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Identification of Ryukyu lacquerwares by pyrolysis-gas chromatography/mass spectrometry and $^{87}Sr/^{86}Sr$ isotope ratio

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ABSTRACT

Six pieces of lacquer obtained from Ryukyu lacquerwares produced in the 17–19th century in the Ryukyu Kingdom were analyzed by pyrolysis gas chromatography/mass spectrometry and $^{87}Sr/^{86}Sr$ isotope ratio measurements to determine the identity of the lacquer source. The results were compared with the standard of natural lacquer film to discriminate lacquer species. Four pieces were made used lacquer sap collected from *Toxicodendron vernicifluum* lacquer tree, and other two pieces were made with lacquer sap tapped from *Toxicodendron succedanea* lacquer tree according to the characteristics of the pyrolysis products. All $^{87}Sr/^{86}Sr$ isotope values of urushiol lacquerwares are over 0.71, suggesting that the origin of lacquer trees were the Asian continent.

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1. Introduction

Lacquer is a kind of special natural resin tapped from lacquer trees belongs to *Toxicodendron vernicifluum*, *Toxicodendron succedanea*, and *Gluta usitata* family. Because it can form a beautiful and durable film after drying, lacquer has been used to prepare crafts or wares not only for protection but also for aesthetic effect in Asian countries for more than 7000 years [1,2], and lacquer culture is still a part of modern life in Asia [3].

In order to make a better conservation and restoration of the valuable ancient lacquerwares, identification of the lacquer origin and species is important. Although solid nuclear magnetic resonance (NMR) [4] and Fourier transform infrared spectroscopy (FT-IR) [5] have been employed, several troublesome pre-treatments are required and cannot obtain clear results. On the other hand, pyrolysis-gas chromatography/mass spectrometry (Py-GC/MS) is an effective method for analyzing lacquer film and other natural resins due to the different characteristic pyrolysis products [6–11]. An improved method with direct probe-Li^+ ion attachment mass spectrometry used to analyze Japanese lacquer film also was reported [12]. Previously, we analyzed six pieces [13] and two pieces [14] of Ryukyu lacquerwares belonging to the Urasoe Art Museum by Py-GC/MS. The characteristics of the pyrolysis products of urushiol (MW = 320), 3-heptylcatechol (MW = 208), 3-heptylphenol (MW = 192), laccol (MW = 348), 3-nonylcatechol (MW = 236), and 3-nonylphenol (MW = 220) were detected, suggesting that these Ryukyu lacquerwares were coated with alone and/or mixed with each other of lacquer sap tapped from *T. vernicifluum* and *T. succedanea* lacquer trees. In this study, three pieces from lacquer boxes, two pieces from lacquer trays, and one piece from lacquer cupboard of Ryukyu lacquerwares belonging to the Urasoe Art Museum were analyzed by Py-GC/MS and $^{87}Sr/^{86}Sr$ isotope ratio measurements. Based on the results, Ryukyu lacquer culture and trade history are discussed.

2. Experimental

The information of six pieces of lacquer that obtained from Ryukyu lacquerwares belonging to Urasoe Art Museum are summarized in Table 1, and the photos of objects are shown in Fig. 1.

2.1. Py-GC/MS measurement

Pyrolysis gas chromatography/mass spectrometry measurements were carried out using a vertical micro furnace-type pyrolyzer PY-2020iD (Frontier Lab, Japan), a Hewlett-Packard 6890 gas chromatograph, and a HPG 5972A (Hewlett-Packard, Ltd.) mass spectrometer. A stainless steel capillary column (0.25 mm

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Fig. 1. Photos of lacquerwares (the Nos. are the same as Table 1).

Table 1
Samples information.

No.	Art name (No. in Museum)	Size (L × W × H, cm)[a]	Age (century)
1	Green wood tray with *Hakue* (4)	36 × 36.4 × 4.1	16–17
2	Black wood box with *Raden* (31)	11.9 × 11.9 × 9	17–18
3	Black wood box with *Raden* (32)	10.3 × 10.3 × 11.3	17–18
4	Black wood lunch box with *Raden* (33)	13 × 14.3 × 18.8	18–19
5	Black/red wood cupboard with *Raden* (43)	46.5 × 77.6 × 131.5	17–18
6	Black wood tray with *Raden* (44)	15.2 × 15.2 × 4.5	17–18

[a] L: length; W: width; H: height.

i.d. × 30 m) coated with 0.25 μm of Ultra Alloy PY-1 (100% methylsilicone) was used for the separation. The sample (0.05 mg) was placed in a platinum sample cup. The cup was placed on top of the pyrolyzer at near ambient temperature. The sample cup was introduced into the furnace at 500 °C, and then the temperature program of the gas chromatograph oven was started. The gas chromatograph oven was programmed to provide a constant temperature increase of 12 °C per min from 40 °C to 320 °C, and then hold for 10 min at 320 °C. The flow rate of the helium gas was 1 ml/min. All pyrolysis products were identified by mass spectrometry. The mass spectrometry ionization energy was 70 eV (EI-mode).

2.2. $^{87}Sr/^{86}Sr$ isotope ratio measurement

The $^{87}Sr/^{86}Sr$ isotope ratio measurement is carried out as the same as our previous report [15] and the detail of method is described as follow.

2.2.1. (1) Sample treatment and separation of strontium

Thirty micrograms of the lacquer sample was treated with 6 ml of 14 M nitric acid at 120 °C for 11 h in a Teflon beaker with a lid to dissolve the organic matter, and the lid was opened in dry heat to remove the organic matter. Then 5 ml of 14 M nitric acid, 0.5 ml perchloric acid, and 0.5 ml hydrogen peroxide were added to the beaker and continuously heated at 120 °C for 9 h covered, and then the lid was opened in dry heat to remove the organic matter. This operation was repeated at least four times in order to completely remove the organic matter. Then 1 ml of 7 M nitric acid was added to the sample and shaken to dissolve it in nitric acid; this was called sample **I**.

Table 2
Pyrolysis products of Ryukyu lacquer films and their species.

No.	Lacquer species	Pyrolysis products			
		Monomer	Alkylcatechol	Alkylphenol	Drying oil
1	*T. succedanea*	Laccol	3-Nonylcatechol	3-Nonylphenol	Fatty acids
2	*T. vernicifluum*	Urushiol	3-Heptylcatechol	3-Heptylphenol	Fatty acids
3	*T. succedanea*	Laccol	3-Nonylcatechol	3-Nonylphenol	Fatty acids
4	*T. vernicifluum*	Urushiol	3-Heptylcatechol	3-Heptylphenol	Fatty acids
5	*T. vernicifluum*	Urushiol	3-Heptylcatechol	3-Heptylphenol	Fatty acids
6	*T. vernicifluum*	Urushiol	3-Heptylcatechol	3-Heptylphenol	Fatty acids

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