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The formation process of « use-wear polishes »

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RÉSUMÉ

Les traits diagnostics des caractères optiques des « polis » ont été reconnus au microscope électronique à balayage. Le développement progressif des « polis » a été observé par placage d'argent et par stéréo-photographie. Les processus se sont avérés définis par l'usure en tribologie. Deux types d'usure ont été observés : type de coupe et type de fracture. Ni accumulation de silice ni fluidité de surface n'ont été observées. L'hypothèse du gel de silice est donc rejetée. Une hydratation de l'ordre de moins d'un micron peut se produire en surface, mais il est admis que le diagnostic des caractéristiques d'un « poli » est principalement produit par la dureté, les propriétés visqueuses et élastiques ainsi que la texture superficielle des matières travaillées.

ABSTRACT

Diagnostic features of optical polish types were recognized with a scanning electron microscope. The progressive development of the « polishes » were observed with the use of silver-coating and stereo-photography. The processes proved to be wear defined in tribology. Two types of wear were observed : cutting type accompanied by striations and fracture type accompanied by pitting. Neither deposition of silica gel layer nor fluidity of the surface weres observed. The silica gel hypothesis is thus denied. Hydration on the order of less than a micron may occur on a surface, but it is believed that the diagnostic features of « polish » types are mainly produced by the hardness, the viscous and elastic properties, and the surface texture of the worked materials.

Introduction

The establishment of the periodical use-wear meeting shows that experimental use-wear studies, especially investigations of « polish », have become popular. Many archaeological studies with « polish » analysis have been reported. Still, the question of why such a « polish » is produced is not agreed upon. Even its recognition is still problematical. Most researchers, in other words, are investigating a topic which they do not understand completely.

A « polish » is often defined in terms like « an altered flint surface which reflects light and which cannot be removed with acids, bases and solvents » (Vaughan, 1981 : 132). That is to say, use-wear analysts cannot define our basic phenomenon in a positive way. This makes the entire study of « polish » somewhat tentative .

The problem of understanding the « polish » mechanism is the basis of all the questions faced by those of us who do use-wear analysis. Does a « polish » type represent the type of materials worked ? How might a « polish » be altered by natural weathering ? Is this study of « polish » applicable to various rock types ? What features should be selected to measure in the computer image analysis for quantification of « polish » ?

The most popular explanation of « polish » among current use-wear analysts is the « silica gel theory » put forth by Anderson (1980 ; see also Mansur-Franchomme, 1983). In this paper, I used scanning electron microscope (SEM) observations to show that a « polish » is produced by « wear » (defined in the field of tribology), and not by the mechanisms proposed by the silica gel theory. Before that, I will explain the terminology used here, and summarize the major competing theories on « polish » formation presented so far. Then I will show how a « polish » looks under the SEM, and discuss the development process of « polish », in order to evaluate the silica gel explanation of the phenomenon.

Terminology

In this paper, I consistently use terms in the meaning defined in tribology which is « the science and technology of interacting surfaces in relative motion and of the practices related thereto. » (OECD, 1969 : 62).

In use-wear studies, the word « wear » implies various phenomena, including « polish » and micro-flaking. In tribology, however, « wear » is defined as « the progressive loss of substance from the operating surface of a body occurring as a result of relative motion at the surface » (OECD, 1969 : 64). Use-wear researchers tend to use the word « abrasion » for this same meaning. In tribology, abrasion – or abrasive wear – refers to a type of wear « by displacement of material caused by hard particles or hard protuberances » (*op. cit.* : 13). Microflaking is, then, a type of fracture, not wear.

Theories on « polish » formation

« Corn gloss », the most distinctive type of use-wear « polish », was recognized by the end of the last century (Diamond, 1979). Other types of « polish » were not recognized until Keeley established the « high power approach » in the late 1970s.

Semenov (1964 : 14) classified « frictional wear » in three categories on a scale from polishing to grinding and finally rasping. In this system « polish » suggests plastic deformation.

A well-known paper by Witthoft (1967) explains corn gloss is a product of fusion on the flint surface developed by high frictional heat, with a large amount of transferential fused silica from the plant body accumulated on the flint surface. This process increases the mass and volume of a stone tool.

In the papers of the first use-wear conference (Hayden ed., 1979), three authors discussed mechanisms of « polish » formation.

First, Kamminga (1979) divided « polish » into « phytolith polish » and « abrasive smoothing ». The former corresponds to corn gloss, and, with SEM observation, a glass polishing model was used to explain its formation. The smooth surface of a corn gloss was said to be the result of chemical wear which removes the soft, hydrated silica gel layer. Phytoliths were suggested to act as abrasives in the process. The « abrasive smoothing » is a worn surface which is produced by foreign dust, sand, micro-chips of stone edge and so on. It is said to be « more akin to the grinding that is preparatory to optical polishing » (Kamminga, 1979 : 151).

Diamond (1979) proposed a combination of « very mild and mild abrasive wear », « fatigue breakage wear » and « surface fatigue wear » to explain « polish ». Finally, Del Bene (1979) divided « polish » into « depositional polish » and « mechanical polish ». He said the former is produced by deposition of some foreign substance such as opaline phytolith, while the latter is produced by « abrasion » and « translocation ». There are no appropriate existing models to explain abrasion and translocation, but, since striations are not frequently observed under SEM, Del Bene suggested that abrasion does not seem to be a dominant factor in « polish » formation. These three authors explained the « polish » formation in tribological terms, although they do not use the term tribology. However, all of them tried to apply existing models without extensive observation of use-wear « polish » itself.

The silica gel theory was originally proposed by Anderson (1980), who undertook extensive SEM studies based on Keeley's high power approach. Her understanding has undergone some change (Anderson-Gerfaud, 1986) but, according to original theory, a « polish » is a « silica gel » layer produced by hydration of a flint surface. It does not mean that the hydrated layer is removed by friction. On the contrary, the layer can reach about 10 m thick, and inorganic micro-residues such as phytoliths are often found to be sinking into the layer. In the case of plant working, transferential silica gel from worked plants is also merged into the layer, although the amount cannot be evaluated.

This theory was supported by M. E. Mansur-Franchomme (1983), who proposed an additional hypothesis of striation formation based on the silica gel theory. It proposed that a developed « polish » entered a fluid state during the work, so that scratched grooves which may have survived on the surface disappear .

Meeks *et al.* (1982) presented a contrary position when they reported that they could not detect « polish » layer with SEM within their resolution on cross sections of carefully prepared specimens. They concluded that the built-up appearance of polished surfaces is an illusion caused by rounded edges of fracture scars. They also indicated that « the optical illusion that the gloss has some depth is caused by the transparency of the flint and the fact that the edges of abrasion features are rounded by polishing » (Meeks *et al.*, 1982 : 337). They explained striations and pits as products of wear, and that gloss is the result of a concurrent polishing process in which some chemical change of silica has occurred. Furthermore, they succeeded in producing polished surface similar to corn gloss with metallurgical polisher impregnated with fine diamond paste.

Masson *et al.* (1981) argued that the silica-gel theory underestimates wear. Based on knowledge of relevant diciplines, such an amorphous layer on the surface must be on an Angstrom or nm scale. In practrice, they could not observe amorphous patterns with X-ray diffraction, and, considering the nature of amorphous silica, they concluded that it cannot survive on archaeological specimens. Even if it is detected, it may be the result of post-depositional alteration. They think that, by changing the plasticity of materials worked, moisture affects « polish » development by increasing the contact area between the tool edge and and the worked surface.

Unger-Hamilton (1984) called attention to the possibility of misindentification of original rock inclusions as residues of use. Still, the existence of a very thin amorphous layer, on an Angstrom scale, which cannot be detected with X-ray diffraction cannot be denied, so that she felt « polish » is produced by both attrition and deposition.

In sum, then, the two competing explanations of « polish » can be termed the « silica gel theory » and the « wear theory » (fig. 1). At this point the silica gel theory probably has more supporters, but neither theory has been conclusively proved and neither can be entirely rejected.

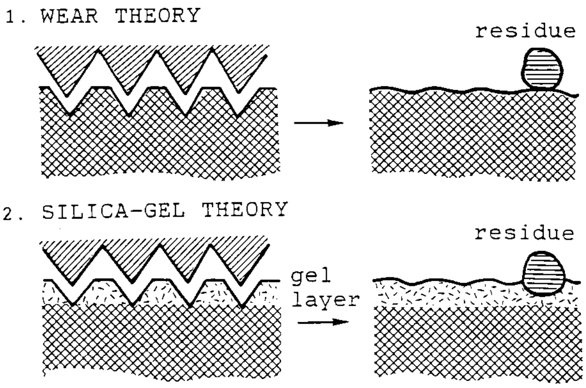

Fig. 1. Theories on « polish » formation.

Method

The specimens shown in table 1 were observed with SEM (JEOL, JSM T-200) and metallurgical microscope (Olympus BHM). To assure accuracy, many photos of each specimen were taken, many at the same spot. Most tests of the observed edges involve movements which were parallel to rather than across the margin.

Siliceous shale was used in most of this test. It is a grey-colored, siliceous deposit which was the prevalent raw material for prehistoric tools in northeastern Japan. Although it is called shale, it is highly silicified and does not have a lamina structure. If it were more silicified, it would be chert, since it is close to chert, but includes more inpurities. Essentially the same use-wear « polish » patterns form on this material and European « flint » (Kajiwara, Akoshima, 1981) so that the result of the present analysis can also be applied to European « flint ».

Siliceous shale	Number of specimens	Number of photos
Worked materials		
unused	3	69
grass	6	365
wood	1	100 including 50 for whittling
soaked antler	6	238 including 40 for whittling
boiled bone	2	130
dry hide	3	165
wet hide	3	118
siliceous shale	2	90
Chert		
wet hide	2	80
Total	28	1355

Table 1. List of experimental specimens.

The features of polished surfaces under SEM

Two closely related questions must be addressed initially : how does « polish » look under SEM ? Can we expect to see similar diagnostic features based on the worked material as have been seen under the optical light microscope (OLM) ?

To answer the first question, images of the same spot taken under SEM and OLM were compared. Next, detailed features were observed under high magnifications of SEM. Such observations show the nature of the respective images which we have to understand in order to interpret « polish » surfaces.

Under the SEM, polished areas which are bright and white under the OLM look relatively dark and do not allow quick identification (fig. 2). This is caused by the « edge effect » of SEM. That is, protrusions and edges of specimen generate more electrons so that worn flat surface becomes relatively dark while unaltered granular surfaces become bright.

Also, by their excellent focus depth, SEM images look like a picture without shadows. This decreases the three-dimensional impression of the image. Under OLM, many people get the impression of an additive polish. This is an illusion caused by the shallow focus depth of OLM. It exaggerates three-dimensional impressions of the subject. If you see the same spot under SEM, it is clear that at least there is not so thick a surface layer as suggested by OLM.

Striations are also exaggerated under OLM. One of the facts that denies the wear theory is that there are no striations on « corn gloss » when observed with SEM. However, this too is an illusion caused by the SEM image and inadequate observation (fig. 3). If there are no striations on corn gloss, why are they so often observed with OLM ?

Observations with SEM further show us an important aspect of the nature of striations. Generally, striations are understood as secondary scratches on a polished surface and are treated as separated from a « polish ». They are believed to be produced by external abrasive agents such as microchips from stone edge or sand grains. However, SEM observations show that « polish » and striation are not separate things. A polished surface consists of numbers of fine striations, although secondary scratches do exist (fig. 4). If you observe the magnified image of one striation, numbers of finer striations are found inside (fig. 5).

At Tohoku University, all the linear features, that is, « scratches » as well as « polish bands », have been defined as striations under OLM, because they are often difficult to separate. The SEM observations described above suggest that our definition was appropriate.

Small circular pits – or « pot-lids » – which are frequently observed under OLM on « polish » produced by dry hide working are another illusion caused by OLM. Under SEM they clearly can be seen to be naturally occurring pits created by friction or the nature of the rock (fig. 2).

In general, differences in the polished surface corresponding to the materials worked have not been reported under SEM. However, if we observe the polished surfaces carefully under SEM, paying attention to the nature of the SEM image, we can find variations of polished surfaces. They are simply not easy to identify unless one is used to the SEM images. Especially « plant polish », which gives a domed impression under OLM, is actually found to be a domed polished surface, and « bone polish », which gives a flat impression under OLM, is found to be a flat polished surface (fig. 6). The difference in profile of polished surfaces was also demonstrated with a roughometer by Akoshima and Kajiwara (1984). At higher magnifications, the scope of the observations becomes narrower so

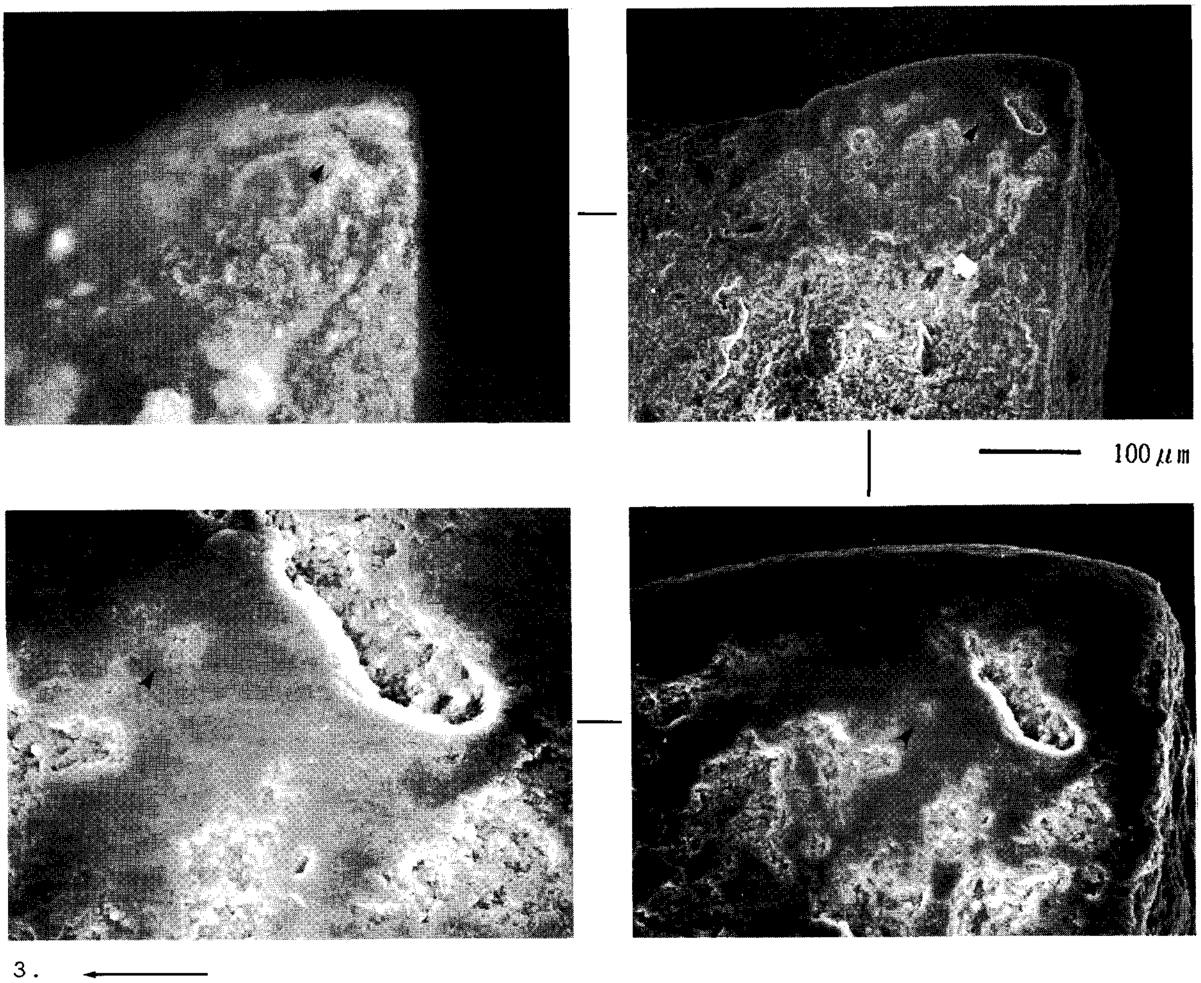

Fig. 2. Comparison of OLM and SEM images. All photos in this paper were taken on siliceous shale.

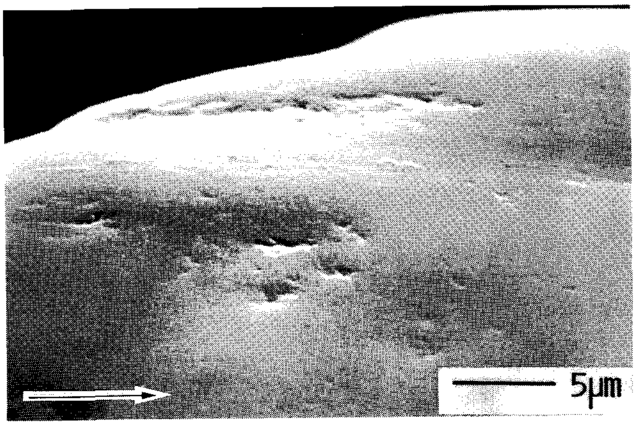

Fig. 3. Striations on « corn gloss ».

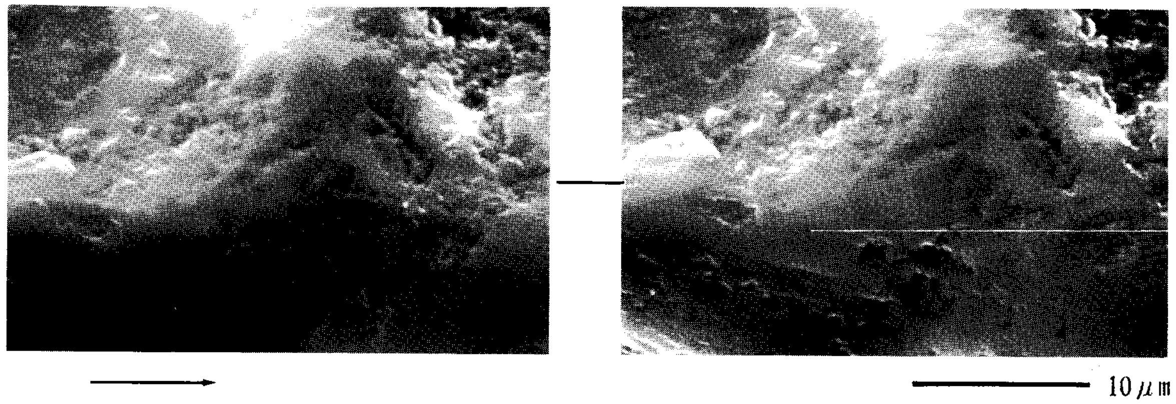

Fig. 4. Emergence of secondary « scratches ».

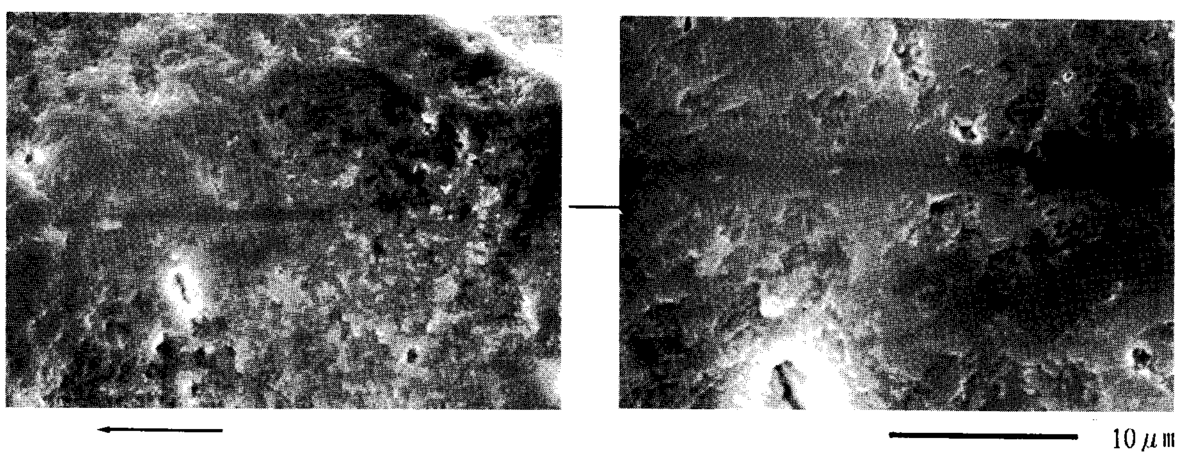

Fig. 5. « A striation ».

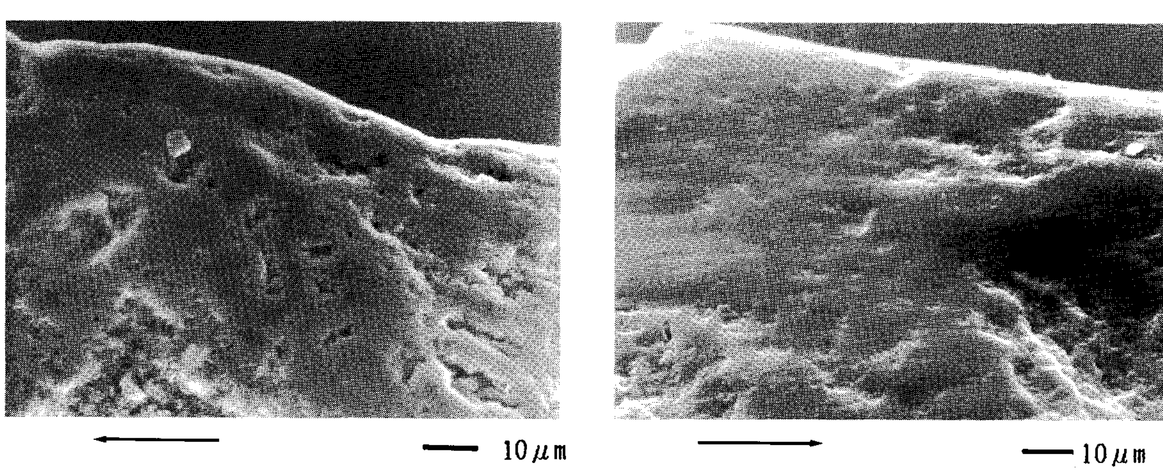

Fig. 6. « Plant polish » and « bone polish ».

that the distribution pattern of polished surfaces cannot be observed.

I do not mean that we can identify worked material types under SEM, only that « polish » variations observable under OLM are observable under SEM as different surface topographies. Whether these correspond to types of worked materials or not is another problem (see Yamada and Sawada in this volume).

One interesting fact revealed by this study found here is that when a « polish » is not very developed, and identification of diagnostic features is difficult with OLM, it is often possible to identify the diagnostics under the magnified image of SEM. A polished surface shows a basically similar pattern between 200-2500x. The fractal nature of polished surface, demonstrated by Rees *et al.* (1988), I think is the cause of the controversy as to whether the materials worked can be identified on undeveloped polished surfaces (Hurcombe, 1988 : 3 ; Bamforth, 1988 : 17 ; Newcomer *et al.*, 1988 : 25). Due to the fractal-like nature of polished surfaces, it is sometimes possible to judge worked materials from a small patch of « polish » and get a high score in blind test. The fact also suggests that a computer simulation of polished surface development is possible.

Whether a « polish » is attritional or additional cannot be determined simply by its appearance. Still, many of my photos show attritional appearance. At least, I cannot find thick layers up to about 10 μm of the sort Anderson reported in 1980. I think the thickly built-up appearance of « polish » is an illusion caused by the microtopography of the original surface as Meeks *et al.* (1982) indicated. Also, when a polished surface is domed, it gives an additional impression of depth.

Based on some reports of electron probe micro-analysis (Anderson, 1980 ; Mansur-Franchomme, 1983) and chemical cleaning which have been repeated since Keeley established the « polish » approach, a « polish » has been demonstrated to be a part of the rock surface rather than an additive to it. This is also admitted by the silica gel theory except for some transferential silica from plants. Micro-residues are secondary elements. Therefore, a « polish » should more properly be called a « polished surface ». The word « polish » is nothing but an optical impression. Since it does not refer to the material, the use of the term « polish » has no meaning for material scientists.

The progressive development of a polished surface

Having come this far, the next question we must ask is, what is the mechanism of the « polish » formation ?

To answer this let us trace the progressive alteration of the surface at a single point on a specimen using stereophotography of SEM. If the silica gel theory is right, distribution of micro-features such as striations and pits will greatly change as the polish develops. If the wear theory is right, such features will gradually be smoothed without changing their locations.

We have a technical problem to overcome first. Vapor deposition of metal is necessary for the SEM observation of unconductive materials such as rock, but how do we manage to remove the coating film after each observation ?

I confirmed that silver coating films are easily removed with H_2O_2, causing little damage to the rock surface. This was checked on one specimen, through 4 repeated treatments, with 230 photos. To check the stability of silver coating film, 3 specimens were examined 2 or 3 times each with 200 photos. Treatment with boiled H_2O_2, 10 wt %, 10 minutes was enough to clean the surface. It was followed by acetone wiping to avoid contamination due to the treatment residues. This technique can be widely applied in the basic research of polished surfaces and in the observation of archaeological specimens for which color change cannot be allowed.

In theory, the method of continuous observation seems easy, but actually it was quite difficult. First, the specimen has to be fixed at the same angle for each observation. Microchipping produced during use and unexpected accidents can also damage the edge and interrupt the observations. To improve the situation, a dorsal ridge was used instead of flake edge for specimens, and the « edge » was rubbed gently to avoid microflaking. Therefore the amount of use shown by strokes in the figures is not comparable to what might be expected in actual use.

Observation at 3500x was found to be most effective, although 1000x is effective to observe alterations over a wider area. At more than 3500x, it is difficult to take continuous photos at the same spot, and errors in the angle of setting a specimen greatly affect the interpretation of observed phenomena.

Results of the continuous observation are as follows. When we compare photos 2 and 3 in figure 7 with stereophotos, we find the domed area to the lower right of the two pits marked in photo 2 becomes the same elevation as the other area in 3. Were this alteration caused by deposition of some elements in the upper left area, the two tiny pits could not have survived. Wear at the lower right area is reasonable. We cannot determine whether the change is additional or attritional from the very minute change found in these photos because the specimen cannot be fixed in precisely the same angle at each time of the observation. However, when we compare no 1 with 4, wear is obvious. Wear is also confirmed in figure 8 : 1 and 2.

In these development processes of polished surfaces, two types of wear are confirmed. One is a cutting type which accompanies striation. The other is a fracture type which accompanies pitting. In figures 7 and 9, new pits are found to be produced. Strictly speaking, we cannot rule out the possibility that these pits were produced by H_2O_2 treatment. However, OLM observations show the process occurred during use. Under OLM, it has been demonstrated that dry hide working causes many pits and a large amount of wear. This may be related to the nature of dry hide.

I observed 10 specimens (5 specimens for grass, 3 for soaked antler, 2 for dry hide, 440 photos in all). The success of these observation depends on how the spot alters and if there are recognizable features at the spot. Furthermore, the interval of the observations has to be appropriate, neither too long nor too short. For these reasons, wear was not confirmed in all the photos. Still, at the very least, there was no evidence of thick surface layers being formed nor of surface flow occurring. However, we have to note that, with this method, it is impossible to interpret the phenomena occurring on the order of less than 1 µm.

Some problems with the silica gel theory

The apparent absence of striation on « corn gloss » is one of the important pieces of evidence against the « wear theory », but I can say that striations are present on polished surfaces. This apparent absence is due to inadequate observation, or because the striations are too fine to observe.

In the silica gel theory, water is assumed to become contained in the « polish » by chemical reaction. Another possibility is however that water acts as a lubricant. Thus, in abrasive wear, as defined in tribology, a lubricant brings the reverse lubricant effect and accelerates wear. We know, for example, that a kitchen knife can be sharpened most effectively with a whetstone and water.

Mansur-Franchomme's hypothesis on striation formation is also not supported by my observations. If striations on the surface disappear due to the surface flow, there must be a deep fluid layer compatible with the depth of striations which can be up to a few µm. However, such deep layers simply could not be detected on the examined surfaces. If such a flow does occur, the features said to be representative of worked materials disappear. The silica gel theory cannot explain why there are different « polish » types corresponding to worked materials. The existence of micro-residues is proposed as one of the agents, but these residues are too small in size and density to greatly affect the OLM image. This by itself is a contradiction that practitioners of the high power approach cite to support the silica gel theory.

It is possible that residues may adhere to the surface, but cannot sink into it because there is no layer to make this possible. Anderson-Gerfaud herself proved this in her paper of 1986, making reference to Andersen and Whitlow (1983)'s ion beam analysis. There the amorphous layer was found to be less than 1 µm. Also she detected transferential silicon by EDAX on bone implements used on plants, and cite this as an indirect evidence of the existence of transferential silica on stone implements. However, silicon was most heavily detected in depressions, and « the silicious residue material is much sparser and thinner, or none at all was detected, in smooth, flattened areas of the gloss surface of the tools » (Anderson-Gerfaud, 1986 : 74). This means that amorphous silica on the surface does not contribute to the smooth appearance. There must be a gel layer which is thicker than a few µm to catch plant opals and to flow out striations. Again, the silica gel theory fails.

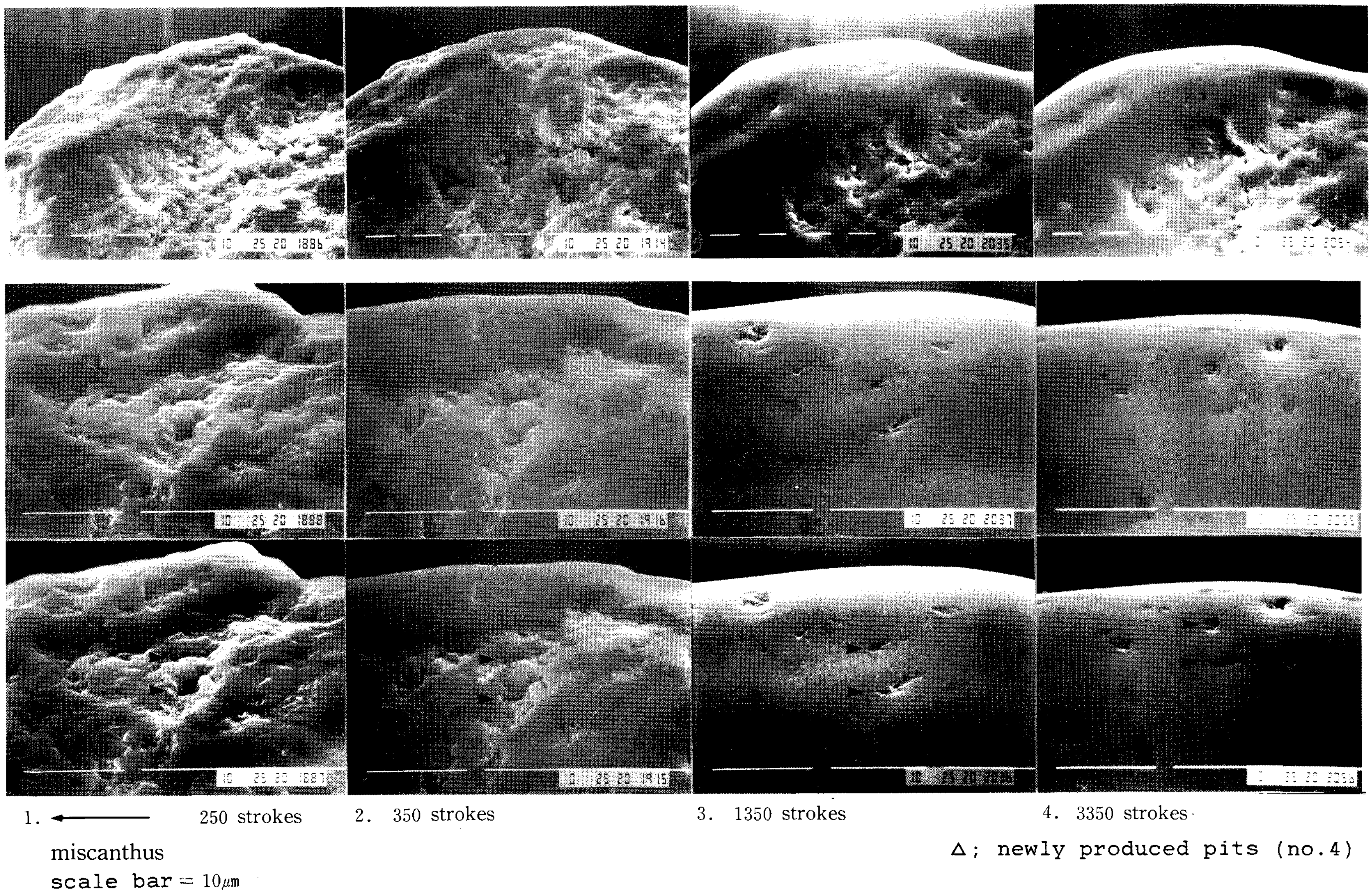

Fig. 7. Progressive development of polished surface. Lower series are stereo-photographies.

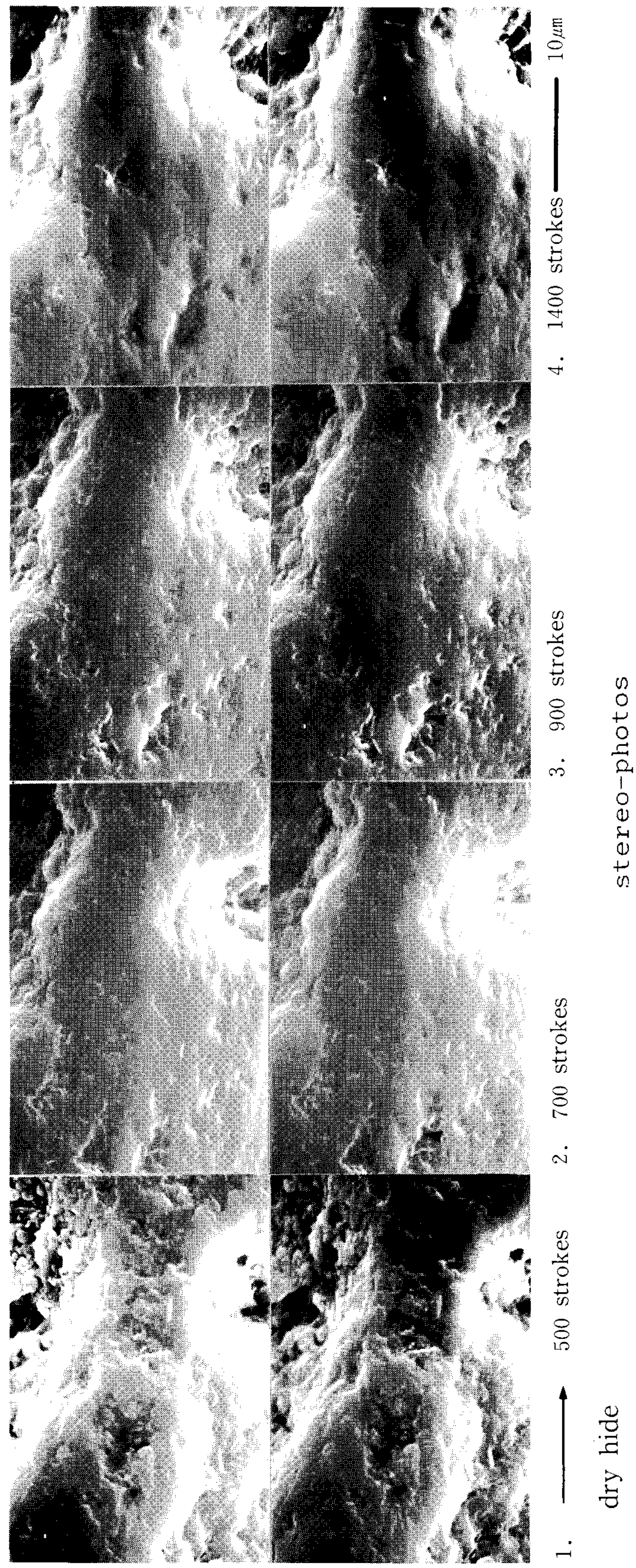

Fig. 8. Progressive development of a polished surface.

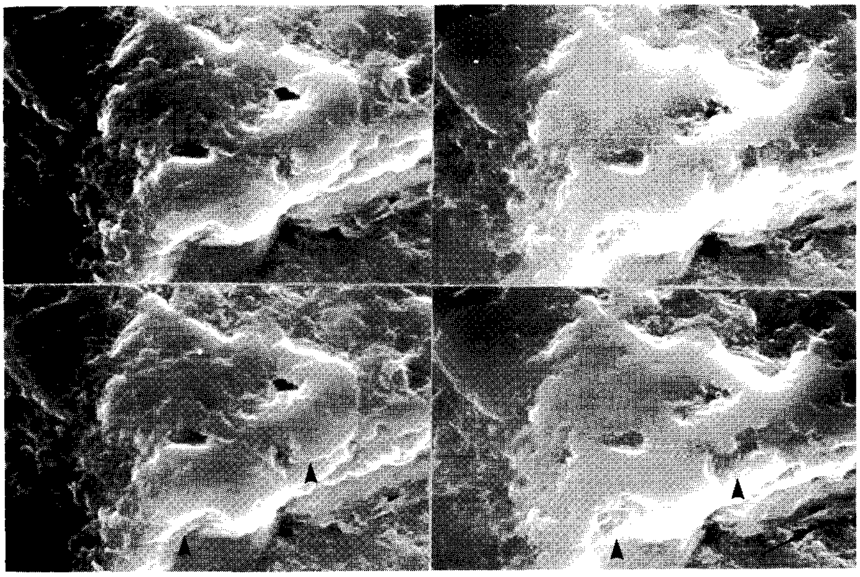

Fig. 9. Development of « fracture type » wear.

Discussion

From all the above results, it is evident that polished surfaces are produced by wear in the strict sense of the term. The mechanisms for the formation of smooth, mirror-like surfaces on the order of less than 1 µm, however, still remain unsolved. For the polishing process in tribology, there are multiple theories – abrasion in micro-scale, plastic flow, fusion, chemical reaction – to explain the phenomenon.

The features of polished surfaces of stone tools show those of abrasion, but the eminent fact that hard rock surfaces are abraded by relatively soft materials may support the possibility of chemical reaction as one of the possible mechanisms of glass polishing which Kamminga (1979) proposed for « use-wear polish ». That is, a hydrated, soft layer is formed on the surface, then removed by friction. However, Levi-Sala (in this volume) denies the possibility of hydration.

Relatively soft, highly viscous and elastic materials such as plants and hide tend to produce domed polished surfaces. Materials which have the opposite nature, bone and antler, tend to produce flat polished surfaces (fig. 10). Such features are basically common on various rock types (Midoshima, 1988). These facts indicate that diagnostic features of polished surfaces are mainly determined by 3 factors of the worked materials: its hardness, its viscous and elastic properties, and its surface texture. Grace (1989 : 60) predicted that all the polished surfaces finally become the same smooth, flat surface. However, there are qualitative differences in polished surfaces such as profile and frequency of pits.

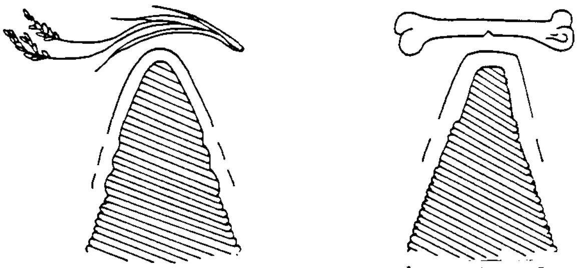

Fig. 10. Idealized profiles of a worn edge.

Conclusions

1. Diagnostic features of « use-wear polish » described with OLM are also recognizable with SEM. Profiles of polished surfaces are especially distinctive. « Bone and antler polish » tends to be flat, while « plant polish » tends to be domed. Identification of « polish » type is more difficult with SEM than with OLM because of the nature of the image.

2. The additive appearance of « polish » is an illusion caused by the nature of OLM as well as by microtopography and domed polished surfaces.

3. Polished surfaces show attritional appearances which actually consist of a number of striations.

4. The process that polishes surfaces has proved to be wear. Two types of wear were observed : a cutting type accompanied by striations and a fracture type accompanied by pits. Neither deposition of silica gel layer nor surface flow were observed with SEM.

5. The effect of moisture on « polish » development may be due to changes of the viscous and elastic properties of the worked materials to, a lubricant effect, to the acceleration of hydration, or to a combination of these three causes.

6. Diagnostic features of « polish » types are mainly produced by the hardness, the viscous and elastic properties, and the surface texture of the worked materials.

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