peridot may crack,
permitting the CO₂ to expand. In this process,
called decrepitation, the crack generally extends
to the edge of the crystal, resulting in complete
emptying of the inclusion. In some gemstones,
however, the crack may extend only partway out
into the host crystal (i.e. partial decrepitation),
just far enough to relieve some of the pressure
difference. This crack may subsequently heal,
yielding a plane of tiny secondary inclusions
around the large original inclusion. Such planes
may appear as halos of specks, fingerprints, etc.,
depending upon how the crack-healing process
proceeded. Such changes in volume from partial
or complete decrepitation are fairly obvious.

If a solid inclusion is ensnared within a crystal
of another substance having different thermal
expansion characteristics, the host crystal will be
put in stress during cooling and may crack. If
only the two solids are present, these fractures
in the host will remain as open cracks. However,
commonly some fluid, silicate melt, dense CO₂,
or aqueous solution is trapped with the original
solid inclusion as a thin film between the inclu-
sion and host. This fluid can move into the new
fracture and cause recrystallization to form the
planar halos of tiny secondary inclusions of fluid
that are called “snowstars” (in aquamarine), “lily
pads” (in peridot), “rosettes” (in Thai-ruby), etc.,
when they occur in gemstones.

Under some rather special conditions, some
constituents may leak out of (or into) an inclu-
sion without changes in inclusion volume. Ev-
dence for such leakage, in or out, has been rec-
novated, but is relatively rare and of little concern
to gemmology.

Significance of fluid inclusions in gemstones
No crystal is truly perfect; all contain imperfec-
tions of various sizes. Although fluid inclusions
in a gem are imperfections in a material that is
prized mostly for its perfection of crystalization,
they are not completely negative features. The
study of inclusions in gemstones can provide
valuable information on the environment of for-
mation of the gems or their subsequent geologi-
cal history. Such information may be useful in
many ways, such as: 1) in the search for gem de-
posits; 2) in the identification of gemstones and
recognition of their sources; 3) in the identifica-
tion of a given stone; 4) in the guidance of at-
ttempts at synthesis; and 5) in the recognition of
natural vs synthetic stones.

The selection of a region, locality or specific
spot for prospecting for any given natural de-
posit, be it metals or gems, is usually based on
some degree of knowledge of the likelihood of
success in the area. Although some deposits
have been found by pure chance, selection
based on knowledge is obviously more effect-
tive, and its effectiveness is more or less directly
dependent upon the amount, nature and particu-
larly the validity of the available knowledge. Suc-
cess comes more often to those who, from the
voluminous lore of gems, can recognize and use
those specific facts that are actually valid and
helpful in prospecting. Many of these facts are
purely empirical and are based on the observa-
tions of generations of keen miners. Thoughtful
miners have frequently found useful signs—
commonly associated minerals in stream grav-
els, colour or texture differences of the host rock,
variations in the nature of the soil, etc. These
signs should never be ignored simply because a
scientific raison d’être has not been established.
Obviously, however, the more that is known about the nature of the process involved in the formation of any given type of deposit, the more intelligent can be the selection of places to prospect or of ways to search for extensions of a given deposit.

Fluid inclusions do not provide a panacea, but do provide some insights into the nature of the processes involved which can, in theory if not in practice, put the mining of known deposits and the exploration for new deposits on a more rational basis.

Over 30 years ago it was shown that the shape and nature of the inclusions, both the liquid and, more particularly, the solid types, can be helpful in the identification of unknown stones and are an exceedingly important factor (along with colour, etc.) in deciding upon the source of a given gem (Gübelin, 1953). Thus the presence of cubes of halite along with liquid and vapour in the inclusions in an emerald has long been considered definitive of a Colombian source for the stone. Although this is generally true, recent work has shown that emeralds from some other localities also have such cubic (presumably halite) daughter crystals. This is not surprising, as halite is by far the most common daughter crystal and is present in inclusions from many geological environments.

Because fluid inclusions contain an assortment of ions other than just those of the host mineral, the ratios of these various trace elements in the fluids probably will vary widely from locality to locality. Such variations might well be much larger than those shown by the trace elements in the gems themselves, because these latter elements are limited by the crystal chemistry of the host structure. It may be predicted that sometime in the future, when analytical procedures are improved, non-destructive analysis of the soluble constituents in fluid inclusions will provide a “fingerprint” method to characterize the locality of origin for a given gem.

The fluid inclusions within a given gemstone can be recorded photographically and are obviously just as useful in verification and recognition of stolen stones as any other internal characteristic that cannot be easily eliminated by slight recutting.

The fluids that have been used in synthesis of most gemstones in the laboratory are usually quite unlike those used by Nature. As a result, the nature of the fluid inclusions can provide the critical evidence to distinguish between natural gems and synthetic stones. Thus, there is a large difference between the low-pressure gas-bubble inclusions in a synthetic boule-grown sapphire and the dense, high-pressure CO₂ inclusions in some natural sapphires. A similarly large difference exists between the fused tungstate-melt inclusions in a synthetic flux-grown emerald and the NaCl-H₂O-CO₂ inclusions found in some natural ones.

Unfortunately, in growing gemstones, Nature does not normally use the pressure-temperature-composition combinations that yield the “rapid” growth necessary for economic synthesis of gemstones by man. However, some of the all-too-frequent problems in the control of cracking and other imperfections of synthetic materials, and particularly in control of colour, might be helped by study of the fluids used by Nature. Caution is necessary in the use of the nature of the fluid inclusions in gemstones to distinguish between natural and synthetic, however, since such knowledge can be self-defeating. As long as a large price differential exists between the synthetic and the natural, knowledge of the characteristic inclusions in the latter may well be used to set-up synthesis conditions which, although perhaps not yielding high growth rates, may yield synthetic stones containing fluid inclusions that label them “natural”.

E. Roedder
Topaz plate from Nigera, between crossed polars. The fluid inclusions do not contain green liquid, but merely change the apparent thickness of the topaz along the light path, and hence the interference colour seen. Width of photo 2 mm.

Photomicrograph E. Roedder, US Geol. Survey, Reston, VA, USA

Oil inclusions in fluorite from Illinois in which the original homogeneous yellow oil filling has undergone changes since becoming trapped in the fluorite. On cooling a shrinkage bubble formed in each inclusion (dark circle with bright centre). During the subsequent millions of years, the oil has undergone degradation to form two immiscible hydrocarbon phases, a dark brown globular mass that is presumably a bitumen, and a light yellow fluid that flashes into vapour on opening to the atmosphere. The bitumen phase preferentially wets certain crystallographic planes on the inclusion walls, yielding the intricate pattern. Width of photo 0.75 mm.

Photomicrograph E. Roedder, US Geol. Survey, Reston, VA, USA

Photomicrograph of a plane of pseudosecondary inclusions on a (111) cleavage fracture in a pale-coloured fluorite crystal from a vug, which sealed off an immiscible mixture of liquid oil (yellow) and brine (colourless). Recrystallization of the fluorite walls of the fracture to isolate individual inclusions occurred almost entirely via the brine. As a result, the inclusions of brine have become three-dimensional and are lined with negative crystal faces, whereas the yellow oil inclusions are still flat and are almost two-dimensional. In addition, the index of refraction of the oil is much closer to that of fluorite. Small irregular masses of dark-brown birefringent matter have formed in the oil phase since capture, and the round gas bubbles (probably methane, under pressure) characteristically occur in the oil rather than in the brine, where both phases are present. Area of photograph is 1 x 1.2 mm. Plain transmitted light. Sample from Hill mine, Cave-in-Rock, southern Illinois fluorite-zinc district.

Photomicrograph E. Roedder, US Geol. Survey, Reston, VA, USA
Colour-zoned fluorite crystal from Illinois that grew upwards as shown here. After the lower zone formed, irregularities in the crystal growth resulted in a series of re-entrants in the crystal surface, which were subsequently sealed over, trapping fluid inclusions of the hot brine from which the crystal grew. Height of photo 1.75 mm.

Photomicrograph E. Roedder, US Geol. Survey, Reston VA, USA

Right

Photomicrograph of a large primary inclusion in zoned purple fluorite crystal from a vug. The crystal grew from bottom to top as a series of subparallel cubic projections, two of which were slightly inclined to each other. These two finally met, embracing a wedge-shaped mass of fluid (a strong brine) as a fluid inclusion. A gas bubble (dark) formed in the thickest part of the inclusion due to shrinkage caused by subsequent cooling. Although not apparent in this two-dimensional view, the bubble comprises only about five percent of the volume of the inclusion. Some of the coloured growth bands are outlined by tiny primary inclusions. A healed cleavage fracture is revealed by the plane of pseudosecondary inclusions in focus at right, which extend to what was the edge of the crystal at the time of fracturing. Two inclined planes of secondary inclusions cut across the crystal from lower right to upper left; one of these intersects the large primary inclusion and probably caused replacement of the fluid in it with new fluid. Area of photograph 2.7 x 3.4 mm, plain transmitted light. Sample from Mill mine, Cave-in-Rock, southern Illinois fluorite-zinc district.

Photomicrograph E. Roedder, US Geol. Survey, Reston VA, USA
Photomicrograph of a cleavage fragment of topaz in transmitted light between crossed polars, showing large amounts of daughter minerals (unidentified) in presumably pseudosecondary inclusions. Most of the daughter minerals are birefringent, although some are so fine-grained that their birefringence is not visible here. A few are visibly isotropic, particularly in the large inclusion, where they abut the vapour bubble, making it angular. The different amounts and kinds of daughter minerals in the several inclusions, and the different volume percentages of vapour phase, indicate that at least some of these inclusions were probably interconnected in the past, and recrystallization (necking down) has resulted in isolating material of diverse composition in different inclusions. Occasionally thin tubes can be seen still connecting such inclusions. Area of photograph 0.8 x 1.1 mm. USNM sample 90595, Rukuba tin mine, Nigeria.

Photomicrograph E. Roedder, US Geol. Survey, Reston, VA, USA

Right

Photomicrograph of a multiply-zoned sphalerite crystal, showing the relation of pseudosecondary inclusions to crystal zoning. The red and yellow colour bands are not growth zones. The colour bands reflect differences in composition between adjacent twin bands, presumably due to preferential uptake of minor constituents such as iron on specific growing crystal faces. The crystal grew from left to right, and successive growth stages are revealed by the nearly vertical hazy grey zones of minute primary inclusions. Fracturing occurred at some time after the growth of this part of the crystal. Healing of a cleavage fracture that was parallel to the plane of this section resulted in the imprisonment of large numbers of thin flat presumably pseudosecondary inclusions, each of which now has a gas bubble (13 volume percent) and several different tiny daughter minerals. One of these is apparently quartz; in the large central inclusion it can be seen as a small prism, holding the bubble away from the wall (lower left edge of bubble). Recrystallization of sphalerite around these inclusions has been controlled in part by the compositional (and twin) zones to yield straight segments in the inclusion borders, and in many areas in this sample, to

yield preferential occurrence along certain compositional zones. Area of photograph is 5.5 x 7 mm; photograph taken in plain transmitted light. USNM specimen R577, Cananea, Mexico.

Photomicrograph E. Roedder, US Geol. Survey, Reston, VA, USA
This Sri Lankan sapphire is host to a delicately aligned group of primary negative crystals, each containing only a gaseous phase. Such fluid inclusions containing only one phase of matter are called monophase or single-phase fluid inclusions. Darkfield and oblique illumination. 50x.

Top left
Photomicrograph of cleavage fragment of topaz in transmitted light between crossed polars, showing daughter crystals in sharply faceted negative crystal inclusion. One faint isotropic octahedral daughter crystal holds the bubble away from the inclusion wall and two other isotropic daughter crystals are faintly visible in the other end, adjacent to a group of smaller birefringent daughter crystals. Area of photograph 0.05 x 0.08 mm. USNM sample 96595. Rukubat tin mine, Nigeria.

Photomicrograph E. Roedder. US Geol. Survey, Reston, VA. USA

Centre left
The same topaz from Nigeria as above shows two inclusions, each with a bubble and daughter crystals, but with different gas/liquid ratios, connected by a thin tube. If this necking-down continued to completion, and sealed off the tube, the resulting two inclusions would give erroneous homogenization temperatures 0.05 x 0.08 mm.

Photomicrograph E. Roedder. US Geol. Survey, Reston, VA. USA

Bottom left
A complex three-phase inclusion testifies to the natural hydrothermal origin of its Colombian emerald host. In addition to the gas bubbles and briny liquid contents, four separate daughter crystals are present as solid phases. Diffused transmitted light. 120x.
A network of very fine fluid-filled channels represent an intermediate stage of fracture repair in this bright blue tourmaline from the State of Maine, USA. Such intricate inclusions in tourmaline are sometimes referred to as trichites. Shadowing, 35×.

Top left
Three-phase inclusions are the variety of fluid inclusions most familiar to the gemmologist, who uses them in distinguishing natural from synthetic gem materials. The three-phase inclusion in the natural Colombian emerald shown here would be proof positive of the gem's natural origin, as long as no hydrothermal synthetic is manufactured with saline solutions, from which three-phase inclusions of analogous composition could form. Shadowing, 60×.

Bottom left
In contrast to the jagged, fringed borders of the primary three-phase inclusions in Colombian emeralds, the syngenetic secondary (i.e. pseudosecondary) three-phase inclusions display rounded contours. These represent the sealed-off fluid relics of partially accomplished healing. Their composition with one or more halite crystal proves that the healing solution was similar to, and perhaps identical with, the original mother-liquor. Transmitted light, 60×.
A cube of halite (sodium chloride) is the solid phase here. Like a tiny dice, it can tumble around in the void, while the much larger gas bubble is held firmly between the walls. The strong saline solution from which the halite crystallized has a refractive index so similar to that of the host fluorite from Africa that the inclusion is almost invisible. Note how the gas bubble stands out from its background in strong relief, thanks to its much higher refractive index. Transmitted light. 40x.
Two immiscible liquids consisting of a watery solution and liquefied carbon dioxide are present together in this flattened cavity in a deep-red African tourmaline. The gas bubbles within the liquid portion are gaseous carbon dioxide (top middle picture). Should the temperature of the host tourmaline be raised to about 31.5 °C, the meniscus between the liquid and gaseous carbon dioxide would disappear (bottom middle picture), i.e., the carbon dioxide would homogenize. Transmitted illumination. 50X.
Top/bottom
Several-negative crystals—from their phenomenal appearance, typically primary—with multiphase fillings, in a blue sapphire from Sri Lanka. A gas bubble can be recognized suspended in a liquid, and a pale as well as a dark solid phase, neither of which could be identified. On heating the sapphire to 315°C, the liquid and gaseous phases homogenized, whereby the proof is furnished that they consist of carbon dioxide. Darkfield. 12 x

Going... carbon dioxide liquid and gas fluid inclusions are very common in Sri Lankan sapphires. The inclusion shown here also contains several small solid phases. One of them, a long thin rod, can be seen traversing the interior of the cavity. Transmitted light. 45 x.
Going . . . when heat is applied to the sapphire, the small volume of gas appears to get smaller. As the critical temperature (31.1 °C) is reached, the carbon dioxides instantaneously homogenize. 4Sx.

Gone . . . the homogenization of the entire volume of carbon dioxide is now complete. i.e. it has attained a supercritical condition. If the sapphire is now laid on a cool surface, the whole sequence is repeated in the reverse. 4Sx.
A hemimorphic negative crystal in an amethyst of unknown origin distinguishes itself by its four-phase content—two immiscible liquids, a gas bubble and a solid crystal. The two immiscible liquids are difficult to distinguish from each other. Darkfield. 40x.

Under the influence of increased temperature (31.5°C), one of the two immiscible liquids has homogenized with the gas bubble, thus proving unmistakably that it consisted of CO₂. The tiny volume of aqueous solution is still visible in the upper right-hand corner. Darkfield. 40x.

In a yellow beryl from Malagasy, hovering above a secondary healing fissure, are several primary one-, two- and multi-phase inclusions in which the gas bubble proves that they contain a liquid. Transmitted illumination. 25x.

In the same yellow beryl from Malagasy, the meniscus disappeared on heating to 31.5°C, i.e. both liquid and gas bubble consist of carbon dioxide, and now exist in a homogenized supercritical condition. Transmitted illumination. 25x.
In another yellow beryl from Malagasy, the various phases of a primary inclusion—two immiscible liquids, a gas bubble and a daughter crystal—are very easy to recognize. Transmitted light, 50x.

A rise in the temperature (to 31.5 °C) has a homogenizing influence within the spherical volume of fluid, which is still surrounded by an aqueous solution. Neither this nor the daughter crystal are influenced by the homogenization of the carbon dioxide. Transmitted light, 50x.

Arranged in a disciplined manner parallel to the main growth direction, negative crystallites with two-phase fillings occupy a prism face in a blue topaz from St. Anne's Mine in Zimbabwe, thus hallmarking their host crystal. Transmitted light, 35x.

Several needle-like and one slightly stronger growth-tube with a multi-phase content still reliably indicate the main growth-direction (parallel to the c-axis) even in this faceted emerald from the Gachala Mine in Colombia, South America. The growth-tubes often start from an obstacle hindering the growth, being filled with a fluid content. Transmitted light 50x.
Around a primary growth-channel following the growth-direction of its host emerald from the Muzo District of Colombia, a syngenetic secondary wing-shaped healing-fissure has subsequently spiralled up. The screw-like turns of the fissures are very specific of beryl (mainly aquamarine and emerald). Usually they are the result of internal tension through subparallel growth. Darkfield. 25×.

**Top left**
The 250× magnification of this inclusion combination exhibits an exemplary growth event before our eyes: In its conflict with an emerald splinter hardly visible in transmitted light, a classical three-phase inclusion has developed from a trapped fluid phase, and at the same time a multi-phase-filled primary growth-tube rises over the obstacle in this emerald from the Gachalá Mine, Colombia. Transmitted illumination. 250×

**Centre left**
Between crossed polars and inserted 1st order red compensator, the splinter imprisoned by the host emerald appears pale, supplying eloquent enlightenment over the happenings which took place here. 250×

**Bottom left**
An extremely rare fluid inclusion in a Mexican opal from Querétaro. The fluid inclusion contents are trapped in a rhombohedral-shaped void which at one time was occupied by a carbonate such as calcite or dolomite. The carbonate dissolved, leaving a void. Note that tiny dark-red solid particles adhere to both the inner walls of the cavity and the gas bubble's surface. Transmitted and oblique illumination. 50×.
This clearly-defined primary multi-phase inclusion in a yellow fluorite from Illinois, USA, owes its strange shape to widely contrasting growth-conditions. These lead to the periodical repair of the growth-tube walls. The brown grains arranged in parallel lines are bitumen. Transmitted illumination. 100x.

Two globules of natural petroleum were trapped by this hydrothermally generated Illinois (USA) fluorite. The gas bubbles present in the brownish-orange petroleum are probably methane gas, while the black substances partially coating the inner surfaces of the sub-spherical voids containing the petroleum are a tar-like substance which developed from the crude oil at a later time. Darkfield and oblique illumination. 45x.

The hydrothermal rock-crystals – mainly those from Illinois, USA, are very susceptible to the development of syngentic primary and secondary fluid inclusions, of which the liquid phase is composed of oil and the solid component of a tar-like substance (bitumen). This portrait of one of these inclusions displays the rhombohedral termination of a negative (primary) crystal. Darkfield. 75x.
Stages in the healing of a crack in a quartz crystal resulting in the formation of secondary inclusions. Solution of the curved surfaces and redeposition as dendritic crystal growth on others eventually results in the formation of sharply faceted, negative crystal inclusions. If this process occurs with falling temperature, the individual inclusions may have a variety of gas/liquid ratios.

*Drawing by E. Roedder, US Geol. Survey, Reston, VA, USA*

If, for any reason, a fissure should develop within the body of a crystal during its growth, this can, on account of the capillary strength, fill with therapeutic mother-liquor. This spreads out more or less *evenly in the fissure*, wetting its walls, as in this Burma sapphire. Transmitted illumination. 25× (Stage a)

The healing process starts along the inner edge of the fissure, where the larger sections heal first, whereby rough, limp and impoverished fluid relics are shut off. Green beryl from Brazil (Stage b).
Stage b) is also illustrated here, in a partially healed fissure of a mauve scapolite from Mogok in Burma, in the form of sealed-off flaccid fluid drops. Darkfield, 25×.

In the course of progressive therapy — mainly in corundum (Thai and Pailin-rubies and -sapphires) — healed islands and protracted plates of unmistakably typical pattern are formed. Transmitted light, 30×.

This photomicrograph illustrates recrystallized plates with hexagonal front-edges which have been held up against a still unhealed surface. If enough mother-liquor had been available, the whole fissure would probably have been completely healed. With increasing healing the repaired planes extend still further, and the impoverished fluid remnant droplets shrink continually. Transmitted illumination, 150×.

This illustration reveals very clearly the difference between the front line and the finished, repaired parts of a healing fissure in a sapphire from Sri Lanka. If sufficient saturated healing-solution is available, the healed fissure received the well-known pattern resulting from sealed off and lixiviated fluid drops and runnels between the healed sections. The comparison between the front-line and the other parts of the “feather” manifests the achievement of such patterns very eloquently. This condition represents stage c) transmitted light, 50×.
As a result of various therapeutic forces, the promoting or impeding of the healing-process within the formative space of the injured gemstone, and its own speed of growth and reaction to temperature, those fantastic, decorative, often fascinating and sometimes typical (of occurrence or species) fluid inclusions, known as "feathers", "veils" or "wisps", are created during stages b) and c), e.g. [left column from top to bottom] in a sapphire from Sri Lanka, in a sapphire from Thailand, in a ruby from Sri Lanka, (right column from top to bottom), in a Burma spinel, in a colourless beryl from Brazil and in a Russian alexandrite.

This Sri Lankan sapphire contains two small primary negative crystals surrounded by secondary fluid "feathers". Darkfield and oblique illumination 35x.

The last phase of healing denotes itself in that the healed regions in the fissure expand maximally, while the depleted fluid remnants keep contracting until they are encapsuled within small negative crystals. This final state is illustrated very beautifully here in a Burma sapphire by zones of more or less densely assembled, tiny secondary-negative crystallites. Transmitted light 50x.
Fluid-Inclusions

Fluid inclusions are small volumes of fluid, trapped within imperfections in crystals. Most are microscopic in size. They are found in essentially all minerals, including gemstones, as most such crystals, including synthetic stones, have grown from fluids of one sort or another. The term fluid refers to a fluid condition at the time of trapping, and does not require that the inclusion contents be all or even partly fluid when observed at room temperature. Thus an inclusion that trapped a hot dilute water solution, will contain liquid water (and a vapour bubble, from shrinkage) when cooled to room temperature, but one that trapped a very highly concentrated water solution of soluble salts at high temperatures, may cool to form a mass of crystals called daughter crystals, which are only wet with a minor amount of liquid. Still other inclusions, such as those in minerals from intrusive igneous rocks and extrusive lavas, trap a fluid silicate melt at high temperatures. On cooling, such fluid inclusions may crystallize, or may merely supercool to form a rigid glass. Still other inclusions trap a high- or low-density gaseous fluid, and exhibit no changes on cooling.

Fluid inclusions trapped by various growth imperfections during the growth of the host crystal, are called primary inclusions; studies of such inclusions have provided us with a wealth of information about the temperature, pressure, density and composition of the fluids deep in the earth from which various gemstones have formed, millions or even billions of years ago.

If a crystal is fractured while it is immersed in a fluid, the fracture may heal, trapping small amounts of that fluid as a plane of secondary inclusions outlining the former fracture. This cracking and healing may occur during the growth of the crystal or millions of years later, and frequently it is only one of a series of such events which the crystal has undergone. The fluid in such secondary inclusions may be very different from that in the primary inclusions, and gives us an insight into some events in the history of the sample, after its original crystallization.

Although fluid inclusions in a gem are imperfections in a material which is prized mostly for its perfection of crystallization, they are not completely negative features. Study of fluid inclusions may prove useful in the search for gem deposits, in the identification of gemstones and the recognition of the source for specific stones, in the recognition of a given stone, even after recutting, in guidance of attempts at synthesis, and (with some caveats) in the recognition of natural versus synthetic stones.

E. Roedder