Effect of Heat Treatment on Siliceous Rocks Used in Prehistoric Lithic Technology

Article in Journal of Archaeological Science · November 1992
DOI: 10.1016/0305-4403(92)90031-W

CITATIONS
104

READS
901

2 authors, including:

John A. Webb
La Trobe University
175 PUBLICATIONS 3,050 CITATIONS
SEE PROFILE

Some of the authors of this publication are also working on these related projects:

Volcanoes of Western Victoria View project

Geoarchaeology and environmental archaeology View project

All content following this page was uploaded by John A. Webb on 06 July 2018.

The user has requested enhancement of the downloaded file.
Effect of Heat Treatment on Siliceous Rocks Used in Prehistoric Lithic Technology

Marian Domanski and John A. Webb

(Received 28 March 1991, revised manuscript accepted 20 January, 1992)

Stone tool manufacture by many prehistoric and recent societies was characterized by deliberate heating of fine grained siliceous rocks to improve their flaking properties. Extensive mechanical testing of heated and unheated cryptocrystalline and macrocrystalline quartz lithologies has shown that thermal treatment causes a consistent marked reduction in fracture toughness. This mechanical property can be used as an objective measurement of the flaking qualities of stone materials, and a reliable criterion for the recognition of intentionally heated artefacts in the archaeological record. X-ray diffraction studies and scanning electron microscopy have demonstrated that the change in fracture toughness with heating is the result of recrystallization. The poorly ordered, strongly interlocking cryptocrystalline fabric of the unheated samples becomes more equigranular and better crystallized with thermal treatment. As a consequence, fractures propagate more readily in heated samples, accounting for their better flaking properties.

Keywords: HEAT TREATMENT, BLADE TECHNOLOGY, FRACTURE TOUGHNESS, MODULUS OF ELASTICITY, FRACTURE PROPAGATION, SILICA RECRYSTALLIZATION.

Introduction

In many of the prehistoric and recent societies that utilized stone tools, certain lithic materials, particularly fine-grained siliceous rocks, were often deliberately heated to improve their flaking properties. Ethnographic and archaeological evidence suggests that such intentional heating of siliceous materials has been practised in many parts of the world, including western Oceania, northern Australia, Bengal, southern Africa and various parts of North America (Hester, 1972; Flenniken & White, 1983; Olausson & Larsson, 1982). Thermal alteration has received much attention from archaeologists since the first experiments verified that the flaking of siliceous lithologies could be improved by heating (Crabtree & Butler, 1964). The most common advantages attributed to heat treatment are improved flakeability, longer flake removals, fewer step and hinge terminations and production of sharper edges (Crabtree & Butler, 1964; Bleed & Meier, 1980; Rick & Chappell, 1983).

Thermal pretreatment has become a popular subject among prehistorians, but it has proved difficult to define both an accepted criterion for identification of heat-treated
material, and an objective measurement of the changes in working quality of thermally altered rocks. Furthermore, there is no clear understanding of the microstructural changes within the rock causing the changes in flaking properties. This paper sets out to resolve these problems, firstly by determining the mechanical changes accompanying heating, and how these explain the improvement in stone tool production brought about by heat treatment. Secondly, the microstructural changes resulting from heating will be discussed.

**Visual Changes Accompanying Heat Treatment**

Heat treatment has been shown to cause a variety of visual changes in microcrystalline siliceous rocks, and these have been used with varying degrees of success as criteria for the identification of intentionally heated stone artefacts in the archaeological record. The most notable and widespread visual effects are darkening in colour, increased lustre of flaked surfaces and obvious heat damage, such as crazing and microfracturing. Colour changes can be very variable, and are strongly dependent on the rock type (Rick & Chappell, 1983). The most common change is from yellow/brown to dark red (Purdy & Brooks, 1971; Price et al., 1982), and is the result of goethite transforming to haematite during heat treatment (Schindler et al., 1982). Some siliceous lithologies, particularly those that are dark in colour to begin with, show little change with heating (Flenniken & White, 1983; Hanckel, 1985). It has been proposed that colour changes would not occur unless suitable iron-containing minerals were present in the material (Purdy, 1974).

Heated siliceous rocks usually have a greasy lustre on flaked surfaces (Purdy & Brooks, 1971), and this may be the most consistent, distinctive heat-induced change visible to the naked eye. However, experimental studies indicate that only a relatively small percentage of flakes may have this lustre (Collins, 1973), and the coarser grained lithologies like silcrete and quartzite may show little variation in lustre with heating (Hanckel, 1985). Weathering and bioturbation in the soil may remove the lustre (Price et al., 1982), so only recently fractured surfaces should be examined for evidence of heat treatment. In addition, a greasy lustre can be caused by other processes like wind polish.

The increased lustre is related to heat-induced microstructural changes within the silica, and these changes are also responsible for the improvement in the mechanical and flaking properties of the heat-treated material. The relationship between heat treatment and flaking of siliceous rocks was initially demonstrated by skilled flint-knappers (Crabtree & Butler, 1964), and subsequently there have been only a few, limited studies on the mechanical properties of heated stone (Purdy & Brooks, 1971; Purdy, 1974; Schindler et al., 1982; Rick & Chappell, 1983). No mechanical test has yet been identified that adequately characterizes the flaking properties of stone (Kamminga, 1982). The need for such a test is illustrated by the place of obsidian in prehistoric lithic technology. Obsidian is regarded as the lithology with the best flaking qualities (Bordes, 1969). It has been proposed that heat treatment of fine grained siliceous rocks yields a material judged second only to obsidian in the manufacture of blades and pressure flaked points (Crabtree & Butler, 1964; Ahler, 1982). Thus there should be a mechanical property of fine grained siliceous rocks which changes as they are heated, to give values typical of obsidian.

**Mechanical Changes Accompanying Heat Treatment**

In an attempt to define an accurate standard mechanical test for quantifying improvement in the flaking qualities of thermally altered stone artefact materials, four properties were assessed: elastic constant, compressive strength, tensile strength and fracture toughness. The standard tests for these four properties, as used in rock mechanics engineering, were applied: Young's modulus of elasticity, uniaxial unconfined compressive strength, Brazilian tensile strength and chevron notch fracture toughness respectively (Gunsallus &
Kulhawy, 1984; International Society for Rock Mechanics, 1988). Separate specimens of each lithology tested were gradually heated in an oven to 300, 400, 500 and 600°C, held at the peak temperature for 2 h, and then allowed to cool slowly. These temperatures were selected because they can be easily reached in the ash of an ordinary campfire (Griffiths et al., 1987). The samples were then subjected to each of the four rock mechanics tests; altogether over 2000 specimens were tested. A complete discussion of the methodology used in these investigations and the full results of the experimental work will be published elsewhere.

The lithologies tested belong to two groups: cryptocrystalline (extremely fine grained) quartz, including flint, chert, jasper, chalcedony and agate, and macrocrystalline quartz, including silcrete, quartzite and crystal quartz. In the latter grouping, samples usually contain quartz crystals visible to the naked eye, although in some silcretes these are embedded in a microcrystalline matrix. None of the samples contained any opal-A or opal-CT; quartz was the only silica mineral present. This was verified by X-ray diffraction analyses of all samples.

Replicate mechanical tests on separate samples of the same lithology often gave different results, confirming earlier observations of a considerable variation in the flaking properties of a given rock type (Crabtree & Butler, 1964; Rick & Chappell, 1983). In rock mechanics engineering, at least 10 replicate tests on a particular sample are recommended, in order to assess the variation in a particular mechanical property (Yamaguchi, 1970). Because of the limited size of many of the rock samples available in the present study, it was impossible to prepare enough specimens to carry out 10 tests for each property. A minimum of three tests was performed for each sample, and more specimens were tested wherever possible. The average from a series of tests is presented as the median, because this is less affected by outlying values than the mean (Rock et al., 1987). As all the data sets are so small, confidence intervals were calculated using the non-parametric asymmetric method (Rock et al., 1987); classical statistical methods like standard deviation are inapplicable, because the underlying population is unknown, and cannot be assumed to be normal.

Of the four properties tested, compressive strength and tensile strength values did not show consistent changes as a result of heat treatment, varying substantially with the raw material. However, the modulus of elasticity and fracture toughness results displayed a consistent pattern.

Modulus of elasticity measures the stiffness of a material, so it indicates the suitability of a lithology for blade manufacture, because stiffness-controlled fracture propagation is basically responsible for blade detachment (Cotterell & Kamminga, 1987). The chert, flint, agate, quartzite and silcrete samples used in the heat treatment experimentation showed a slight, consistent increase in the Young's modulus of elasticity when heated to 300, 400 and 500°C. Thus, these materials, which are often used in blade technology, become slightly stiffer and therefore easier to flake after controlled heat treatment.

Fracture toughness is a measure of the resistance of a material to fracture propagation (Lawn & Wilshaw, 1975), and has been regarded as the most important mechanical property of raw materials used in prehistoric lithic technology (Cotterell & Kamminga, 1987). The present study showed that the fracture toughness of eight different cryptocrystalline and macrocrystalline samples decreases after heat treatment (Table 1, Figure 1). The cryptocrystalline samples (chert, agate, flint and jasper) almost all displayed a marked reduction in fracture toughness when heated to 300 and 400°C. Many of the samples heated above this temperature suffered extensive microfracturing (crazing) and excessive brittleness. Similar damage was induced by very rapid heating or cooling. The coarser grained macrocrystalline samples (quartzite, silcrete, crystal quartz) also showed a significant reduction in fracture toughness with increased temperature. However, many of
Table 1. Changes in fracture toughness of siliceous rock samples with heat treatment. Samples are representatives of the 30 different lithologies tested. Percentage change always calculated with reference to unheated specimens. Tests on the quartz specimens heated to 500 °C are invalid because the fractures did not propagate along the notches.

<table>
<thead>
<tr>
<th>Sample number</th>
<th>Rock type</th>
<th>No. of specimens</th>
<th>Fracture toughness before heat treatment (N/mm²)</th>
<th>Fracture toughness after heat treatment (N/mm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>300°C</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>No. of specimens</td>
<td>Median and 95% uncertainty</td>
</tr>
<tr>
<td>119</td>
<td>Chert</td>
<td>8</td>
<td>48.54 ± 1.92</td>
<td>-3.01</td>
</tr>
<tr>
<td>196</td>
<td>Silcrete</td>
<td>11</td>
<td>80.45 ± 2.79</td>
<td>-2.19</td>
</tr>
<tr>
<td>208</td>
<td>Flint</td>
<td>10</td>
<td>65.85 ± 7.65</td>
<td>-7.68</td>
</tr>
<tr>
<td>223</td>
<td>Quartzite</td>
<td>15</td>
<td>54.85 ± 2.74</td>
<td>-5.67</td>
</tr>
<tr>
<td>232</td>
<td>Quartz</td>
<td>7</td>
<td>46.67 ± 5.05</td>
<td>-10.11</td>
</tr>
<tr>
<td>242</td>
<td>Flint</td>
<td>12</td>
<td>51.52 ± 3.02</td>
<td>-3.02</td>
</tr>
<tr>
<td>324</td>
<td>Agate</td>
<td>3</td>
<td>81.96 ± 14.78</td>
<td>-11.43</td>
</tr>
<tr>
<td>333</td>
<td>Jasper</td>
<td>9</td>
<td>72.41 ± 10.01</td>
<td>-6.88</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>400°C</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>500°C</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>119</td>
<td>Chert</td>
<td>8</td>
<td>48.54 ± 1.92</td>
<td>-3.01</td>
</tr>
<tr>
<td>196</td>
<td>Silcrete</td>
<td>11</td>
<td>80.45 ± 2.79</td>
<td>-2.19</td>
</tr>
<tr>
<td>208</td>
<td>Flint</td>
<td>10</td>
<td>65.85 ± 7.65</td>
<td>-7.68</td>
</tr>
<tr>
<td>223</td>
<td>Quartzite</td>
<td>15</td>
<td>54.85 ± 2.74</td>
<td>-5.67</td>
</tr>
<tr>
<td>232</td>
<td>Quartz</td>
<td>7</td>
<td>46.67 ± 5.05</td>
<td>-10.11</td>
</tr>
<tr>
<td>242</td>
<td>Flint</td>
<td>12</td>
<td>51.52 ± 3.02</td>
<td>-3.02</td>
</tr>
<tr>
<td>324</td>
<td>Agate</td>
<td>3</td>
<td>81.96 ± 14.78</td>
<td>-11.43</td>
</tr>
<tr>
<td>333</td>
<td>Jasper</td>
<td>9</td>
<td>72.41 ± 10.01</td>
<td>-6.88</td>
</tr>
</tbody>
</table>
Figure 1. Changes in fracture toughness of siliceous rock samples after heat treatment. Median values plotted; see Table 1 for uncertainties.

Table 2. Fracture toughness of obsidian

<table>
<thead>
<tr>
<th>Sample number</th>
<th>Rock type</th>
<th>Collection locality</th>
<th>No. of specimens</th>
<th>Fracture toughness (N/mm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Median and 95% uncertainty</td>
</tr>
<tr>
<td>235</td>
<td>Obsidian</td>
<td>Lost Valley, Umleang, Lou Island, P.N.G.</td>
<td>11</td>
<td>24.01 ± 3.32</td>
</tr>
<tr>
<td>236</td>
<td>Obsidian</td>
<td>Lost Valley, Umleang, Lou Island, P.N.G.</td>
<td>17</td>
<td>26.30 ± 1.18</td>
</tr>
<tr>
<td>237</td>
<td>Obsidian</td>
<td>Lost Valley, Umleang, Lou Island, P.N.G.</td>
<td>17</td>
<td>27.42 ± 1.10</td>
</tr>
<tr>
<td>240</td>
<td>Obsidian</td>
<td>Glass Butte Series, Lake County, Oregon, U.S.A.</td>
<td>6</td>
<td>25.82 ± 1.38</td>
</tr>
</tbody>
</table>

these samples required higher temperatures than cryptocrystalline silica (at least 500°C) to achieve the same changes in fracture toughness. Almost none of the macrocrystalline samples revealed any effects of overheating, even at 500 and 600°C.

The clearly marked reduction in fracture toughness with temperature, consistent for all siliceous lithologies tested, indicates that this property is a reliable method for quantifying the heat-induced mechanical changes within these materials. As the decrease in fracture toughness correlates with an improvement in flaking properties, fracture toughness can be regarded as an objective measurement of the flaking quality of stone. Furthermore, the present tests also show that the fracture toughness of heated cryptocrystalline quartz is similar to that of obsidian (Table 1, 2; note particularly the heated samples of chert and
fracture toughness can be used to assess the relative quality of different rock types for artefact manufacture, particularly for blade tools.

In addition, fracture toughness may provide a reliable criterion for the recognition of intentionally heated artefacts in the archaeological record. For this test to be definitive, comparable values of fracture toughness need to be obtained from unheated samples of the same lithology. If this material is not available, as is often the case, a preliminary indication of heat treatment can be obtained by comparing the artefact’s fracture toughness with that of heated and unheated samples of similar lithologies tested during the present study (Table 1). However, the main drawback to the widespread fracture toughness testing of artefacts is the fact that the size of sample required (a cylinder 15 mm in diameter and 21.75 mm long) will inevitably result in at least partial destruction of the artefacts tested. Fracture toughness testing is perhaps best used to confirm an initial suspicion of heat treatment based on visual indications, particularly lustre. It would then only be necessary to test one of a number of artefacts with very similar lustre, in order to confirm that heating had occurred.

The chevron notch fracture toughness test is also useful in that it results in well-defined fracture surfaces. This makes it very easy to define the validity of each test. In a valid fracture toughness test, the fracture initiates from a well-defined point, the tip of the chevron notch, and propagates along the notches. The test also provides surfaces ideal for microstructural studies.

**Microstructural Changes Accompanying Heat Treatment**

The changes in mechanical properties and fracture propagation in heat-treated silica must reflect significant modifications of the microstructure within the material. In order to investigate this more fully, attention was focused on two samples of cryptocrystalline silica, agate (324) and flint (208 and 209), that showed the greatest reduction in fracture toughness after heat treatment to 400°C, without suffering any damage from overheating (Table 1). Flint specimens 208 and 209 represent the same material. The agate and flint samples were examined by X-ray diffraction, infra-red absorption spectroscopy and scanning electron microscopy.

Previous workers have proposed five different ways in which heat-induced microstructural modifications may have occurred. These will now be discussed in turn, assessing each in the light of the results obtained during this study.

The most widely quoted theory invokes recrystallization of the chalcedonic matrix in flint, so that the fossil fragments and lepispheres are bound more tightly (Purdy & Brooks, 1971; Olausson & Larsson, 1982; Bradley & Clayton, 1987; Griffiths et al., 1987). During heat treatment, impurities in the matrix are believed to act as a flux, effectively welding together the whole flint structure. As a result, fractures propagate equally through the lepispheres, fossil fragments and matrix, giving smooth fracture surfaces. In untreated flint, the fractures run preferentially through the matrix and around the lepispheres and fossils, so that fracture surfaces appear granular under the scanning electron microscope.

Although this explanation has achieved wide currency in the archaeological literature, it does not explain how welding together the silica microstructure more tightly could result in a decrease in fracture toughness; intuitively, the opposite would be expected. It is also unclear how this theory would apply to siliceous lithologies which lack a chalcedonic matrix around larger grains, e.g. many varieties of chert, jasper and silcrete. Thus, this theory would appear to suffer from a number of problems. Furthermore, the scanning electron micrographs on which this hypothesis is based are almost all of unetched surfaces. The details of the microstructure of siliceous lithologies are only clearly visible when the surfaces have been etched with hydrofluoric acid for a few minutes.
The second theory involves the role of water in the microstructure. During heat treatment the water held within the silica lattice and along grain boundaries is believed to migrate into microfluid inclusions (Griffiths et al., 1987). This water movement causes microfracturing, which decreases the mechanical strength of the material. NMR (nuclear magnetic resonance) spectroscopy of one flint sample showed a broad liquid water peak when heated to 380°C, but lacked this peak when heated to 280°C or less (Griffiths et al., 1987).

There is little doubt that water movement occurs during the heat treatment, and several authors (e.g. Purdy & Brooks, 1971; Mandeville, 1973) have recorded water losses on heating. The amount of water lost varies between samples (Collins & Fenwick, 1974), and heated specimens frequently take up water again on cooling (Purdy, 1974), so the significance of the water movement is uncertain. In order to check this, infra-red absorption spectra were run on the two selected samples of agate and flint, both of which showed major changes in fracture toughness on heating. Infra-red absorption spectroscopy is useful for detecting water loss in silica samples, because the strengths of several peaks in the absorption spectra are proportional to the amount of water present (Langer & Florke, 1974). The 950 cm⁻¹ peak in the quartz infra-red spectrum has been ascribed to the Si-O stretching of SiOH groups (Moenke, 1974), and the 1630 cm⁻¹ peak results from the H-O-H bending vibrations of molecular water (Langer & Florke, 1974). The spectrum of heated agate (Figure 2(a)) clearly shows a reduction in the intensity of the 950 cm⁻¹ shoulder, reflecting a loss of water bound into the silica lattice as OH groups. However, the heated flint sample has a larger 950 cm⁻¹ peak than the unheated sample (Figure 2(b)); presumably it re-absorbed water on cooling. There is a slight decrease in the 1630 cm⁻¹ peaks of both lithologies, representing a drop in the free water content with heating. Thus there is no evidence for the migration of water into microfluid inclusions after heat treatment, and indeed the role of water in the heat-induced microstructural changes would appear to be minimal. As all the samples used in the present and previous studies were composed of quartz, the amount of water contained within their structures is very small (Wilding et al., 1977), and its loss would be unlikely to affect the mechanical properties of the material significantly.

The third theory for the microstructural changes proposes that the heating induces microcracking or local strain, and effectively breaks up the silica crystals (Flenniken & Garrison, 1975). An X-ray diffraction analysis showed a broadening of the 324 quartz peak in heated chert samples (Weymouth & Mandeville, 1975), and this was assumed to be due to the breakup of the quartz crystals within the chert. This result would be consistent with microfracturing (Rick & Chappell, 1983). Almost all authors who have studied heat treatment found that high temperatures, or too rapid heating or cooling, will result in crazing and microfracturing (e.g. Crabtree & Butler, 1964; Griffiths et al., 1987). However, this effect has not been noted at lower temperatures, even on scanning electron micrographs of fracture surfaces. In any case, detailed X-ray diffraction studies during the present study (Figure 3) clearly show an increase rather than a decrease in crystal order after heating (see discussion below).

The fourth explanation was based on jasper from Bald Eagle, Pennsylvania, U.S.A. (Schindler et al., 1982). This jasper contains small amounts of scattered goethite, which recrystallizes to haematite on heating; relatively low temperatures (< 100°C) are required for this transformation (Langmuir, 1971). It was postulated that heating caused the haematite crystals to be concentrated in "channels", along which fractures propagated, and this explained the greater ease of flaking of heat-treated material (Schindler et al., 1982). Although this hypothesis might be applicable to the jasper studies, it cannot explain the same changes in mechanical properties that occur in iron-free lithologies like many flints, cherts and chalcedonies.
Figure 2. Infra-red absorption spectra of heated and unheated samples of (a) agate - 324 and (b) flint - 209. Arrows indicate position of peaks due to water.
Figure 3. X-ray diffraction traces for heated and unheated samples of (a) agate -324 and (b) flint -209.
The most likely explanation is that heating causes "recrystallization of the ... silicic materials, which results in reduced crystal size" (Crabtree & Butler, 1964:2), and this is confirmed by X-ray diffraction data and scanning electron micrographs obtained during the present study. The X-ray diffractograms, run on a high resolution Scintag Pad V Diffractometer, show differences in the 212 peaks (1.3820 Å) in the spectra of the heated and unheated specimens (Figure 3). The relative intensities of these peaks can be used to calculate a crystallinity index for quartz; this is largely a function of crystal size (Murata & Norman, 1976). Large euhedral quartz crystals have indices of 8.0–10.0, whereas the index of poorly crystallized cryptocrystalline quartz ranges from 1.0 to 3.0. The crystallinity indices of the raw and heated samples of agate and flint illustrate the microstructural changes which occur when cryptocrystalline siliceous rocks are heated. The crystallinity of the unheated agate is 4.4; that of the agate heated to 400°C is considerably greater, 8.7. A smaller trend is shown by the flint; the index increases from 1.2 to 2.2. This is evidence of recrystallization of the microcrystalline silica in the unheated samples into better ordered crystals after heating. This recrystallization can be detected only in the high angle peaks of the X-ray diffractograms, and is most easily recognizable in the 212 peaks. Most previous workers have found no significant changes in the diffraction patterns of materials subjected to heat treatment (e.g. Purdy & Brooks, 1971), because they did not examine the 212 peaks.

This recrystallization is also undetectable by thin section examination under the standard petrological microscope. However, under the scanning electron microscope obvious changes are visible on fractured, etched surfaces of raw and heated samples of the agate and flint. The unheated agate specimen is characterized by tightly packed, interlocking fibres of chalcedonic silica (Figure 4(a)). In the sample heated to 400°C, these fibres are transformed into more granular, equidimensional crystals of quartz, with well-defined crystal faces (Figure 4(b)). This increase in crystallinity correlates with the higher crystallinity index of the heat-treated sample. A similar recrystallization is observed in the heated flint (compare Figures 5(a) and (b)), although it is less obvious, and this is reflected in the small increase in crystallinity index after heating. Nevertheless, more equidimensional crystals of quartz, with clearly defined faces, can be seen in the specimen of flint heated to 400°C. It should be noted that other samples of flint from different sources might well show more pronounced recrystallization.

The recrystallized samples are structurally different to the original lithologies, and will therefore respond differently to mechanically induced stress. Thus the recrystallization is responsible for the changes in mechanical properties and fracture propagation in the heat-treated siliceous artefact materials. The resistance to the passage of a crack through a
brittle elastic solid is determined by the energy of the cohesive bonding between the constituent atoms or molecules along the path of the fracture (Lawn & Wilshaw, 1975). Fracturing within micro- or macrocrystalline quartz is more likely to occur along crystal boundaries rather than through crystals, because the bonding between crystals is much weaker than the very strong interatomic silicon–oxygen bonds within the silica lattice. The shape and size of individual quartz crystals will therefore determine fracture propagation. In unheated agate, fractures must break through the strongly interlocking fibrous crystals of chalcedony. Heat-treated agate is composed of shorter, more equigranular quartz crystals, which interlock less strongly. Fractures can propagate around these crystals more easily than in unheated agate, so the fracture toughness of the heated material will be reduced. Similarly, fractures will propagate more readily in heated flint, which is also composed of more equidimensional crystals, thereby decreasing the fracture toughness of this lithology.

This explanation of the heat-induced microstructural changes is applicable to all heat-treated siliceous lithologies, and can also account for the observations of previous authors. For example, the flint described by many authors (Purdy & Brooks, 1971; Olausson & Larsson, 1982; Griffiths et al., 1987) as composed of lepispheres, skeletal fragments and chalcedonic matrix, is made up of different grain sizes and shapes of microcrystalline quartz. Heat-induced recrystallization will cause the quartz crystals of the different components to be more similar in size, so that fractures will propagate equally well through lepispheres, fossils and matrix, giving smoother, more lustrous fractures. This was not recognized by most previous authors because they did not examine etched fracture surfaces. The only etched surfaces of heated flint to be illustrated (Griffiths et al., 1987) have a granular appearance, and this is more easily interpreted as the result of recrystallization rather than the etching out of submicroscopic fluid inclusions (see discussion above).

In the case of the iron-rich Bald Eagle jasper (Schindler et al., 1982), the recrystallization of the goethite to haematite may well contribute to the changes in mechanical properties, but the dominant cause is still likely to be the recrystallization of the silica itself. Tools produced after the heat treatment of chert have sharper edges (Rick & Chappell, 1983). As finer grained lithologies give sharper edges, this observation is easily explained if the recrystallization during heat treatment gives rise to finer, more even grained textures in the silica of the tool material.

Compared to the microcrystalline artefact materials, the coarser grained macrocrystalline siliceous lithologies show a more gradual decrease in fracture toughness with
heating (Table 1). This fact has been noted by other authors (e.g. Hanckel 1985). Whereas cryptocrystalline quartz suffers heat-induced microfracturing above 400°C, temperatures as high as 600°C do not cause significant heat damage in macrocrystalline samples. Presumably the coarser grain size of these lithologies means that heat-induced recrystallization will be less marked, and there will be correspondingly smaller changes in the mechanical and flaking properties. In the case of metamorphic quartzite, the texture was formed at high temperatures, and is unlikely to be affected by further heating. Because the macrocrystalline materials are less sensitive to the temperature applied and the rate of heating and cooling, they allow the stoneknapper a better control of the heat treatment process.

Conclusions

Heating of cryptocrystalline siliceous artefact lithologies causes a consistent marked reduction in fracture toughness, which accounts for the improvement in flaking properties. Fracture toughness can be regarded as the best independent measure of the flakeability of an artefact material. The changes in properties during heating are the result of the original poorly ordered, strongly interlocking microfabric becoming more equigranular and better crystallized.

These results suggest several ways in which intentionally heated artefacts can be recognized in the archaeological record. Firstly, a preliminary indication of heat treatment is likely to be given by the visual appearance of the artefact. An increased, greasy lustre on flaked surfaces is the most consistent feature, although this needs to be compared to unheated material of the same lithology and from the same source for a definite indication. Mechanical testing will determine if this initial suspicion is correct: thermally treated cryptocrystalline siliceous material will have a reduced fracture toughness compared to unheated samples of the same lithology. Although this is probably the most reliable method for recognizing heat treatment, it suffers from the drawback that the size of sample required will inevitably result in partial destruction of the artefact tested.

Less destructive ways to identify heat-treated artefacts are scanning electron micrographs of etched fracture surfaces and X-ray diffraction. The presence of moderately equidimensional crystals of quartz with well defined faces in a cryptocrystalline siliceous lithology could indicate that it has been subjected to heat treatment, particularly if the sample also has a high index of crystallinity. However, for positive identification, comparison with unheated samples of the same lithology would be necessary.

Acknowledgements

We would like to thank the following people for assistance with the analyses and testing: A. Marshall (SEM—Zoology Department, La Trobe University), T. Finlayson (XRD—Physics Department, Monash University), I. Potter (IR—Chemistry Department, La Trobe University), R. Frank (sample preparation—Archaeology Department, La Trobe University), T. Ryan (sample preparation—Department of Geology, La Trobe University), R. Kajewski, R. Cox and J. Boland (modulus of elasticity and compressive strength—Division of Geomechanics, CSIRO), J. Griffiths (tensile strength—Department of Materials Engineering, Monash University) and W. Bamford (fracture toughness—Department of Civil Engineering, Melbourne University).

References


