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STRUCTURE OF HYALITE FROM THE SPRUCE PINE PEGMATITE DISTRICT, NORTH CAROLINA

WILLIAM J. FURBISH
Department of Geology, Duke University, Durham, North Carolina 27708, U.S.A.

ED L. SCHRADER, JR.
Department of Geology and Geography, University of Alabama, University, Alabama 35486, U.S.A.

ABSTRACT

Hyalite opal from the Spruce Pine, North Carolina, pegmatite district gives an X-ray pattern similar to the pattern of vitreous silica, on which peaks of quartz, tridymite or cristobalite may be superimposed. Scanning electron micrographs show particulate, relatively even-sized spherical structures that occur in semi-close-packed arrays as well as randomly dispersed spheres of various sizes. The matrix consists of layers of apparently structureless and loosely consolidated particles; particulate spherical structures are either enclosed within single layers or lying across the layer interfaces.

SOMMAIRE

Une opale hyaline pegmatitique de Spruce Pine (Caroline du nord) donne aux rayons X le diagramme de poudre de la silice vitreuse, auquel se superposent, dans certains cas, les pics du quartz, de la tridymite ou de la cristobalite. Au microscope électronique à balayage, on voit des particules sphériques de même diamètre en assemblages quasi-compacts, ainsi que des sphères de différentes tailles dispersées au hasard, dans une pâte de particules mal-consolidées, à l'aspect amorphe, disposées en couches; des amas réguliers de particules sphériques se trouvent soit au sein d'une couche soit sur l'interface entre deux couches.

(Traduit par la Rédaction)

INTRODUCTION

After Greig (1932) indicated the presence of crystallinity in certain opals, Taliaferro (1935) pointed out that apparently amorphous hyalite gave definite β–cristobalite X-ray patterns. Jones *et al.* (1963) concluded that most hyalite is near-amorphous. In 1964 Jones *et al.* reiterated that hyalite gives an X-ray diffraction pattern with only a diffuse band at 4.1Å. Jones & Segnit (1971) used X-ray diffraction patterns as their principal criterion for the classification of opals into three structural groups. Hyalite was placed in category "Opal–A," characterized by a diffuse X-ray band at 4.1Å.

Controversy has continued over the specific nature of the crystalline components of opal since Greig's (1932) work. Quartz, α–cristobalite, disordered α–cristobalite, cristobalite, α–tridymite, disordered α–tridymite, β–tridymite, disordered β–tridymite, tridymite and highly disordered to near-amorphous phases have all been reported by various authors. Flörke *et al.* (1973) felt that, even though all natural hyalite they examined seemed near-amorphous, there remained a possibility that hyalite with disordered cristobalite and tridymite components might form under suitable natural conditions. This conclusion followed from their experimental work, in which they obtained such material in their high-temperature or high-depositional-rate hydrothermal runs (or in both).

The discovery by Pense (1963) of the existence of discrete particulate structures in opal led to re-evaluation of the physical and optical properties of opal. Jones *et al.* (1964) first related the X-ray pattern to internal particulate structures. Sanders (1964) considered the "color" of precious opal to be caused by diffraction by the lattice of voids between regularly packed and sized particulate spheres within the opal host.

Little thought was given specifically to the origin of hyalite until the paper of Flörke *et al.* appeared in 1973. They conducted a series of experiments in which silica glass with the physical properties and appearance of natural hyalite was formed by vapor transport in a hydrothermal system. They suggested that this might also be the mode of formation for hyalite in at least two natural occurrences and, if so, the origin of hyalite is then genetically distinct from most opal deposits that originate through deposition from solution or colloids.

SAMPLE DESCRIPTION

Hyalite quite commonly occurs in the Spruce Pine, North Carolina, pegmatite district in fractures in alaskite, in pegmatites enclosed by the alaskite, or in pegmatites peripheral to the alaskite. The Spruce Pine pegmatite district is located in the extreme west-central portion of North Carolina and is geographically limited to Avery, Mitchell and Yancey counties.

Generally, hyalite from this area occurs in thin, sheet-like masses on fracture surfaces. The hyalite-coated surface consists of botryoidal to stalactitic projections of transparent, clear to light yellow, green or blue glass-like masses that may cover all or only part of the fracture surface.

Samples for this study were collected from various geological environments within the area. They were from: (1) a large pegmatite enclosed conformably by a hornblende gneiss and located about 450 m from any known alaskite body, (2) a pegmatite enclosed within the host alaskite, (3) various fractures within the alaskite body either in close proximity to the enclosing biotite and hornblende gneisses and schists or well within the body.

Refractive indices from the hyalites of these sample locations vary from sample to sample: 1.443 to 1.445 (one value 1.458); in some instances, they even vary slightly within individual samples. A random difference in intensity and reaction to various wavelengths of ultraviolet radiation was noted among the specimens. This was taken to indicate the presence of different chromophors among samples.

EXPERIMENTAL DATA

Differential thermal analysis

Differential thermal traces of four hyalite

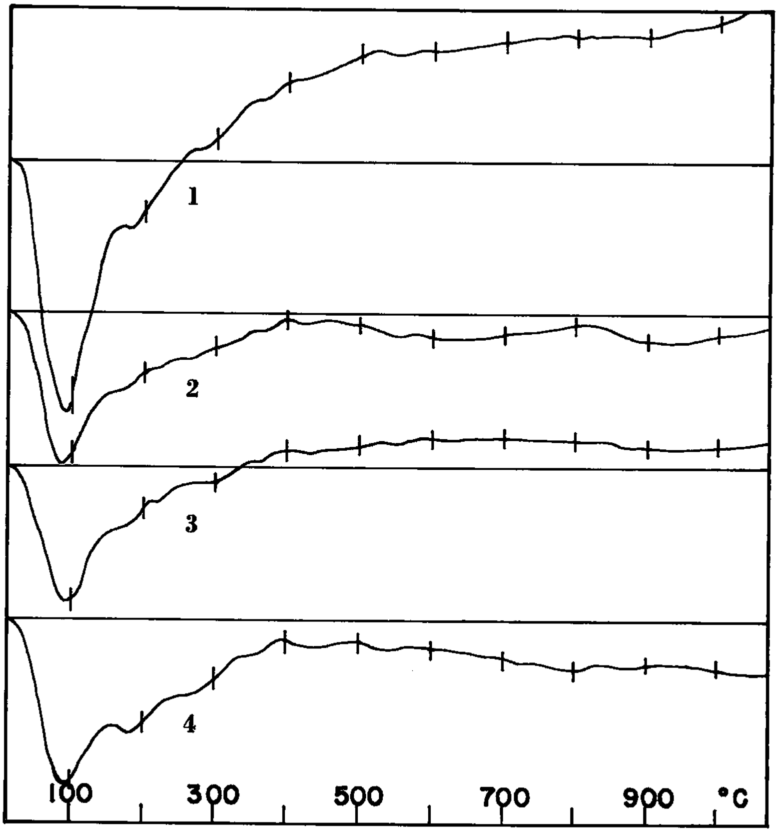


FIG. 1. Differential thermograms of four hyalite samples showing reactive variations. Sample 1 from large pegmatite, 450 m from known alaskite body; samples 2 and 3 from separate fractures (15 m apart) in alaskite body; sample 4 from fracture in alaskite body 75 m from a large included pegmatite. Heating rate 10°C/min, maximum T 1140°C.

samples chosen from different local depositional environments are shown in Figure 1. All samples were dry-mortar ground, sized to 200 mesh, desiccated over silica gel for ten days and analyzed in a uniform fashion. Variations in intensity and position of reaction peaks are shown to exist, and the contrast between samples can readily be seen. Numerous other sample analyses, not shown in this paper, gave a similar variation in differential-thermal-analysis patterns to the four shown in Figure 1, even though many were obtained within one metre of each other in the same depositional environment.

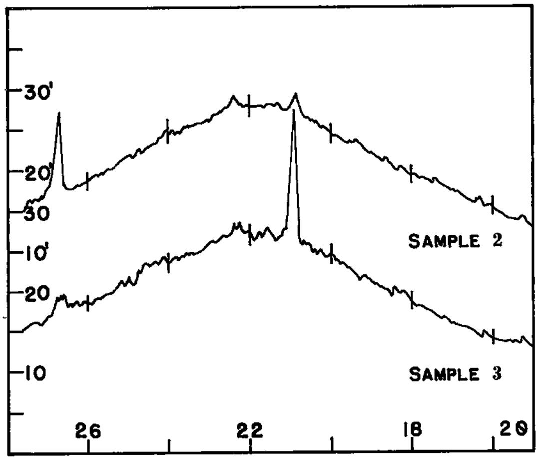


Fig. 2. X-ray diffraction patterns of samples 2 and 3 (of Fig. 1) from separate fractures (15 m apart) in alaskite body. Cu *Kα* radiation.

X-ray diffraction

X-ray diffractograms of both heat-treated and normal unheated samples were made. A wide variation in patterns exists from sample to sample in both the heat-treated and unheated material. Unheated hyalite samples give X-ray diffractograms with peaks that range from a broad swell at 4.1Å to a well-defined α–cristobalite pattern. Superimposed upon these patterns, in some instances, are subdued peaks of either quartz or tridymite (Fig. 2). The specimens were heat-treated in open crucibles. The successive temperatures to which the samples were raised, before being X-rayed, were correlated with the postreactive areas of D.T.A. patterns for a portion of the same sample that was used for D.T.A. Preliminary control-heating runs were done on both unground and ground material from single samples. X-ray diffraction patterns from the controlled runs showed that far less intense peaks were obtained from the preground material. Whole material was used for all sub-

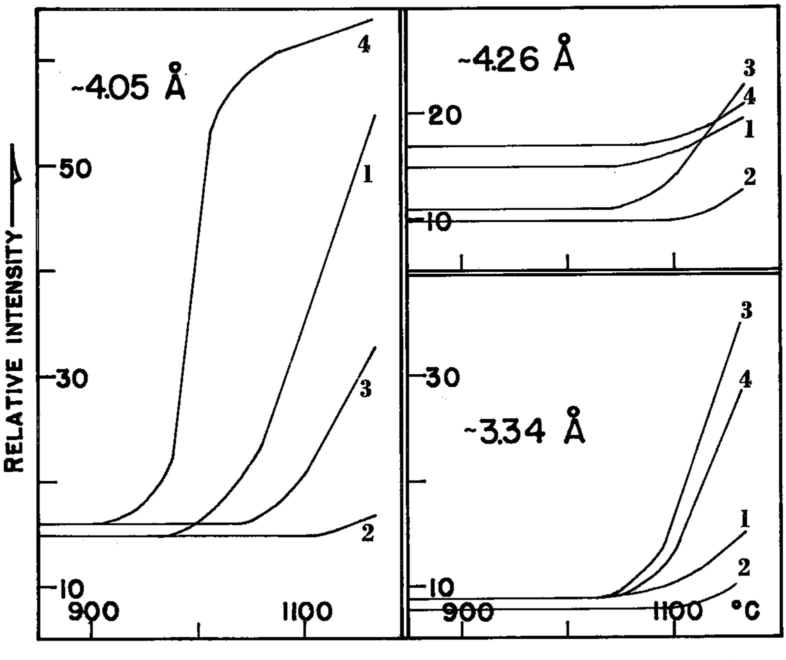


Fig. 3. Heat-treatment curves of samples 1 through 4 based on relative X-ray peak heights with increased T. Curves were drawn on change of 20°C T increments between 800 and 1140°C.

sequent heat runs and ground for X-ray analysis after heat treatment was complete. Beyond 800°C, to a maximum T of 1140°C, temperature increments of 20°C were used, and each increment peak temperature was held for twenty-four hours. Conversion to α–cristobalite was

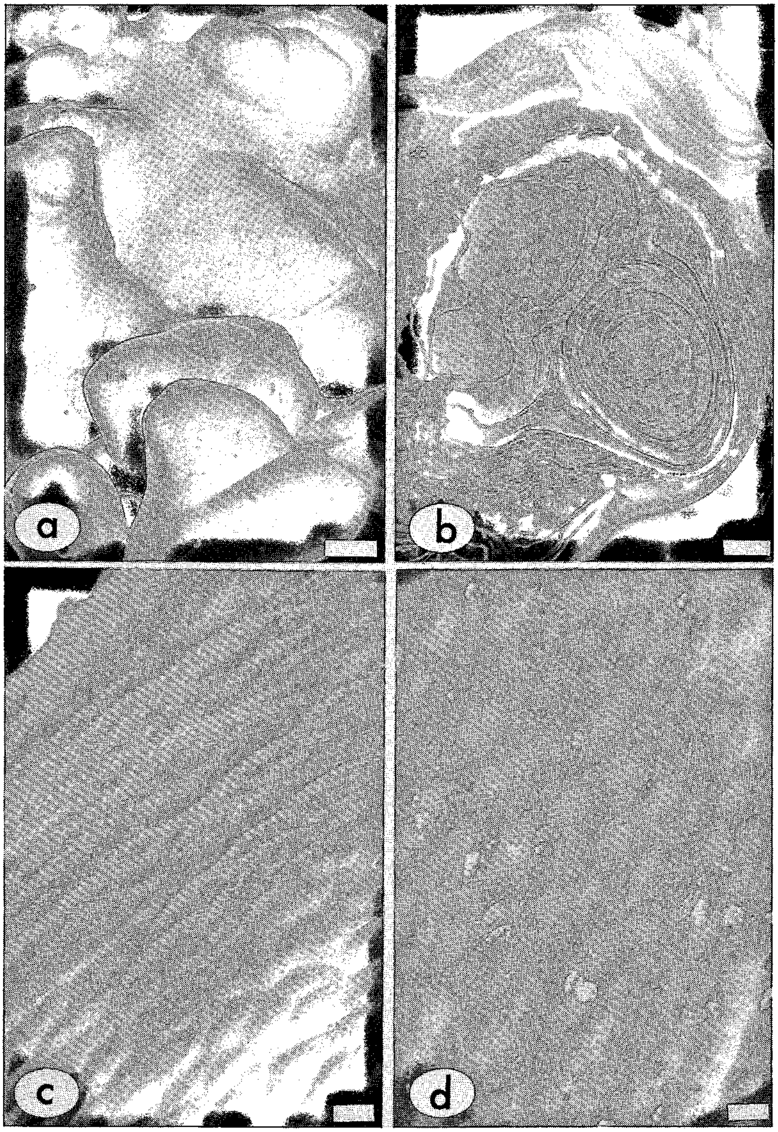
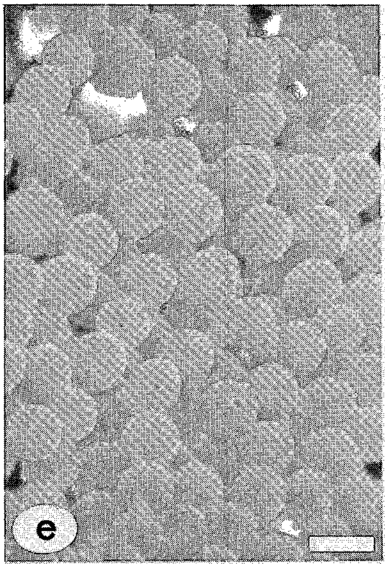
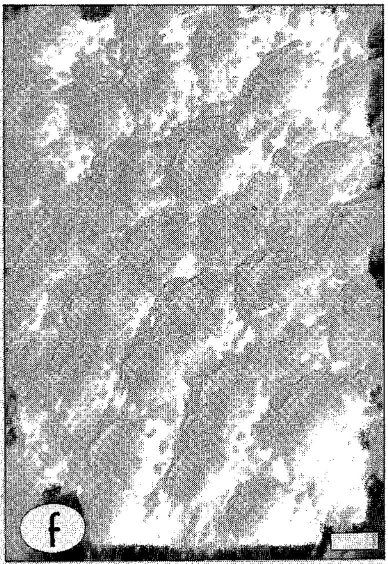


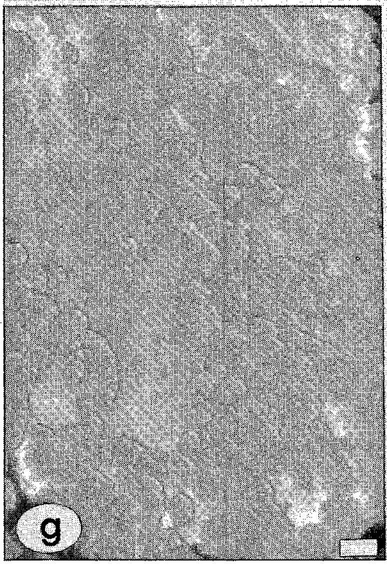
FIG. 4a. Scanning-electron micrograph of hyalite growth-surface showing stalactitic growth forms, smooth and rough surfaces. Bar scale = 100 μm.

FIG. 4b. Scanning electron micrograph of the etched fracture surface on a single stalactitic growth unit. Concentric growth layers can be seen. Bar scale = 60 μm.

FIG. 4c, d. Scanning electron micrographs of normal hyalite sample surfaces showing a sequence of surface morphologic expression from lens-shaped raises of uncertain origin through semi-spherical forms that are definitely a surface expression of an internal structure. Bar scale = 30 μm.







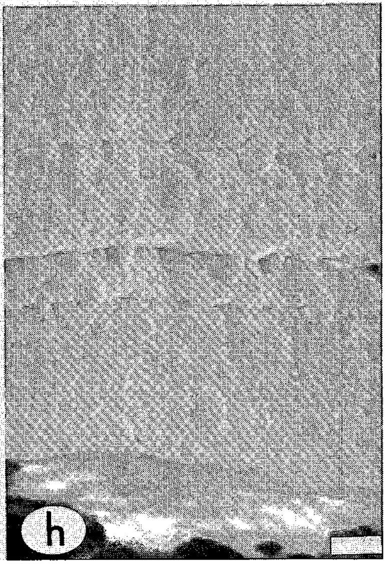


FIG. 4e. Scanning electron micrograph of a normal hyalite growth surface showing particulate-sphere growth configuration. The lack of structureless silica material and the interconnecting necks are especially noticeable. Bar scale = 2 μm.

FIG. 4f. Scanning electron micrograph of a single growth layer in which the volume of spheres and structureless material are similar. Bar scale = 2 μm.

FIG. 4g. Scanning electron micrograph of a single growth layer in which the deposition of structureless silica material predominates over deposition of particulate spheres. Bar scale = 1 μm.

FIG. 4h. Scanning electron micrograph of a cross-section of growth layers. This surface has been etched and shows particulate spheres dispersed at random in structureless silica. Some spheres cross over the boundary between layers. Bar scale = 1 μm.

initiated over a temperature range from ~880 to ~1100°C with those samples that started to convert at the lowest temperature reaching the maximum total conversion (Fig. 3). Usually those samples with an initial X-ray pattern similar to that of vitreous silica converted to α–cristobalite most readily (1 and 4, Fig. 1), whereas those initially with a more definite crystalline pattern showed slight to only moderate amount of reaction (2 and 3, Figs. 1 and 2). No tridymite patterns were produced.

Scanning electron microscopy

Scanning electron micrographs were obtained from hyalite samples that were on either normal growth surfaces, etched growth surfaces, fracture surfaces or etched fracture surfaces. A record of both external morphological and internal structures was thus made available.

The external surface of the hyalite is not planar but rather uneven and consists of blunt stalactite-like projections up to 0.5 mm across (Fig. 4a); these coalesce at their base to form a planar growth zone on the initial hyalite–host rock interface. These projections consist of successive layers of opaline material (Fig. 4b: broken, etched surface). The unbroken, unetched surface of the stalactitic projections may have a perfectly flat surface or may show various sizes and shapes; some cases definitely show internal structure but in others this is less certain. This habit seems to have totally random distribution. Figures 4c and 4d reflect what appears to be a transition from lens-shaped forms to spherical external expressions of internal particulate structural forms. The lens shapes are somewhat exaggerated in Figure 4c because of the inclined electron beam; in Figure 4d the beam is not inclined, and non-spherical forms exist.

Figure 4e represents an unetched natural surface growth area involving mostly particulate spheres in a semi-close-packed array. In this figure the continuous necked interconnection between particulate spheres can be seen. A small amount of structureless, loosely consolidated material can also be seen filling the voids between the spheres. Unlike Figure 4e, Figure 4f represents a growth surface in which the particulate spheres are fewer and more dispersed in the structureless, sponge-like silica material. The layering of spheres is present but no interconnecting necks between spheres are visible.

Figure 4g shows still fewer particulate spherical units than Figure 4f, and where the growth layer has developed it consists predominantly of apparently non-structured siliceous material.

Figure 4h, representing a pattern prevalent throughout the study, shows an etched cross-section surface of hyalite in which the spongy, structureless siliceous material forms layers and is the predominant siliceous form. Particulate spheres are dispersed at random throughout the mass and some are seen to lie across layer interfaces.

DISCUSSSION AND CONCLUSIONS

This study shows that hyalite opal may exhibit crystallinity (Fig. 2) and may contain particulate spherical structures (Fig. 4f). Neither of these features, however, necessarily occurs in methodical patterns in the material studied. Both vary from sample to sample or within a single sample.

Two varieties of solid material make up the hyalite body. Particulate units of spherical or ovoid shape may be present (Figs. 4c and 4d). These units either lie in layered semi-close- to open-packed arrays (Fig. 4e) or occur at random throughout the opal body (Fig. 4h). Their size may be relatively constant within a single layer or sample (Fig. 4e), or variation may occur (Fig. 4h). A porous unit of loosely aggregated particles constitutes the remainder of the opal body (Fig. 4h). It is layered, but the thickness of the layers varies.

The physical relationship of the two units is well documented by the electron micrographs taken during this study. Single or multiple layers of particulate spheres may accrue on the growth surface with no infill of aggregate particles (Fig. 4e). Such a situation is probably only a temporary surface-growth feature. Particulate units together with interspherical infill of aggregate particles may constitute a single layer (Fig. 4f), or the spherical units may be dispersed at random throughout the aggregate unit particles. Where this occurs the spherical units may be contained within a single layer of aggregate material, or they may cut across layer boundaries and be contained within two or more layers (Fig. 4h).

Such relationships indicate that two independent but interrelated modes of depositional processes are contributing to the hyalite growth patterns; predominance of (1) production of one unit or (2) its deposition over the other during hyalite growth results in the final spatial relationships and ratios of the two units involved.

In 1964 Sanders suggested that visible color in precious opal is the result of light diffraction from a three-dimensional array of close-packed silica particles within the range of 1500 to 4000Å in diameter. Sanders & Darragh (1971) later refined refined this concept, pointing out that the minimum diameter of spheres able to diffract visible light in the violet part of the spectrum is 0.16 μm and that the maximum diameter of spheres able to diffract in the red part of the spectrum is 0.28 μm. Requirements for the diffraction of light within the visible range are generally not met by the hyalite opal material of this study. Although close packing in a three-dimensional pattern does occur, it is not extensive, and the size of the particulate silica spheres usually varies so that a visible diffraction effect cannot be obtained.

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