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Scientific approaches for adhesives in the conservation of Japanese paintings

Characteristics of Japanese paintings

There are three characteristics to note about Japanese paintings. First, they are composed of flexible and delicate materials like silk, paper and animal glue. These materials tend to suffer physical damage if they become wet or if they are even slightly stressed by the application of light pressure. Second, the pigments in Japanese paintings are not covered with coating materials; rather, they are simply applied using animal glue only. Thus, they tend to chalk as the glue deteriorates. Third, the paintings do not have a rigid form. The majority of Japanese paintings are mounted as scrolls, panels or books. In most of these cases, their forms differ when they are on display from when they are stored; for example, scrolls can be rolled and unrolled, and folding screens can be opened and closed.

Therefore, the conservation of Japanese paintings involves sophisticated skills with regard to the selection and application of conservation materials and treatments. These skills are aimed not only at conserving the fragile constituent materials, but also at handling the paintings themselves. To meet these opposing demands, Japanese conservators have become more skilful as they continue to develop a complex conservation process. Nonetheless, they do not have a sufficiently scientific point of view.

In this paper, the findings of my research regarding certain conservation materials will be presented in terms of three scientific approaches that can be applied to the analysis and development of conservation materials as described below. However, as I am also interested in the conservators' selection and application of materials, I hope that my scientific findings may be incorporated into the conservators' selection and application of conservation materials.

Scientific approaches

Scientific approaches for conservation materials in Japan can be grouped into three categories, with the first being the characterization of materials. In Japan, the chemical compositions of some materials used in conservation have not yet been clarified. For example, *furunori*, aged paste that is used only in lining mounting scrolls, is a very specific material. It is prepared in each conservation studio by fermenting wheat-starch paste for approximately 10 years, but no definition of *furunori* composition has yet been established. Another example of an undefined material used in Japanese conservation is *mamenori*. It is said that this is paste made from soybeans, but its preparation method has not yet been identified. At present, it is only known that this paste was used before the thirteenth century for paper joining. Research is ongoing.[1]

The second approach to which scientific techniques are applied is the improvement of materials after clarifying their components. For example, *funori* is a popular adhesive in Japan that is made from a kind of red agar, which is also used as a foodstuff and a cleaning agent. In conservation studios, *funori* is prepared by cooking sheets of *funori* agar in water. Then it is used as an adhesive for facing and as a thickener. Its general chemical components are evident, but they are altered by extraction temperature.

1 Noriko Hayakawa, 'Analysis of an Ancient Adhesive, the So-called "*Mamenori*" (Unfermented Soybean Paste)' [in Japanese], *Hozon Kagaku* 53 (2014): 81–94.

Therefore, we have proposed *funori* preparation techniques, research of which will be discussed later, that can be adjusted to suit required purposes.[2]

Another potential for improvement of materials involves wheat starch. In Japanese conservation, two kinds of wheat-starch paste are used: one is aged paste, as mentioned previously, while the other is fresh starch paste. The latter is sticky and very adhesive, so it is used when a strong adhesive is required. In cases when a medium-strength adhesive is required, conservators mix fresh and aged paste. However, this solute mixture often coheres, resulting in precipitation, and the mixture cannot be used properly. I assume that this occurs because aged paste is acidic. Therefore, we are attempting to create paste that is unaffected by this problem, but has the characteristics of partially aged paste. This attempt involves maintaining fresh starch paste at a low temperature in order for the starch to retrograde.

The third approach involves the pre-testing of new materials. Recently, attempts have been made to use new materials in Japanese conservation. Before any new materials are applied, however, it is necessary to examine and evaluate them. Therefore, some aspects of our experiments concerning the evaluation of new materials in order to facilitate suitable selection are also presented in this paper.

2 Noriko Hayakawa, Tominori Araki, Satoshi Kainuma, Tokuichi Taguro and Wataru Kawanobe, 'Characterization of *funori*—extraction from the red seaweed—as a restoration material' [in Japanese], *Bunkazai Hozon-shufuku Gakkai shi: kobunkazai no kagaku* 48 (2004): 16–32.

Characterization of materials: *furunori*

As an example of the first approach, material's characterization, the focus will be placed on aged paste, *furunori*.[3] As has already been said, there are two types of wheat-starch paste used in the conservation of Japanese paintings: fresh and aged. The latter, *furunori,* is prepared by storing fresh wheat-starch paste for approximately 10 years, and it is used only to paste lining paper for scrolls. *Furunori* has weak adhesiveness, so conservators use a beating brush method, known as *uchibake*, to support the adhesive strength when they apply *furunori*.

Furunori is also a special material from a cultural point of view. It is traditionally prepared by the studios' novice restorers. Starch paste becomes *furunori* approximately 10 years later, which is equivalent to the period of time required for apprentice conservators to finish their training. A share of the *furunori* that they themselves prepared when they were novices is presented to them as they become independent restorers.

The characteristics of both types of wheat-starch paste (fresh and aged) are shown in Table 1. *Furunori* has high flexibility, low tension and high repeelability after drying. These advantages are the reasons it is used only for scrolls. Repeated rolling and unrolling of scrolls causes tension and pressure load on the paintings. It is said that these loads are reduced through the use of *furunori*. However, *furunori* is acidic and its components are unclear, facts that are disadvantages as far as conservation is concerned. Thus, we have approached *furunori* from a chemical perspective.

3 Noriko Hayakawa, Rike Kigawa, Tomoyuki Nishimoto, Kurara Sakamoto, Shigeharu Fukuda, Takayuki Kirnishima, Yasuhiro Oka and Wataru Kawanobe, 'Characterization of *furunori* (aged paste) and preparation of a saccharide similar to *furunori*', *Studies in Conservation* 52, no. 3 (2007): 221–32.

Table 1 Characteristics of fresh paste and aged paste (*furunori*).

	Aged Paste (*furunori*)	**Wheat starch paste**
Colour	white or pale coloured	white
Acidity	pH 4-5	pH 5-6
Viscosity	low	high
Chemical component	unclear	clear
Flexibility	high	low
Tension	low	high
Repeelability	high	middle

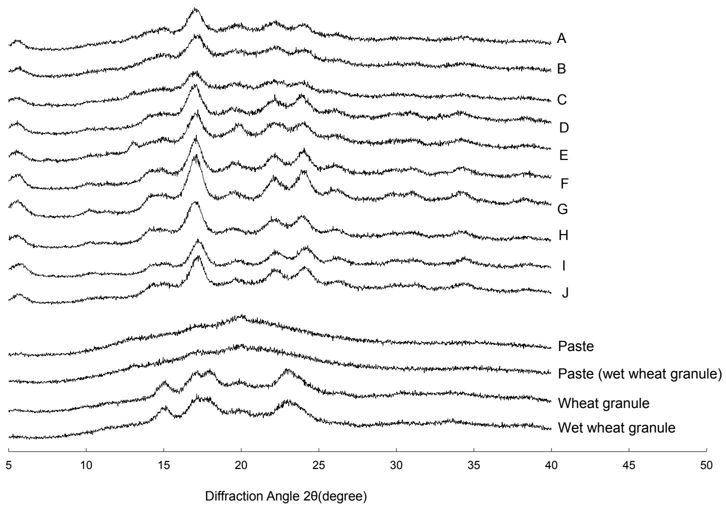

Fig. 1 X-ray diffraction patterns of *furunori,* fresh paste and starch granules.

Figure 1 shows the X-ray diffraction patterns of *furunori*. The bottom two samples are raw starch, and they exhibit patterns corresponding to A-type starch, which means that the raw material has crystallinity. When raw starch is heated in water, the peaks disappear ('paste' samples), and this means that the molecules are in an amorphous state. The alphabetized patterns are all *furunori* samples that were prepared in different studios under various conditions. When starch retrogrades, the molecules of the starch exhibit re-crystallinity. All analysed *furunori* samples exhibited a re-crystallized pattern, that is, the peaks reappeared. This means that *furunori* has crystallinity; thus, *furunori* is well retrograded, a feature which is one of its central characteristics.

Gel filtration chromatograms (GFC) of *furunori*, which indicate the molecular weight, are shown in Figure 2. The black line represents the molecular weight of fresh starch paste, which is the raw material, while the coloured lines are those of *furunori*. These samples were again prepared in different studios under various conditions. However, all the coloured lines show lower molecular weight than that of fresh starch paste. This means that the main component of *furunori* has decomposed. This is the second significant characteristic of *furunori*.

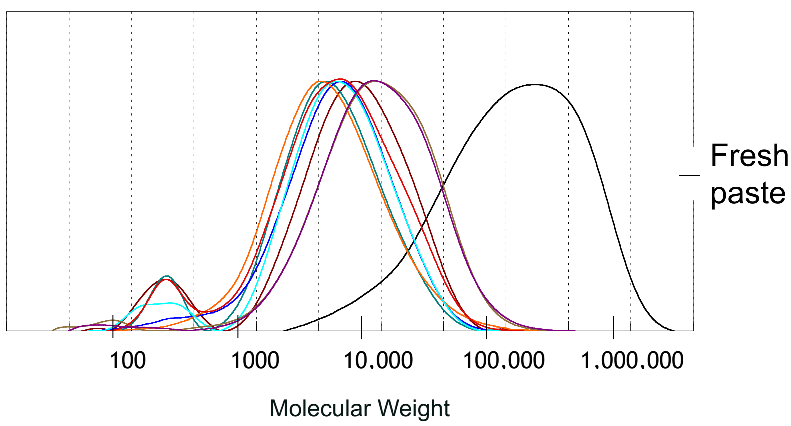

Fig. 2 Gel filtration chromatograms of *furunori* and fresh starch.

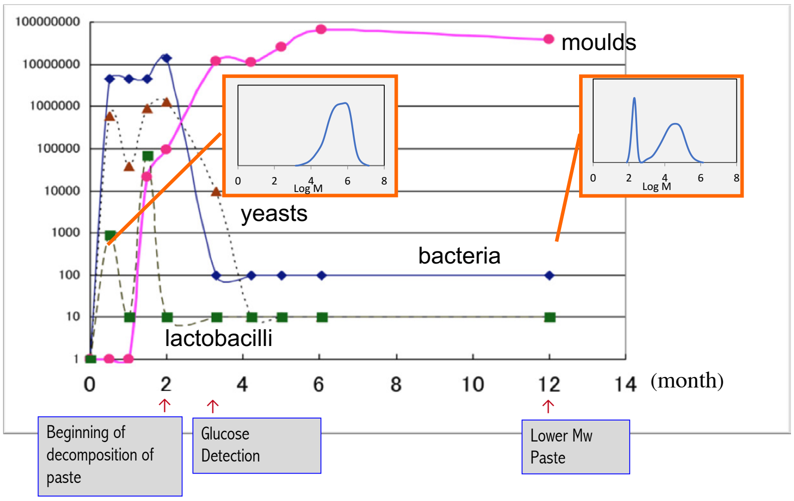

Fig. 3 Relation of micro-organisms for the generation of *furunori.*

The next attempt was to clarify the source of this decrease in molecular weight. Fresh starch paste was prepared in winter, which is the best time for retrograding paste because the phenomenon of retrogradation tends to be shown in low temperatures. We then sampled our specimens at regular intervals and monitored changes in the micro-organism count and accompanying changes in the molecular weight. We monitored the samples for bacteria, yeast and mould. Two months later, we detected mould (Fig. 3). At that time, GFC analyses indicated that the main part of the paste was exhibiting the initial stages of decomposition. Then, four months later, the mould became dominant and, at that time, we detected a fraction in the GFC results that was assumed to be glucose. The various types of mould remained dominant until the end of the year, when the main fraction in the GFC began to shift to a lower molecular weight and the glucose fraction became larger. Hence, it has been suggested that mould growth contributes significantly to *furunori* preparation, as several types of mould generate alpha-amylase. Starch molecules are decomposed by alpha-amylase into glucose units and lower molecular-weight polysaccharides.

Based on the above findings, the characteristics of *furunori* can be summarized as follows. The first feature is the retrograded starch that develops when the paste is stored for a long period of time. The second characteristic is the lower molecular-weight polysaccharides, the main component of *furunori*, which are generated by starch decomposition caused by enzymes in the developed mould. The third feature is the acidity, as indicated by the presence of organic acids that are the degradation products of starch decomposition. The fourth characteristic is the disappearance of intermediates that were detected after one year of storage during our study. These substances were not detected in the *furunori* chromatograms, so it is assumed that they were consumed by various micro-organisms during the 10-year storage period. When *furunori* is prepared for the first time in a studio or outside Japan, care must be taken to retain the first two characteristics. If micro-organisms decompose the starch before it retrogrades sufficiently to re-crystallize, all of the starch will be utilized by micro-organisms. Therefore, it is suggested that the starch first be maintained at a low temperature for a certain period of time, that is, until it has retrograded sufficiently. Thus, the starch can retrograde in preparation for *furunori* production, because some of the crystalline component of the retrograded starch will not be decomposed by the alpha-amylase, which is generated by the developed mould.

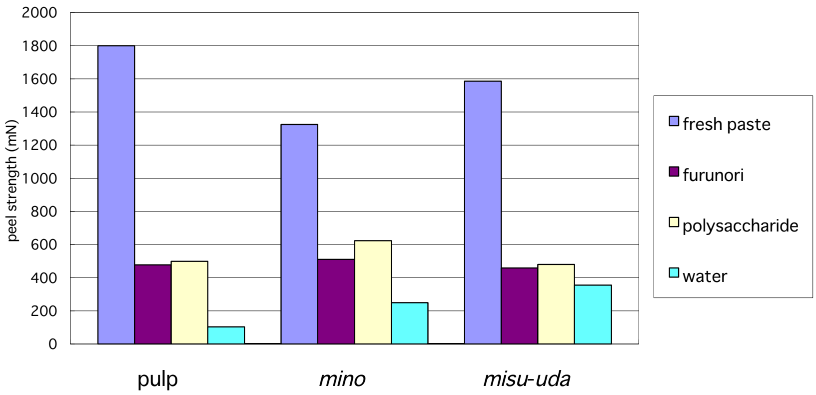

Fig. 4 Peel strength of samples pasted using the *uchibake* method.

Based on these findings, an attempt was made to prepare the polysaccharide similar to *furunori*. Fresh starch paste was prepared by maintaining a temperature of 5º C for retrograding. Then, alpha-amylase was added in order to decompose the starch. The sample was washed sufficiently to remove the intermediate products, instead of waiting for micro-organisms such as oligosaccharides to utilize these components (as in the case of *furunori*). Subsequent analyses indicated that the adhesive strength of this polysaccharide is similar to that of *furunori*, even when applied to different paper combinations (Fig. 4).

As has already been mentioned, we also considered the combination of materials and technique. When conservators apply *furunori*, they use the beating brush method called *uchibake*, a term which refers to the beating of the reverse side of the lining paper with a brush so as to enhance the adhesive strength of *furunori*. Three kinds of paper combinations were prepared for testing: Western paper with Western paper (wood-pulp paper), *mino* paper with *mino* paper (Japanese *kōzo* paper), and *misu* paper with *uda* paper, which are types of Japanese paper with natural clay as filler. Three kinds of adhesives were applied to the paper combinations: fresh paste, aged paste and water. The resultant peel strength was then measured. Fresh paste was found to have the strongest adhesiveness, followed by *furunori* (Fig. 5). It should be noted that all of the samples to which the *uchibake* method was

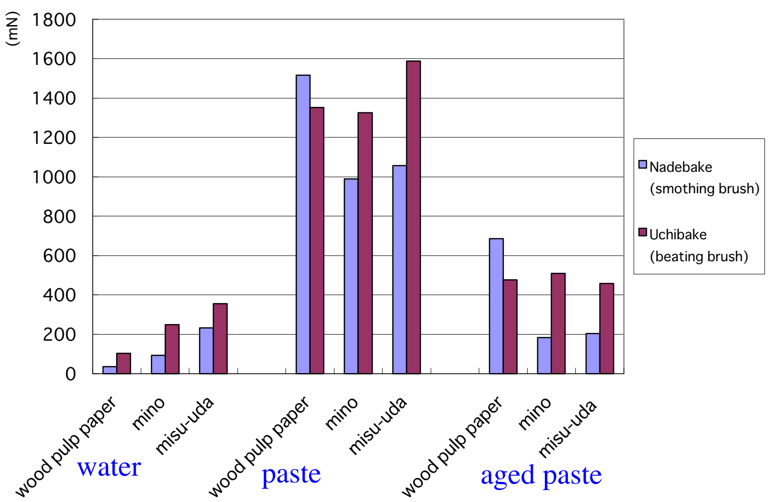

Fig. 5 Adhesive effect of *uchibake* (beating brush)

Fig. 6 (a) Red agar (*funori*). (b) Bleached using sunlight. (c) *Funori* sheet.

applied exhibited higher adhesiveness than those without this treatment, except in the case of wood-pulp paper. This means that the combination of three elements, that is, *uchibake*, *furunori* and long fibre paper (such as Japanese paper) is effective. So, this demonstrates an accurate, sophisticated combination of both material and technique.

Improvement of materials: *funori*

The second approach, materials' improvement, is discussed in this section, using *funori* as an example.[4] *Funori* is a kind of red agar harvested in the tidal zone along the coast of Japan. After harvesting, it is bleached by agents and sunlight, and then dried to make *funori* sheets (Fig. 6). Japanese conservators have found that *funori* paste has two characteristics: first, it is easy to remove by the application of water, even after drying; and second, it has high viscosity. So, in Japan, *funori* is used for three purposes: for facing, as a thickener and for consolidation. I have heard that *funori* is used as a consolidant by itself in Europe, but in Japan *funori* is rarely used alone in this manner; rather, it is mixed with glue or starch because the adhesive power of *funori* by itself is not sufficient for consolidation.

There are three species of *funori* agar: *ma-funori* (*Gloiopeltis tenax*), *fukuro-funori* (*Gloiopeltis furcate*), and *hana-funori* (*Gloiopeltis complanata*). Recently, *ma-funori* and *fukuro-funori* have become major components of *funori* sheets. The chemical structures of the extracts of each of these components have already been clarified to a great extent,[5] but the analysed samples have only been extracted at high temperature, apart from one sample being considered in a current study.[6] Japanese conservators, however, have noted that the *funori* extract (*funori* paste) conditions vary with extraction temperature. Therefore, we have conducted research into *funori* extract, considering both

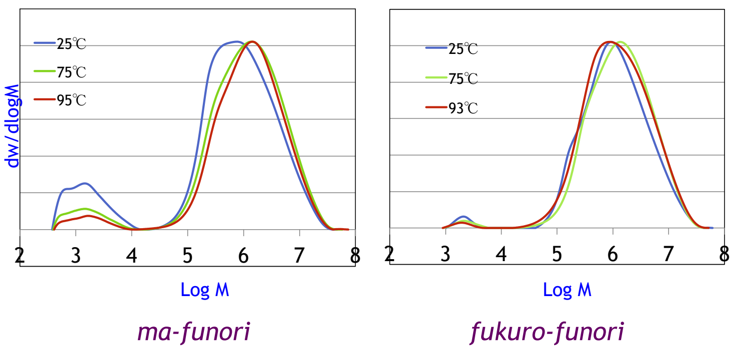

Fig. 7 Comparison of the molecular weight of *funori* at different extraction temperatures.

4 Hayakawa et al., 'Characterization of funori—extraction from the red seaweed'; Noriko Hayakawa, Keiko Kida, Takuya Ohmura, Noriko Yamamoto, Kyoko Kusunoki and Wataru Kawanobe, 'Characterization of funori as a conservation material: Influence of seaweed species and extraction temperature', *Studies in Conservation* 59, Supplement 1 (2014): 230–31.

5 Ryo Takano, Kaeko Hayashi, Saburo Hara and Susumu Hirase, 'Funoran from the red seaweed, *Gloiopeltis complanata:* polysaccharides with sulphated agarose structure and their precursor structure', *Carbohydrate Polymers* 27, no. 4 (1995): 305–11; Ryo Takano, Hiroko Iwane-Sakata, Kaeko Hayashi, Saburo Hara, Susumu Hirase, 'Concurrence of agaroid and carrageenan chains in funoran from the red seaweed *Gloiopeltis furcata* Post. et Ruprecht (Cryptonemiales, Rhodophyta)', *Carbohydrate Polymers* 35, nos. 1–2 (1998): 81–87; Karin Catenazzi, 'Evaluation of the use of *Funori* for consolidation of powdering paint layers in wall paintings', *Studies in Conservation* 62, no. 2 (2017): 96–103.

6 Hayakawa et al., 'Characterization of *funori* as a conservation material', 230–31.

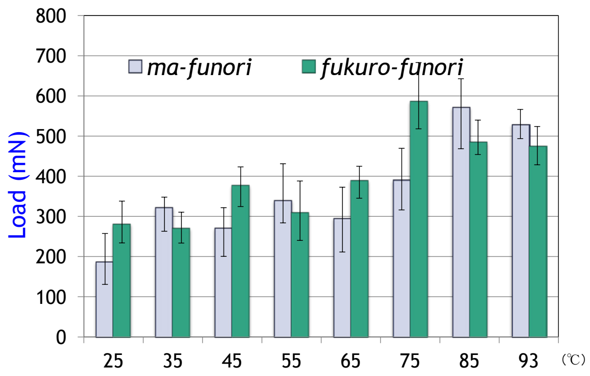

Fig. 8 The relationship between adhesive strength and extraction temperature.

the species of *funori* and the extraction temperature. Figure 7 shows GFC chromatograms for *ma-funori* and *fukuro-funori* samples, which demonstrate that the main components in the extracted samples of both species shifted to higher molecular weight with higher extraction temperature. Figure 8 shows the relationship between adhesive strength and extraction temperature. It is apparent that the strength also changed with the extraction temperature. The highest value, which did not occur at the highest temperature, is indicated. In addition, for *fukuro-funori*, the difference with respect to temperature was larger than in the case of *ma-funori*. In Japan, the ratio of *ma-funori* and *fukuro-funori* contained in a given *funori* sheet depends on the district in which the agar is harvested. As the extraction characteristics of these two species differs, it is now possible to select *funori* for specific purposes based on desired extraction conditions, whether for use as a thickener or for facing.

Pre-testing: application of new materials

The pre-testing approach is a very popular method in conservation science. In Japan, there are few mural paintings on plaster in tumuli, and only two such paintings have been discovered and restored. In these cases, before restoration began, it was necessary to determine the appropriate adhesives, considering both natural and industrial materials.[7] As it would be difficult to discuss all of the relevant evaluations we conducted here, selected results

7 Noriko Hayakawa, Eriko Nakau, Rike Kigawa, Akiko Okimoto and Wataru Kawanobe, 'Basic research of conservation materials for painting surface' [in Japanese], *Bunkazai Hozon-shufuku Gakkai shi: kobunkazai no kagaku* 53 (2008): 1–19.

Table 2 List of materials used in penetration tests.

			Solvent	Supplier
(a)	*funori*	harvested in Wakayama prefecture	water	-
(b)	cow glue (pellet type)	No.1 (JIS)	water	Sun-Orient Chemical
(c)	cow glue (*sanzenbon*)	produced in Himeji prefecture	water	
(d)	paraloid B72	acrylic resin	xylene	Rohm & Haas
(e)	paraloid B72	acrylic resin	ethyl acetate	Rohm & Haas
(f)	AC3444	acrylic emulsion	o/w emulsion	Rohm & Haas
(g)	hydroxypropyl cellulose	Mw : 10×10^4	ethanol	Aldrich
(h)	hydroxypropyl cellulose	Mw : 10×10^4	water	Aldrich
(i)	hydroxypropyl cellulose	Mw : 100×10^4	ethanol	Aldrich
(j)	hydroxypropyl cellulose	Mw : 100×10^4	water	Aldrich
(k)	methyl cellulose	400cps at 2wt%	water	Aldrich
(l)	methyl cellulose	4000cps at 2wt%	water	Aldrich
(m)	ethyl cellulose	4cps at 5wt% (80/20=toluene/ethanol)	ethyl acetate	Aldrich

from this research will be presented.

Evaluation tests were conducted to determine the compressive strength, penetration strength, colour change and susceptibility against mould of the examined adhesives. The penetration tests will be discussed here. The adhesives in question were: *funori*, methyl cellulose (MC), hydroxypropyl cellulose (HPC), cow-hide glue made in Japan, paraloid B72 and AC3444, which is an acrylic emulsion (Table 2). Five grams of each adhesive was dropped on a foundation of $CaCO_3$. The load and depth were measured when a 0.70 mm ϕ needle penetrated the samples. Figure 9 shows the results at 50% relative humidity (RH); the vertical and horizontal axes indicate depth and load, respectively. This result shows that cow-hide glue and AC3444 penetrated deeply and with strong consolidation. On the other hand, paraloid B72 penetrated deeply but exhibited weak consolidation. *Funori* penetration was both shallow and weak. Figure 10 shows the results of a penetration test conducted at 100% RH: all adhesives exhibited weaker consolidation, but even under these conditions paraloid B72 maintained good performance. *Funori*, HPC and EC exhibited a low load value, demonstrating that *funori* is not a perfect material for use in conservation. At present, restoration works for mural paintings are being conducted using several materials that were selected based on the results of this series of pre-tests.

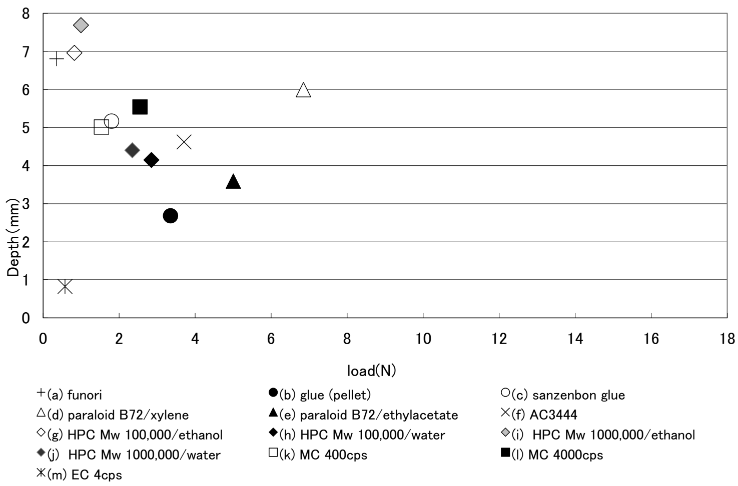

Fig. 9 Penetration test for adhesives at 50% RH.

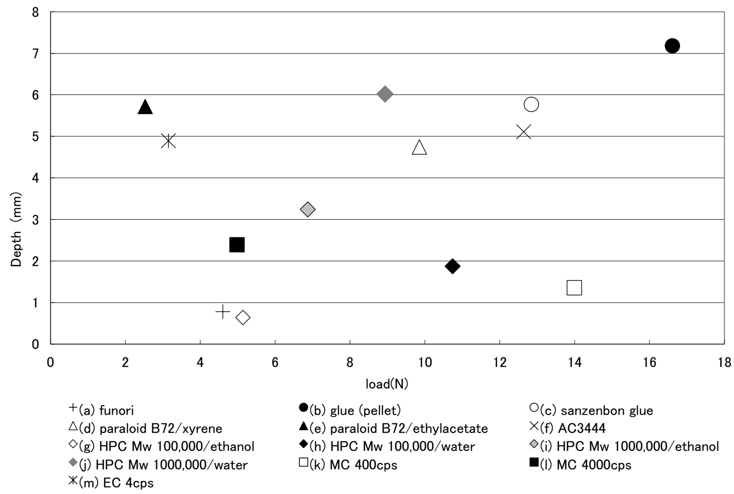

Fig. 10 Penetration test for adhesives at 10% RH.

Conclusion

The research presented in this study focused on Japanese adhesives and their use in the conservation of Japanese paintings. Three investigative approaches were applied: characterization, improvement and pre-testing. These approaches are not independent of each other but are, in fact, related. For example, *funori* must be characterized in order to improve its properties. Further, characterization of *furunori* has aided in attempts to prepare a polysaccharide with similar properties. In addition, this characterization has assisted in the development of a partially aged paste that is retrograded only; this paste was briefly mentioned above. Pre-testing has facilitated more scientific selection of materials for application in current conservation works. In the future, I hope that both conservators' perspectives, which are based on their experiential knowledge, and scientific approaches, such as those introduced in this paper, will be closely incorporated in conservation projects.

Abstract

Scientific approaches to the analysis and development of conservation materials in Japan can be classified in three categories. The first involves the characterization of conservation materials, leading to the confirmation of the rationality of conservators' experience-based techniques. An example of such an approach is the examination of the application of aged wheat-starch paste, *furunori*, in conservation of Japanese paintings. The second approach to conservation materials involves the clarification and improvement of the conservation materials. Such materials include, for example, *funori*, a popular adhesive in Japan made from a kind of red agar, which is also used in conservation. The third approach involves the pre-testing and examination of new materials for use in conservation, particularly for specific cases. In this paper, several current studies pertaining to Japanese adhesive agents for use in conservation are presented, and the combination of scientific research with conservators' experiential knowledge is discussed.

Biography

Noriko Hayakawa studied polymer science at the Tokyo Institute of Technology, obtaining her MEng in 1998. In 1998 she began work as a researcher in the Materials Science Section, Restoration Technique Department of the National Research Institute for Cultural Property, Tokyo. Since 2011 she has been senior researcher. Her principal interests are the characterization and application of restoration materials for Japanese paintings and traditional organic objects.

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