Aluminium Diboride-type Structure in Ethiopian Opal-CT Revealed by Fast Fourier Transform

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Invisible on a scanning electron microscope image of the surface of an Ethiopian opal rough fracture, a periodic arrangement was detected by fast Fourier transform. Using a mask to eliminate the continuous background and keeping only the bright spots in the reciprocal space (fast Fourier transform pattern), an image reconstructed by inverse fast Fourier transform (IFFT) emphasizes a very regular bidisperse array. Taken on a vicinal plane, the image of the successive steps of the stacking allows identification of the crystallographic structure and estimation of the parameters of this aluminium diboride-type photonic crystal. In addition, another more complex IFFT image allowed confirmation of the structure and determination of the crystallographic indexing of the steps, despite image deformation due to the tilt of the vicinal plane under the electron beam.

1. Introduction

The structure of precious opal is known to be a three-dimensional periodic array of hydrated silica spheres all having the same diameter. The diffraction of light through this photonic crystal causes its famous play of colour. Besides this most frequent monodisperse arrangement, only a few cases of bidisperse opals have been documented. Such structures can be described in terms of crystallographic unit cells, as for binary crystals $A_mB_n$. Here, both kinds of spheres $A$ (with a radius $R$) and $B$ (radius $r$) arrange in compact stacking, and the resulting structure depends on the ratio $m/n$ of the numbers of spheres $A$ and $B$ and on the ratio of their radii $R/r$.

After the discovery of the structure of opal by John Sanders in 1964, Sanders & Murray (1978) found bidisperse opal arrays on electron micrographs of surface replicas. They identified an $AB_{13}$ structure, similar to the binary compound NaZn$_{13}$, and an $AB_2$ one (Sanders, 1980), like aluminium diboride. Later, Gauthier et al. (1995) detected another type of $AB_2$ structure in a Brazilian $A$ opal by transmission electron microscopy followed, for identification, by scanning electron microscopy (SEM). In this Brazilian sample, the spheres were visible individually, with voids in between, like for opals formed in a sedimentary environment. Another bidisperse stacking was found in a fractured cristobalite–tridymite (CT) opal from Jalisco, Mexico (Gauthier et al., 2004). Formed in volcanic areas, CT opals do not fracture to display individual spheres, but exhibit instead lepispheres cemented by additional silica nanograins filling the voids. In this case, gentle reaction with dilute acid revealed the core of the spheres, preferentially eroded (Smallwood et al., 2008). The fracture appeared along a vicinal plane – a crystallographic plane with relatively high
Miller indices $hkl$, slightly inclined to a plane of low indices – so that the lattice of spheres is cut at a small angle with respect to the main planes of the stacking. Thus, the three-dimensional structure was identified by sequential observation of the neighbouring steps emphasized by the fracture along the vicinal plane. Finally, the unit cells of the amorphous opal from Brazil and the opal-CT from Jalisco were similar to that of binary crystals in the Laves phase series, like MgCu$_2$ or MgZn$_2$.

Recently, we observed by SEM various broken pieces of an Ethiopian opal from the province of Wollo. Despite an intense play of color, the fractures did not reveal any periodic structure because, as noticed earlier for opals formed in volcanic environments, a siliceous cement filled the voids of the opals. We decided then to submit the SEM images to a fast Fourier transform (FFT) analysis. This image treatment was earlier tested on an Australian opal from a sedimentary deposit, to justify the interpretation of the reconstructed images of the monodisperse Ethiopian opals (Stephant et al., 2014).

During this exploratory experiment, we obtained images given by an inverse FFT calculation (IFFT), which seemed to indicate a bidisperse arrangement. The aim of this paper is to analyse the IFFT image to obtain a complete crystallographic description of the lepisphere stacking.

2. Materials and method

2.1. Materials

Samples were collected in Ethiopia (Province of Wollo) by BR and Francesco Mazzero and observed with SEM. After an FFT treatment of the images, we detected in many samples a periodic structure in a binary system of silica lepispheres. The samples were freshly broken fragments, lightly coated with platinum and then positioned under the beam so as to be as horizontal as possible.

The method has been described previously (Stephant et al., 2014). The aim of the procedure is to eliminate the random signals in reciprocal space and rebuild a filtered image with the periodic spots.

Initially, the scanning electron microscope is adjusted to obtain a suitable image according to the preliminary adjustments of Stephant’s method: we chose the highest image size offered by our microscope (5120 × 4096 pixels), fixed a very long exposure time to acquire the image (28 min) and operated at low magnification (800×). This careful setup must be done to emphasize the resolution during the next step. Then each image was treated using the DigitalMicrograph software (Gatan Inc., Pleasanton, CA, USA). We calculated first the autocorrelation of the SEM image, then the FFT of this autocorrelation. The output of this process clearly shows bright spots related to periodic matter of the sample. Only those spots were selected with a masking tool of the software and used to calculate an IFFT image. This last image gives evidence of periodic matter on the SEM image and acts as a filter for the random part. Images were acquired with a thermal field emission gun scanning microscope (JSM 7600F, JEOL Ltd, Akishima, Japan) using an in-lens detector of secondary electrons.

2.2. Preliminary considerations

Such experimental conditions are not typical for the determination of a complicated unknown structure. Therefore, we will first discuss the difficulties and the advantages one can encounter during the analysis of the transformations realized from initial SEM images.

2.2.1. Difficulties. (1) A priori, a three-dimensional structure cannot be solved, or is very unlikely to be solved, from an image taken on a two-dimensional surface.

(2) In opals that come from a volcanic environment, silica lepispheres are mostly not visible by SEM. Sometimes, a gentle chemical attack reveals the core of the lepispheres (destroyed by dilute acid), but not their effective diameter. Their outer contour, more resistant to the acid, is masked by the additional filling of voids by silica nanograins, similar to those constituting the crown of lepispheres (Fritsch et al., 2006). Thus, the real diameter is unknown, as is the ratio of $A$ and $B$ sphere diameters.

2.2.2. The positive points. (1) To ensure the stability of the structure, we must assume that the stacking is as compact as possible. The different spheres must be in contact.

(2) Mono- or bidisperse opals form by sedimentation of spheres. One can thus expect growth to occur plane by plane. Help could be offered by local configurations already observed in binary systems of spheres in colloids (Bartlett et al., 1992; Velikov et al., 2002; Schofield et al., 2005).

(3) Observed symmetries and characteristic lengths (distances between repeating units) will be useful elements for this structural determination.

(4) The presence of steps, due to fracturation along a vicinal plane, affords three-dimensional information which would not be given by a cleavage plane (crystallographic plane). This kind of structure identification from stairs of a fracture has previously been achieved on a bidisperse system for opal (Gauthier et al., 2004).

(5) The FFT pattern can also give usable information.

(6) The reconstructed image (IFFT) is not an exact visualization of the surface observed by SEM. In these CT opals, the nanograin structure means that the electron beam cannot detect all of the lepispheres, only a fraction in dispersed islets. Nevertheless, the IFFT gave a continuous representation of the regular lattice.

3. Results

3.1. First sample

3.1.1. SEM image, FFT diagram and IFFT image. An image from the surface of the fracture was recorded from a piece of the sample numbered 167. It presents an apparently smooth surface, except for cracks visible on the right (Fig. 1a). Using the application DigitalMicrograph for achieving the fast Fourier transformation, we get the pattern of Fig. 1(b). Keeping only the bright spots, the mask of Fig. 1(c) is used for
inverse Fourier transformation. The IFFT image (Fig. 1d) displays a regular lattice, with bands of large or small rounded features, representing the images of two kinds of lepispheres (for simplification, we will henceforth use the term ‘spheres’, instead of ‘lepispheres’, except if necessary).

As expected, the main rows of the FFT pattern are perpendicular to the bands of the IFFT image. These regular bands mean that the fracture takes place along a vicinal plane, near a dense one, slightly turned around a main row, oriented south-west–north-east. Crossing the structure, the fracture surface is in fact a stair composed of steps parallel to a basal hexagonal lattice. That would allow the building of a three-dimensional structure by considering the features of each step, one after another.

3.1.2. Determining the structure from the steps. An enlargement of the IFFT image is shown in Fig. 2(a). The large spots represent large spheres in a hexagonal arrangement, lying at the edges and at the centre of a large hexagon (band M, right). The matter around each, appearing as a hexagonal and continuous lattice, represents the siliceous cement together with sections of small lepispheres. The resolution does not allow for more precision than shown. Between the bands of large spheres, a hexagonal array represents a plane of small spheres, in hexagonal arrangement (band N, right). But, the centre of each hexagon is empty, revealing part of the large sphere nestled under it. Only two types of layers are thus recorded: M, O, Q, S for one kind (large spheres in hexagonal arrangement), and N, P, R for the second (honeycomb arrangement of small spheres).

The spacing between steps M, O, Q and S is very regular, as shown by the double-headed arrows of the same length (Fig. 2a, right). These steps being quite large, the fracture surface corresponds to a vicinal plane only slightly inclined with respect to the basal plane, with the horizontal row as rotation axis. The repeat distance between equivalent rows of large spheres lying on two neighbouring steps (i.e. the length measured on the vicinal plane) corresponds to about 7.28 spacings between two neighbouring rows of large spheres on the same step. Its projection on the basal plane differs only slightly from this value, and thus we infer that the separation between bands corresponds to the width of exactly seven rows of large spheres in the basal plane.

Note that there is a visible distortion of the hexagons, probably due to a tilt with respect to the horizontal during SEM image acquisition. Although the entire surface of the
sample was positioned under the beam so as to be as horizontal as possible, the observed surface is not horizontal at the scale of the observation because, by nature, the material conchoidally fractures.

3.1.3. Crystallographic cell. The preceding observations lead us to a simple description of the structure. Only two kinds of planes are alternately stacked: one of small spheres (B) in a hexagonal array with regular vacancies like a honeycomb, and another hexagonal array of large spheres (A) settled on the vacancies, above and below each hexagonal seat. The sheet formed by one plane of small spheres surrounded by one plane of large spheres is then replicated along the vertical axis. A unit cell can be defined and is illustrated in Fig. 3. The respective positions are 000 for the large sphere, and 1/3, 2/3, 1/2 and 2/3, 1/3, 1/2 for the small ones. The unit cell corresponds to the formula $AB_2$, and the structure is related to that of aluminium diboride, belonging to the space group $P6/mmm$ of the hexagonal system.

3.1.4. Estimation of the diameter of small spheres. The existence of the steps M, O and Q in Fig. 2 supports a model wherein the small spheres of the honeycomb touch one another. That was not the case for the aluminium diboride structure found by Sanders (1980) with an opal of amorphous type, where the large spheres were in a compact arrangement. The present planar arrangement has been already found in a synthetic binary colloidal crystal (Velikov et al., 2002). Thus, the main horizontal row of large spheres allows us to estimate the diameter of the small spheres. The distance between the centre of two neighbouring large spheres corresponds to $2(3^{1/2})r$. Measuring ten distances, and comparing with the scale bar, we get an approximate value of 0.22 µm for the diameter 2r. However, we do not know if the beam was exactly perpendicular to the fracture, so a precise value cannot be given by this experiment.

3.1.5. About the $R/r$ ratio. In this structural model, to compare an experimental value of $R/r$ with the theoretical one, we can express the height $H$ between two identical planes (of small or large spheres) by (see Fig. 4)

$$H^2 = 4(r + R)^2 - 16r^2$$

or, with $r = R/r$, $H^2/r^2 = 4r^2 + 8r - 12$. \hspace{1cm} (1)

$H$ can be calculated for two extreme theoretical situations and an intermediate one:

(i) The planes of small spheres are in contact (Fig. 4a). Thus $H = 2r$, and equation (1) becomes $r^2 + 2r - 4 = 0$, having a positive root $r = 1.236$ as the lowest possible value (see Discussion for this theoretical case, where large spheres are interpenetrating).

(ii) The large spheres are in contact through the median plane of small spheres (Fig. 4b). The distance between two planes of small (or large) spheres is $H = 2R$, and equation (1) is reduced at the first degree with the solution $r = 1.5$.

(iii) The large spheres are in contact in the horizontal plane (Fig. 4c). Assuming this condition [realized in Fig. 5(b)], $l_1 = 2R = 2r(3^{1/2})$, or $r = R/r = 3^{1/2}$ as the highest possible value, and equation (1) gives $H^2 = 8(3^{1/2})r$ and finally $H = 3.722r$.

Later, we will discuss these different cases. But now, we will try to estimate from Fig. 2 the value of $R/r$. We cannot measure directly the diameter of the large spheres. Remember that the limits of the lepispheres are difficult to determine, because the voids between them are filled with silica. Moreover, the observed size may correspond approximately to the core of the lepispheres, not necessarily to their external diameter.

We tried to calculate $r$ by comparing the distance $L$ between the rows of large spheres in the vicinal plane and the distance $l$ between the corresponding rows in the basal plane. The reference for $r$ is given by the distance between two large spheres on horizontal rows, i.e. $l_1 = 2r(3)^{1/2}$. Vertically, depending on the $R$ value, the distance $H$ between two large spheres...
spheres is given by equation (1). Thus, from Fig. 5(a), we can write, with \( r^2 = 441 \),

\[
L^2 = H^2 + I^2 = (4r^2 + 8r + 438)r^2. \tag{2}
\]

Finally, with \( r^2 = l_l^2/12 \), the \( \tau \) value can be deduced from the equation

\[
4r^2 + 8r + 429 - 12(L^2/l_l^2) = 0. \tag{3}
\]

With a measured value of \( L/l_l = 6.2 \), the solution is \( \tau = 2.00 \).

This value, being beyond the limits indicated above, cannot be accepted. Analysing equation (2), we note an important contribution of the term \( r^2 \), which itself depends on the measurement of \( l_l \). A slight inclination of the preparation under the electron beam can strongly affect the final value of \( \tau \), even through a slight error on \( l_l \).

Therefore, because the vicinal plane is only slightly inclined with respect to the basal plane, we consider that it is not reasonable to deduce the \( \tau \) value from Fig. 2.

Moreover, there is great uncertainty because we do not know the tilt of the surface sample with respect to the electron beam. We tried as much as possible to place the fracture surface perpendicular to the electron beam. But, when breaking an opal piece, one obtains a rough surface with steps (those of the vicinal plane) and we do not know the tilt of these steps under the electron beam. This is the main fact that forbids any determination, even approximate, for the \( R/r \) ratio between 1.236 and 3\(^{1/2} \).

3.1.6. Indexing of the vicinal plane and tilt angle \( \phi \) with respect to the basal plane. The main horizontal row (Fig. 2) in the basal plane is the rotation axis which can be easily indexed as [100] (see Fig. 3). The vicinal plane of Fig. 2 cuts the axes \( a \), \( b \) and \( c \) at 7, 7 and 1, so its indexing is 117.

The tilt angle \( \phi \) around this row is given theoretically by \( \tan \phi = H/L \). Unfortunately, since we were unable to determine the \( R/r \) value, we cannot obtain the experimental value of \( \phi \).

We can only give the extreme and intermediate values of \( \phi \) for the same three scenarios described previously: \( \tau = 1.236 \), then \( \tan \phi = 2/21 \) and \( \phi = 5.44^{\circ} \); \( \tau = 3^{1/2} \), \( \tan \phi = 2(3^{1/2})/21 \) and \( \phi = 9.57^{\circ} \); \( \tau = 1.5 \), \( \tan \phi = 2.25/21 \) and \( \phi = 6.12^{\circ} \).

3.1.7. The different cells visible on the IFFT image. The IFFT image displays three different unit cells (Fig. 2b). Two have identical parameters corresponding to the horizontal cells on each type of basal plane of the staircase-like fracture. They can be seen in bands M, O, Q and S for the large spheres (cell C1) and in N, P and R for the small spheres (cell C2). In addition, we can observe a supercell corresponding to the periodicity of the vicinal plane. Its large parameter extends from the central row of one band to the second analogous row (for instance from M to Q, and not from M to O). This is because the central row of the intermediate band O is shifted with respect to the central rows of M and Q. The horizontal shift corresponds to half of the distance between two large spheres, i.e. half of the small parameter \( l_l \).

3.1.8. The FFT pattern. As for a diffraction pattern, the FFT pattern (Fig. 6) shows the presence of families of ‘crystallographic planes’ as families of parallel ‘lines’ visible on the IFFT image. To study it independently from the reciprocal lattice of the actual three-dimensional opal structure, we will denote the main rows of this two-dimensional FFT pattern as the \( h0 \) (vertical) and \( 0k \) (horizontal) axes. Then, it is interesting to point out some features displayed by this pattern.

**Along the \( 0k \) row.** This row displays numerous spots close together, of high, middle or low intensity, or extinguished. Taking pairs of opposite points with respect to the centre, and using the scale bar, it is possible to obtain the distance values in the reciprocal and direct lattices (Table 1), i.e. \( D \) and \( d \), respectively.

Attention must be paid particularly to spots B (the most intense) and G (almost absent). The distance of 0.169 \( \mu \) calculated for the first corresponds quite closely to 1/28th of the distance of 8.746 \( \mu \) measured on the IFFT image between the centres of the M and Q bands, i.e. 14 intervals between two adjacent rows of large spheres. That confirms our observation (made in §3.1.2) concerning the number of intervals (seven) between Q and S.

For the superstructure in the vicinal plane, the shift of one row in two leads to the intensity of \( 0k \) spots with uneven \( k \) values being extinguished or reduced substantially.

**Along the \( h0 \) row.** The first spot L along the \( h0 \) reciprocal row is almost extinguished. This is due to the face-centred unit cell C1. However, other contributions, namely the large spheres appearing under the hexagonal seats of small spheres, disturb this extinction.

3.2. Second sample

3.2.1. Structure deduced from the IFFT image. From a different piece of the opal sample numbered 167, we obtained

<table>
<thead>
<tr>
<th>Spot</th>
<th>L</th>
<th>J</th>
<th>H</th>
<th>I</th>
<th>G</th>
<th>F</th>
<th>E</th>
<th>D</th>
<th>C</th>
<th>B</th>
<th>A</th>
</tr>
</thead>
<tbody>
<tr>
<td>( D ) (( \mu )m)</td>
<td>1.249 &amp; 1.73 &amp; 2.535 &amp; 3.054 &amp; 4.232 &amp; 4.627 &amp; 5.054 &amp; 5.486 &amp; 5.92 &amp; 6.346</td>
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<tr>
<td>( d ) (( \mu )m)</td>
<td>0.089 &amp; 0.394 &amp; 0.327 &amp; 0.236 &amp; 0.216 &amp; 0.198 &amp; 0.182 &amp; 0.169 &amp; 0.158</td>
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<tr>
<td>Cell C3</td>
<td>0.6 &amp; 0.8 &amp; 0.12 &amp; 0.14 &amp; 0.20 &amp; 0.22 &amp; 0.24 &amp; 0.26 &amp; 0.28 &amp; 0.30</td>
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<tr>
<td>Cells C2, C3</td>
<td>0.4</td>
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![Figure 6 Central part of the FFT pattern, showing the spots of the superstructure consecutive to the vicinal cut.](image)
a SEM image where lepispheres are visible (Fig. 7); their cores are either empty or constituted by a silica different from that of the external envelope. But the fracture section does not fit with a simple crystallographic section and it is not easy to distinguish a periodicity. This time, the corresponding IFFT pattern is more complicated (Fig. 8(a)). Careful analysis leads us to propose a unit cell reproduced in subvertical bands. However, from one band to the next, a slip of 1/3 of the vertical parameter is observed, so the real periodicity corresponds to three unit cells, horizontally.

Another feature affords a precious detail that can be used to build a model of the fracture: some regularly spaced bands are banked approximately 17° to the left from the horizontal. Along these bands, we can recognize the elementary pattern of the two-layer sheet: a large sphere seated on a hexagon of small spheres. Visibly, the fracture surface is again inclined with respect to the basal plane. Moreover, it is not tilted around a simple row, as was the case for the first vicinal section.

Taking into account all the features displayed by the IFFT image (sphere positions, repetition distances, obvious cells and sets of spheres A settled on a hexagonal seat of spheres B), we reconstructed a bidisperse model, placing both types of spheres, one by one, as closely as possible with respect to the positions observed on the IFFT image. So we were able to propose a structure showing the different steps crossed by the fracture (Fig. 8(b)). The unit cells and their organization found on the IFFT image are also shown on the model. The lattice cells appear rectangular on the model, whereas they are rhombic on the IFFT image. However, this assembly fits with the AlB₂ structure described above.

3.2.2. About the cell distortions. To show that the distortion observed in the lepisphere lattice image depends on the tilt of its normal with respect to the electron beam, we photographed the cross section of a square fiber optic network cable (Gauthier et al., 2017). The first picture was taken with the electron beam normal to the sample (Fig. 9(a)), clearly showing the square lattice. After the sample has been tilted to an angle of 27°, the square mesh visible on the first image appears rhombic (Fig. 9(b)).

This experiment allows an understanding of the discrepancy between the experimental image and structural model. Even if the average surface of the preparation was oriented as precisely as possible perpendicularly to the electron beam, the steps of the sample fracture may be more or less tilted with respect to the electron beam, explaining the observed deformation.
3.2.3. Indexing of the steps and tilt with respect to the basal plane. It is now possible to index the crystallographic plane constituting each step, since they display a periodic arrangement. In Fig. 10, we simplify the preceding model, keeping only the large spheres. We represent the vicinal plane arrangement. In Fig. 10, we simplify the preceding model, keeping only the large spheres. We represent the vicinal plane arrangement. 

\[ H = 2r, \quad \tan \phi = 4/468^{1/2}, \quad \phi = 10^\circ 47, \] (6)

\[ H = 3r, \quad \tan \phi = 6/468^{1/2}, \quad \phi = 15^\circ 50, \] (7)

\[ H = 3.722r, \quad \tan \phi = 7.444/468^{1/2}, \quad \phi = 19^\circ. \] (8)

This time, the vicinal plane is a little more tilted with respect to the basal plane.

4. Discussion

Our method has several limitations, as described by Stephant et al. (2014). First, the resolution of the FFT image strongly depends on the original image resolution and pixel size. The FFT cannot differentiate between patterns if they are too alike (same direction and similar parameters) and interpretation can be complicated when too many patterns are present (overlapping of spots), as arises if there are too many different layers exposed on the sample or if the pattern induced by the steps of layers has similarities to other patterns present in the layers of the opal. Finally, the multi-step process of the method degrades the resolution, which can hide the separation between nearby spots on the FFT image.

These limitations did not cause major problems for this work. The structure of this bidisperse Ethiopian opal has been clearly identified using the fast Fourier transformation. The apparently very poor information in the direct SEM image was treated by FFT, and the periodic arrangement is emphasized by masking the continuous background.

Two vicinal planes have been recorded and indexed. Crossing successive steps of the alternating planes of small and large spheres, they allowed an understanding of their arrangement along the vertical axis and enabled us to determine that the unit cell is similar to the atomic aluminium diboride structure.

However, because the fracturation of CT opals gives a fine-grained surface instead of circular sections of silica spheres as in amorphous opals, and also because the lepispheres are detected under the electron beam more by their core than by
their external shape, the ratio \( R/r \) is difficult to assess. Moreover, the unknown tilt of the beam with respect to the cut section eliminates the possibility to estimate, even roughly, this ratio from vicinal planes.

The stability of the structure depends on the ratio \( R/r \). A discussion was published by Murray & Sanders (1980) after the first discoveries of bidisperse natural opals. For the present \( AB_2 \) structure, where two honeycombs overlapping vertically trap a plane of large spheres, the latter must be settled below and above the hexagonal seats of small spheres. For such isolated sandwiches, the ratio \( R/r \) is bound between two values, determined by the extreme cases of the most stable stackings:

(i) Two planes of small spheres are in contact (Fig. 4a), with \( \tau = 1.236 \).

(ii) The large spheres are in a close-packed arrangement (Fig. 4c), with \( \tau = 3^{1/2} \).

But, between \( \tau = 1.236 \) and \( \tau = 1.50 \), the large spheres would interpenetrate (similarly to a covalent bonding for atoms). Mechanically, that is not possible, so ultimately, the ratio \( R/r \) must be between \( \tau = 1.50 \) and \( \tau = 3^{1/2} \). However, in an earlier study (Gauthier et al., 1995), we encountered this problem in bidisperse Brazilian opal-A, having an MgCu\(_2\) structure. In this structure, a large sphere is located at the centre of a tetrahedron of small spheres with opened hexagonal seats on each face (Fig. 11). The large central sphere touches the 12 closest small spheres and crosses the median plane of the four faces, thus impinging upon the neighbouring large spheres. In that case, the problem was solved by the prevailing pressure during formation, which flattened the large spheres where they met the median planes. Thus, on the SEM images, the large spheres appear ‘polyhedrized’.

In the present case, such a mechanism cannot be observed on the lepispheres and, without any other evidence, we cannot estimate more precisely the domain of possible \( \tau \) values.

If the local pressure does not act in that way during the sedimentation of lepispheres, the domain of \( \tau \) values will be restricted between 1.5 and 3\( ^{1/2} \).

Our method of sample preparation by fracture is a convenient way to reveal the periodic patterns within the material because it allows image contrast between lepispheres and cement, but it does not lend itself to a perfect orientation of the observed surface, thus causing unwanted distortions of the FFT. As far as we know there is no method that can fracture opal in a preferred direction and we were not able to determine a better position for the sample before its introduction into the SEM chamber (assuming that a suitable surface exists on the fractured sample). It might be possible to cut opal with a focused ion beam or another instrument designed to polish a surface with an ion beam. This could produce a perfectly flat surface that is suitable for our method, but it may erase the essential contrast between lepispheres and cement.

5. Conclusion

From SEM images of the fracture surface of an Ethiopian play-of-color opal, an FFT treatment leads us to the following results:

(1) The discovery of a bidisperse structure never seen in natural CT opals, but already obtained on natural opal-A and by synthesis.

(2) The possibility to determine a structure from a two-dimensional IFFT image of vicinal planes. This was previously done from a direct SEM image, but never with an IFFT image.

(3) The determination of the complete structure of the cut section.

However, for this structure, which is a stack of alternating planes of small and large spheres, we are unable to determine the ratio \( R/r \).

In conclusion, the FFT–IFFT method is powerful for highlighting the periodicities in CT opals when the lepispheres are not visible on SEM images. But elucidating a bidisperse structure by this approach requires many randomly recorded images and luck still plays a large role in ultimate success.

References


