Distribution of Lead-barium Glasses in Ancient Japan

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Introduction

Lead-barium glass is considered to be unique to ancient China[1]. It is one of the earliest types of glass in Japan, and first appeared in the third century BCE, accompanied by advanced bronze-age culture and a small amount of iron tools. These facts indicate that lead-barium glass was also a marker for interaction and trade relationships between Japan, China, and Korea in the early Iron Age[2]. Further evidence suggests that the distribution of most of the lead-barium glasses stopped by the end of the second century CE. In Japan, tubular beads are the most common lead-barium glass artefacts, although other objects, including *bi* 璧 discs, eye-beads and comma-shaped beads, also exist. In this paper, we categorize lead-barium glass artefacts found in Japan based on form, manufacturing methods and chemical composition, and discuss each production area and distribution route.

Samples

The targeted objects are lead-barium glass beads buried in tombs, chosen in order to identify each period accurately. Glass beads are often excavated from dwelling remains, post holes or layers containing relics, but it is difficult to avoid contamination in cases that were subjected to an open burial process. Such erroneous samples are undesirable and make it difficult to clarify the transitions by period.

The investigated era covers the Yayoi period and the Kofun period. In the Jōmon period, which is just before the Yayoi period, glass artefacts did not exist. The Yayoi 弥生 period and Kofun 古墳 period are each divided into early, middle, late and ending periods. Further subdivision is possible. In order to date unearthed glass artefacts as precisely as possible, artefacts found in the same archaeological context, such as earthenware or bronze mirrors made in China,

1 Seligman 1937, 5-30; Seligman and Beck 1938, 1-64.
2 Oga and Tamura 2013, 35-65.

are used as an indicator. The starting age of the Yayoi period is not clear and still under discussion. The middle Yayoi period was between the third century BCE and the first century BCE, the late Yayoi period is from the first century to the first half of second century CE, and the ending Yayoi period is the second half of the second century. For the Kofun period, the early, middle, late and ending periods are, respectively, between the third and fourth century, the fifth century, between the sixth and first half of seventh century and between the second half of the seventh and beginning of the eighth century.

Lead silicate glasses are not preserved in good condition under the general burial environment in Japan. As a consequence, few chemical examinations have been conducted, namely on 56 glass beads unearthed from 18 different sites (Table 1). We can also refer to useful data published in earlier research. Regarding the changes of the glass beads distributed in ancient Japan, all objects were considered.

Table 1 Beads examined by analytical methods

Site	Prefecture	Age	Type													
			Tubular					Comma			Eye beads	Small				
			I	II		III	others	I	II	III		I	II			III
				Old	New								Old	New		
Hakuji No.1 tunnel tomb	Miyagi	AD7									1					
Tabata-nishidaidori site	Tokyo	AD2												1		
Karako-kagi site	Nara	BC1-AD1												3		
Higashiyama site	Hyougo	AD1				13										
Oshinooka site	Hyougo	AD1				6										
Dasaka No.3 kofun	Hyougo	AD2							1							
Nagasuna No.3 site	Tottori	unknowm			1											
Sakai-yaishi site	Tottori	unknowm			1											
Yokota No.1 site	Hiroshima	AD1				1										
Takagi site	Fukuoka	BC2										1	6			
Mitoma No.B8 kofun	Fukuoka	AD6-7														1
Hazezuka No.1 kofun	Fukuoka	AD6								1						
Nakabaru site	Saga	BC2-AD1	3									1				
Mutabe site	Saga	BC2										1				
Ikisa-nakabaru site	Saga	AD1	1													
Higashiyamada-ipponsugi site	Saga	AD1-2				2		1								
Otomo site	Saga	unknowm					1									
Fukiage site	Oita	BC1		9												

Analytical Methods

Microscopic Observation

The bead-making technique was estimated by observation through a stereo microscope. Clues showing bead-making techniques include the shapes of bubbles contained in the glass, the surface condition and the perforations, all of which can be observed with the stereo-microscope.

Measurement of Specific Gravity

As mentioned above, lead-barium glass artefacts are not preserved in good condition, and it is often difficult to analyse the precise chemical compositions of lead-barium glass artefacts by non-destructive methods. Measurement of specific gravity is helpful to estimate the concentrations of PbO in lead-barium glass artefacts excavated in Japan. In the present study, we measured the specific gravity of some well-preserved artefacts using the Archimedes' principle.

X-ray Fluorescence Analysis

In this research, an energy-dispersive X-ray fluorescence analyser (EDAX, EAGLE III) was used to conduct non-destructive chemical composition measurements. As the diameters of the investigated beads are small (less than 10 mm), micro-area analysis was required. In X-ray fluorescence analysis, both surface weathering and surface curvature can be the cause of measurement errors. The irradiation area of the device used in this research is 112 μm. This range is small enough compared to the size of the targeted objects. Therefore, we judge that the influence of the glass bead curvature is negligible.

The measurement results are normalized by Fundamental Parameter method (FP method) in such a way that the total amount of the oxides of elements detected will be 100%. The FP method was calibrated using glass standards similar to the samples measured, but whose compositions are known.

The target of the X-ray tube is molybdenum (Mo) and the X-ray tube voltage is set to 20 kV for quantitative analysis by the FP method, and to 50 kV for some objects in order to obtain spectra above 20 kV for qualitative analysis. The X-ray tube current is set to 100 μA, the X-ray irradiation diameter is set to 112 μm and a measuring time (live time) of 300 seconds is used. The measurements are conducted in vacuum.

Computed Radiography Method

In this research, Computed Radiography method (CR method) was used to complement the X-ray fluorescence analysis for material examination. In the CR method, an Imaging Plate (IP) is used for detection instead of a film. The filming principle is identical with film, but the IP has a high sensitivity and wide dynamic range, and it is possible to obtain pictures as digital data.

The CR method is used to distinguish alkali silicate glasses and lead silicate glasses. When the densities of alkali silicate glasses and lead silicate glasses are compared, the specific gravity of the latter is much greater and the X-ray ab-

sorption coefficient of the latter is also larger. Therefore, when the X-ray transmission photographing is done on alkali silicate glasses and lead silicate glasses under the same conditions, the lead silicate glasses look darker in a positive image, helping to distinguish the two[3].

A micro-focus X-ray Magnification Imaging System (Fujifilm, μFX-1000) and Imaging Analyser (Fujifilm, BAS-5000) were used for the CR method. The IP used was a BAS-SR2025. The X-ray tube voltage, the X-ray tube current and the exposure time were varied depending on the measured samples, but the typical operation ranges were 30 to 50 kV for the voltage, 40 to 60 μA for the current 60 to 180 seconds for the exposure time.

Results and Discussions

Form and Production Techniques

Several kinds of beads, such as tubular beads and comma-shaped beads, are predominant in lead-barium glasses in Japan, but *bi* discs, hairpins, cullet and others also exist. Concerning the glass beads, they are divided into several groups, based on the production techniques and the distribution periods. All the artefacts investigated in this study are beads, including tubular beads, comma shaped beads, eye beads and small beads. First of all, we explain the subgroups based on the production techniques and the distribution periods.

Tubular beads are the most common lead-barium glass artefacts in Japan. Therefore, in this study, we mainly discuss the tubular beads. Lead-barium tubular beads are divided into three types: Tubular I (Fig. 1), Tubular II (Fig. 2, 3) and Tubular III (Fig. 4), based on the manufacturing methods and shapes.[4] A few tubular beads, which do not belong to Tubular I, II or III, are called Tubular others (Fig. 5). Tubular I contains large misshapen beads made by the winding method. They have translucent or opaque blue appearances coloured by copper. They appeared at first around the third century BCE and were distributed only in northern Kyūshū until the second century BCE. The number of Tubular I is small in Japan, but the same kind of tubular beads have also been excavated in Jilin province in China[5] and in the south-western part of the Korean Peninsula[6], and they belong to almost the same age.

3 Koezuka and Wakiya 2003, 38-39.
4 Oga 2010a, 213-230.
5 Kitsurin-shō hakubutsukan 1988; Fujita 1994; Yanagida 2008, 254-274.
6 Okauchi 1993, 35-54.

Fig. 1 Tubular I (Ikisa-nakabaru No.1 jar burial 伊岐佐中原 1 号甕棺墓)

Fig. 3 Tubular II not containing Chinese blue pigments (Nagasuna No.3 site 長砂第 3 遺跡)

Fig. 4 Tubular III (Yokota No.1 Site 横田 1 号遺跡)

Fig. 5 Tubular Others (Otomo site 大友遺跡)

Fig. 2 Tubular II containing Chinese blue pigments (Arimoto site 有本遺跡; Koezuka 2002)

The second type of tubular beads, Tubular II, consists of well-shaped tubular beads made by drawing and twisting the glass. The feature of Tubular II is that most of them are coloured by Chinese blue, an artificial blue pigment.[7] There are also some exceptions that do not contain Chinese blue pigments (Fig. 3). Tubular II beads appeared in the first century BCE, and the most important

7 Koezuka 2002, 705-717.

fact is that they appeared together with glass *bi* discs[8]. And the number of Tubular II is greater, compared to other types of lead-barium glass beads in Japan.

Tubular III consists of middle-sized beads, and they show a slight convex curve in the shaft. They are probably made by folding the hot glass around an iron rod. Copper is used as the colouring agent, and the appearance is opaque light greenish blue. Tubular III appeared in the first century CE, and they were distributed simultaneously with Tubular II. The distribution period of Tubular III, however, was short; they were distributed only in the first century CE[9].

Comma-shaped beads are also common among lead-barium glasses found in Japan. However, compared to tubular beads, the number of the comma-shaped beads is relatively small, since they were rarely buried in large quantities in single burial environments. As for making techniques, a casting method using an open mould is the most common, and the drawing with twisting method follows. There is also a unique technique of bending tubular beads. There are differences between the provinces concerning the details of the shape. There are various types of comma-shaped beads, and a systematic classification has not been established. In this study, we analysed three specimens. Since the features of the three differ greatly, we distinguish them from each other and call them Comma I, Comma II, and Comma III, respectively (Fig 6).

Fig. 6 Comma-shaped beads
Left: Comma I (Higashiyamada-ipponsugi site 東山田一本杉遺跡)
Middle: Comma II (Dasaka No.3 tomb 駄坂 3 号墳)
Right: Commma III (Hazezuka No.1 kofun 砂魚塚 1 号墳)

8 Oga 2010a, 213-230.
9 Oga 2010a, 213-230.

Comma I is the most common type among the comma-shaped beads. They have a transparent blue-green appearance, and seem to be made using an open mould technique. Comma I beads were distributed continuously from the first century BCE to the second century CE, and the time of their first appearance dates possibly back to the second century BCE. We suppose that this type of comma-shaped beads was made in Japan, since moulds and cullet of the same colour were unearthed[10].

The Comma II type is also thought to have been made by an open mould technique, but this is not certain because this type has very few bubbles and the surface is remarkably polished. These beads have a translucent blue appearance. Small numbers of this kind of comma-shaped bead have been found in tombs of the second to the fourth centuries CE.

Comma III contains very few bubbles and the production technique is unclear. There are many cracks inside of the Comma III type, and this feature is considered to be related to the making technique. These beads often feature a marble pattern which consists of transparent light-blue portions and colourless portions. A small number of Comma III has been unearthed so far, only from tombs dating to the fifth and sixth centuries CE.

As for eye beads, only six have been found in Japan. Most of them were unearthed from sites dating to the third to second centuries BCE, but one specimen analysed in this study was excavated from a tomb dating to the latter half of seventh century CE (Fig. 7). It is considered to have been handed down for an exceptionally long period of time. A number of similar beads have been found from the tombs of the Warring States period (475–221 BCE) in China[11].

The winding method is the most common production technique for small lead-barium glass beads. We call the small beads made by a winding method Small I type. There are also small beads made from cullet by cold working and those made by cutting Tubular II beads in cold condition. We call the former Small II and the latter Small III types.

As for the shape of Small I, globular or spheroid beads are common (Fig. 8). As special examples, ring-shaped beads also exist (Fig. 9). In this study, we analysed two specimens of Small I that were found in tombs from the second century BCE. Other examples show that Small I were distributed until the second century CE. Small I beads vary in colour, and there is a high possibility that chronological changes in chemical compositions also exist. However, these issues are subjects for future investigation.

10 Oga 2010c, 27-35.
11 Zhao 2012, 177-215.

Fig. 7 Eye bead
(Hakuji No.1 tunnel tomb
白地 1 号横穴墓)

Fig. 8 Small I
(Nakabaru site 中原遺跡)

Fig. 9 Small I
(Mutabe site 牟田辺遺跡)

Fig. 10 Small II Old
(Takagi site 高木遺跡)

Fig. 11 Small II New
(Karako-kagi site 唐古・
鍵遺跡)

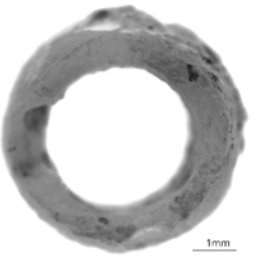
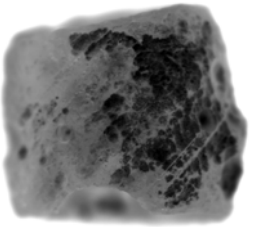


Fig. 12 Small III
(Mitoma No.B8 kofun
三苫 B8 号墳)



Small II beads have misshapen appearances caused by the making technique. Although beads of the Small II type have a translucent or opaque blue appearance, when they date to the second century BCE (Fig. 10), those dating later than the first century BCE often have a transparent blue-green appearance (Fig. 11). We call the former Small II Old, and the latter Small II New. Some of the

Small II New type were manufactured at the Naguoka 奈具岡 site, a settlement site of the first century BCE[12].

Small III beads are short and cylindrical in shape (Fig. 12). Although they were made by cutting Tubular II beads, Small III coloured by Chinese blue pigment have not yet been found. In addition, Small III type beads were found in tombs dating to the first to second centuries CE, and they were therefore later in appearance than Tubular II beads. The distribution of all of the lead-barium glass artefacts mentioned above stopped by the end of the second century CE, with minor exceptions.

Chemical Compositions

Based on the results of X-ray fluorescence analysis, lead-barium glasses in ancient Japan can be classified into more than five compositional groups. We also detected a chronological change in chemical compositions, and a relationship between chemical composition and bead-making technique (Table 2).

Lead-barium glasses from the third to second centuries BCE, the oldest group in Japan, including an eye bead, Tubular I, Small I and Small II Old, have lower concentrations of lead oxide (PbO) and barium oxide (BaO) (Fig. 13). On the other hand, they have higher concentrations of sodium oxide (Na_2O). These compositional characteristics are correlated to their lower specific gravity. In addition, they have low concentrations of calcium oxide (CaO). However, although one example of Small I and one of Small II Old belong to same period, they have different characteristics in their chemical compositions. That is, they have higher concentrations of PbO, moderate BaO and Na_2O. Such characteristics of chemical compositions differ from those of any other lead-barium glass artifacts analyzed in this study.

Lead-barium glasses distributed in the first century BCE, consisting of Tubular II Old beads, have high concentrations of BaO, moderate PbO and quite low concentrations of Na_2O and CaO. Tubular others, and most of the Small II New, which were distributed between the first century BCE and the second century CE, have high concentrations of PbO, moderate BaO and Na_2O, and lower levels of CaO. The compositional results are consistent with their higher specific gravity. On the other hand, single examples of the Small II New and Comma I types, that belong to same period, have different features of chemical composition. Comma I beads are similar in composition to Tubular III beads described below. The one specimen of Small II New with different

12 Oga 2010c, 27-35.

features of chemical compositions can be judged as an artefact which belonged to the first group and had been handed down for periods.

Table 2 Chemical compositions of lead-barium glasses by XRF

Site	Age	Type	Specific gravity	Chemical Composition														
				Na_2O	MgO	Al_2O_3	SiO_2	K_2O	CaO	MnO	Fe_2O_3	CoO	CuO	PbO	Rb_2O	SrO	ZrO_2	BaO
Nakabaru site	AD1	Tubular I		2.0	0.6	1.9	69.0	1.2	0.5	—	0.16	—	0.40	19.1	0.00	0.05	0.12	3.4
Nakabaru site	unknowm	Tubular I		4.9	0.8	3.4	56.1	0.9	1.0	—	0.60	—	0.80	21.6	0.02	0.04	0.19	8.4
Nakabaru site	unknowm	Tubular I		3.8	0.5	2.5	63.5	0.2	1.0	—	0.73	—	0.67	19.5	0.02	0.09	0.19	6.4
Ikisa-nakabaru site	AD1	Tubular I	3.0	8.6	0.7	0.9	58.1	0.1	0.6	0.03	0.19	0.02	0.58	20.2	0.01	0.10	0.34	9.6
Fukiage site	BC1	Tubular II Old		1.3	0.2	1.2	51.0	0.1	0.5	0.07	0.22	0.02	0.72	30.5	0.01	0.12	0.29	13.8
Fukiage site	BC1	Tubular II Old	3.5	2.3	0.1	0.8	43.8	0.2	0.3	0.08	0.19	0.03	0.69	33.6	0.01	0.18	0.23	17.6
Fukiage site	BC1	Tubular II Old	3.7	2.2	0.2	0.8	47.9	0.2	0.4	0.09	0.24	0.03	0.58	30.3	0.01	0.10	0.42	16.7
Fukiage site	BC1	Tubular II Old	3.2	1.7	0.2	1.0	41.7	0.1	0.5	0.07	0.30	0.03	0.78	35.2	0.02	0.17	0.45	17.7
Fukiage site	BC1	Tubular II Old		1.8	0.2	0.5	46.8	0.1	0.4	0.06	0.16	0.02	0.72	34.7	0.02	0.17	0.24	14.2
Fukiage site	BC1	Tubular II Old		2.2	0.2	0.8	50.1	0.1	0.4	0.04	0.29	0.02	0.64	30.7	0.01	0.09	0.35	14.0
Fukiage site	BC1	Tubular II Old		0.9	0.2	0.7	60.1	0.1	0.3	0.04	0.17	0.03	0.58	26.6	0.03	0.10	0.19	10.0
Nagasuna No.3 site	unknowm	Tubular II New		5.1	0.3	0.2	47.3	0.1<	1.5	0.10<	0.13	0.01<	0.42	31.3	0.01<	0.12	0.01<	13.6
Sakai-yaishi site	unknowm	Tubular II New		7.2	1.5	0.2	48.1	0.0	2.7	0.11	0.12	0.03	0.09	26.7	0.01<	0.07	0.24	12.9
Higashiyama site	AD1	Tubular III		6.2	0.7	0.6	45.7	0.1	1.6	0.11	0.23	0.02	0.30	34.0	0.03	0.12	0.00	10.4
Higashiyama site	AD1	Tubular III		5.5	0.1	0.5	45.3	0.1	0.8	0.07	0.27	0.01	0.37	35.8	0.00	0.10	0.32	10.8
Higashiyama site	AD1	Tubular III		3.4	0.0	0.3	44.4	0.1	0.8	0.07	0.26	0.03	0.45	38.0	0.00	0.16	0.41	11.7
Higashiyama site	AD1	Tubular III		6.2	0.7	0.6	45.6	0.1	1.6	0.06	0.24	0.02	0.28	33.7	0.01	0.08	0.25	10.6
Higashiyama site	AD1	Tubular III		5.8	0.5	0.4	46.0	0.0	1.1	0.08	0.27	0.02	0.30	33.9	0.00	0.15	0.08	11.3
Higashiyama site	AD1	Tubular III	3.6	5.9	0.3	0.4	45.3	0.0	1.1	0.08	0.26	0.02	0.30	34.3	0.00	0.11	0.27	11.6
Higashiyama site	AD1	Tubular III		5.1	0.3	0.6	45.1	0.1	0.8	0.07	0.23	0.02	0.38	37.3	0.02	0.13	0.05	9.9
Higashiyama site	AD1	Tubular III		5.0	0.2	0.5	44.7	0.0	0.8	0.09	0.24	0.02	0.35	37.1	0.00	0.11	0.35	10.4
Higashiyama site	AD1	Tubular III		3.5	0.0	0.3	44.6	0.1	0.9	0.11	0.26	0.03	0.38	38.0	0.00	0.08	0.38	11.5
Higashiyama site	AD1	Tubular III		5.2	0.2	0.6	45.6	0.1	0.8	0.06	0.23	0.02	0.38	36.4	0.00	0.15	0.18	10.1
Higashiyama site	AD1	Tubular III	3.7	3.4	0.0	0.4	45.0	0.1	0.9	0.08	0.25	0.02	0.41	37.8	0.06	0.18	0.69	10.8
Higashiyama site	AD1	Tubular III		4.7	0.3	0.5	44.3	0.1	0.8	0.06	0.24	0.03	0.36	38.5	0.02	0.18	0.00	10.0
Higashiyama site	AD1	Tubular III	3.8	4.9	0.2	0.5	44.0	0.1	0.8	0.08	0.26	0.04	0.42	37.7	0.00	0.16	0.15	10.8
Oshinooka site	AD1	Tubular III		4.4	0.0	0.3	44.5	0.0	1.5	0.09	0.26	0.04	0.54	37.4	0.00	0.14	0.00	10.8
Oshinooka site	AD1	Tubular III	3.4	4.7	0.2	0.4	44.9	0.0	0.8	0.10	0.22	0.03	0.42	37.1	0.00	0.11	0.20	10.8
Oshinooka site	AD1	Tubular III	3.2	6.0	0.2	0.4	44.5	0.0	0.9	0.10	0.21	0.03	0.37	35.4	0.00	0.10	0.43	11.4
Oshinooka site	AD1	Tubular III		4.9	0.0	0.2	45.6	0.0	1.3	0.08	0.30	0.02	0.30	34.9	0.00	0.12	0.36	11.9
Oshinooka site	AD1	Tubular III	3.3	5.3	0.1	0.6	45.8	0.0	0.9	0.07	0.23	0.01	0.39	35.0	0.00	0.12	0.21	11.3
Oshinooka site	AD1	Tubular III		5.8	0.2	0.5	44.5	0.1	1.3	0.11	0.28	0.02	0.56	35.4	0.03	0.09	0.32	11.0
Yokota No.1 site	AD1	Tubular III		2.9	0.3	0.9	61.7	0.0	1.2	—	0.21	—	0.39	24.0	—	0.08	0.27	8.1
Higashiyamada-ipponsugi site	AD1	Tubular III	3.5	4.7	0.2	0.5	46.8	0.1	0.9	0.08	0.34	0.02	0.73	33.6	0.02	0.16	0.18	11.8
Higashiyamada-ipponsugi site	AD2	Tubular III	3.5	5.9	0.3	0.8	46.3	0.1	0.8	0.07	0.36	0.04	0.72	33.1	0.04	0.14	0.18	11.2
Otomo site	unknowm	Tubular others		4.3	0.2	1.5	34.2	0.2	0.5	0.08	0.15	0.03	0.64	44.0	0.03	0.13	0.20	13.9
Higashiyamada-ipponsugi site	AD2	Comma I		4.4	0.2	0.6	44.7	0.1	0.9	0.06	0.26	0.02	0.42	36.3	0.00	0.14	0.31	11.6
Dasaka No.3 kofun	AD2	Comma II	3.7	5.7	0.3	0.5	48.0	0.1	1.8	0.08	0.18	0.02	0.26	30.7	0.02	0.05	0.11	12.3
Hazezuka No.1 kofun	AD6	Comma III		6.2	1.4	0.2	48.8	0.0	2.6	—	0.07	—	0.03	27.6	0.02	0.05	0.19	12.8
Hakuji No.1 tunnel tomb	AD7	Eye beads	3.0	10.3	1.1	0.9	59.8	0.1	0.9	0.03	0.12	0.00	0.97	19.5	0.00	0.06	0.27	5.9
Nakabaru site	BC2	Small I		3.0	0.7	3.0	56.7	0.3	0.8	—	0.33	—	0.26	22.1	0.01	0.09	0.26	12.6
Mutabe site	BC2	Small I		10.0	0.7	0.6	49.5	0.0	0.6	0.06	0.15	0.03	0.42	30.2	0.01	0.13	0.36	7.2
Takagi site	BC2	Small I		4.8	0.1	0.4	45.3	0.0	0.8	0.09	0.16	0.04	1.26	31.9	0.02	0.13	0.46	14.6
Takagi site	BC2	Small II Old	3.3	9.3	0.7	1.3	53.0	0.1	0.6	0.05	0.31	0.04	0.96	23.9	0.02	0.08	0.09	9.5
Takagi site	BC2	Small II Old	3.5	8.0	0.7	0.6	53.3	0.2	1.0	0.04	0.12	0.01	0.97	29.1	0.00	0.12	0.34	5.7
Takagi site	BC2	Small II Old	3.3	8.0	0.4	0.7	53.8	0.1	0.8	0.06	0.19	0.01	0.85	25.0	0.04	0.12	0.20	9.9
Takagi site	BC2	Small II Old	3.3	8.9	0.3	0.7	54.3	0.0	0.8	0.03	0.20	0.03	0.67	24.1	0.01	0.13	0.29	9.6
Takagi site	BC2	Small II Old	4.0	4.0	0.2	0.8	42.8	0.3	1.0	0.08	0.16	0.02	0.52	34.3	0.02	0.12	0.12	15.6
Takagi site	BC2	Small II Old	3.5	8.9	0.4	0.9	50.4	0.1	0.8	0.05	0.18	0.02	1.09	29.5	0.00	0.06	0.09	7.6
Karako-kagi site	unknowm	Small II New	4.6	2.6	0.1	0.3	39.1	0.1	0.4	0.06	0.07	0.02	0.77	44.0	0.00	0.15	0.39	11.9
Karako-kagi site	unknowm	Small II New	4.3	3.7	0.2	0.3	38.6	0.1	0.5	0.11	0.08	0.02	0.57	45.0	0.00	0.12	0.47	10.2
Karako-kagi site	BC1-AD1	Small II New	4.3	3.6	0.2	0.3	37.0	0.2	0.4	0.11	0.09	0.05	0.69	43.1	0.00	0.05	0.36	13.8
Tabata-nishidaidori site	AD2	Small II New		4.0	0.7	1.9	62.0	0.6	0.8	0.05	0.23	0.02	0.52	22.1	0.00	0.12	0.15	5.6
Mitoma No.B8 kofun	AD6-7	Small III		2.9	1.0	0.5	63.0	0.1	1.8	0.06	0.11	—	—	21.9	—	—	—	8.6

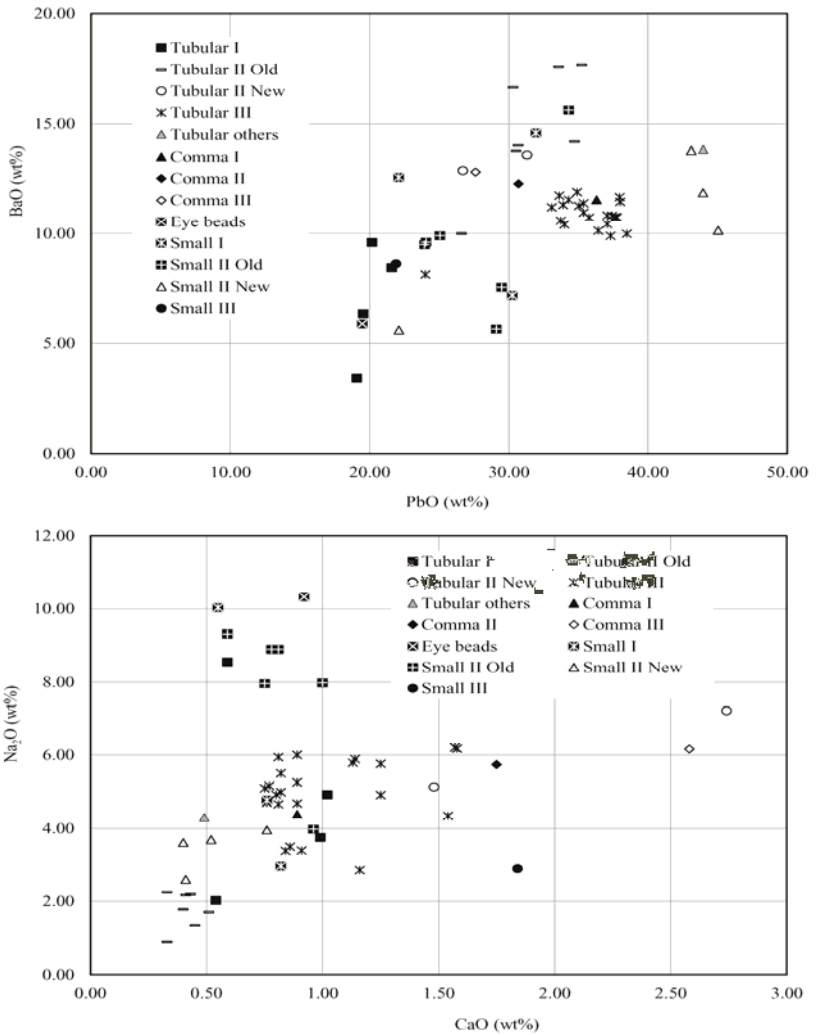


Fig. 13 Comparison of the element content between lead-barium glass groups (upper): PbO vs. BaO, (lower): CaO vs.Na_2O.

Tubular III beads were distributed in a limited period of time in Japan. The chemical compositions of the Tubular III type are also distinctive. Their concentrations of PbO, BaO, Na_2O and CaO are all moderate. The differences in chemical compositions among Tubular III beads are relatively small.

Lastly, it should be noted that the latest lead-barium glasses after the second century CE, including Tubular II New, Comma II, Comma III and Small III

types, show the tendency of higher concentrations of CaO. The concentrations of PbO, BaO and Na_2O are at moderate levels. Lead-barium glasses with high levels of CaO are reported in the Korean peninsula.[13] They belong to almost the same period. Therefore, some of the high calcium types of lead-barium glass could have some relations with those in Korean peninsula.

Production Areas

Next, we discuss the production area and distribution routes of lead-barium glasses found in Japan. Lead isotope ratios are effective in presumption of production areas of the glasses containing high PbO like lead-barium glasses. Fig. 14 shows the lead isotope ratios of lead-barium glasses and several bronze artefacts as references.

First of all, we explain the lead isotope ratios of bronze artefacts found in Japan. The lead isotope ratios of bronze artefacts dating from the Yayoi to Kofun period are concentrated on several ranges. Reference group Area D is set based on the lead isotope ratios of bronze arms and bells made in the Korean peninsula or Japan in the beginning to early Middle Yayoi period (third century BCE). Although the mining region of Area D is assumed to be in China, it is difficult to narrow down the location.

Reference group Area A is set based on the lead isotope ratios of bronze mirrors made in China in the Western Han 西漢 Dynasty (206 BCE–8 CE). The lead isotope ratios of the bronze arms and bells made in Japan in the mid- to latter half of the Middle Yayoi period (second to first centuries BCE) fall in the same range. The mining region of Area A is considered to be in the northern part of China. As the lead isotope ratios of the bronze arms and bells made in Japan in the late to ending Yayoi period (first to second centuries CE) are concentrated in the limited range among the Area A, they probably originate from a single mine. However, since this fact is not the point of argument in this study, we did not distinguish them from other specimens of the Area A.

Although the lead isotope ratios of the reference group of Area B fall in the same range, they can be divided into three sub-groups which differ from each other in production area. Area B1 is based on the bronze mirrors made in China in the Eastern Han 東漢 Dynasty (25–220 CE), and the mining region of Area B1 is considered to be in the southern part of China. Area B2 consists of the lead isotope of the bronze mirrors made in the Wei 魏 Dynasty (220–265

13 Kim Gyuho (2013), 130-141.

CE), which originated in mines from the territory of the Wei Dynasty. Area B3 consists of the bronze artefacts, including mirrors, made in the northern part of China or north-western part of the Korean peninsula in the fourth century CE, and the mining region is considered to be in the northern part of China.

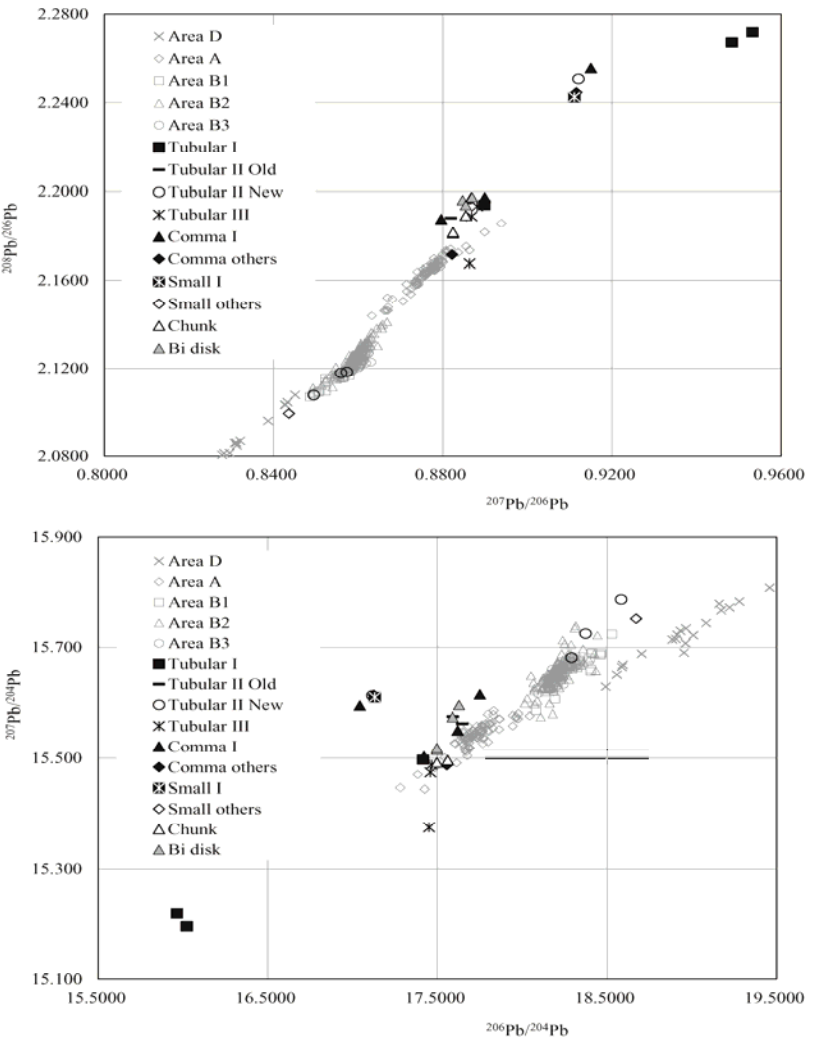


Fig. 14 Lead isotope ratios for compositional groups of glass beads excavated in Japan, (upper): $^{207}Pb/^{206}Pb$ vs. $^{208}Pb/^{206}Pb$, (lower): $^{206}Pb/^{204}Pb$ vs. $^{207}Pb/^{204}Pb$

In Fig. 14 we show the lead isotope ratios of lead-barium glass artefacts that are clear with regard to the location of their discovery, the shape, and making techniques. All of these lead isotope ratios have been published in previous studies.[14] It is necessary to keep in mind that since there are few specimens with both the reliable chemical compositions and lead isotope ratios in previous studies, it is difficult to compare the lead isotope ratios directly with the results of chemical compositions mentioned above.

The lead isotope ratios of lead-barium glasses are divided into four groups: Group I (0.9400〜0.9600 for $^{207}Pb/^{206}Pb$), Group II (0.9000〜0.9200 for $^{207}Pb/^{206}Pb$), Group III (0.8800〜0.8900 for $^{207}Pb/^{206}Pb$) and Group IV (0.8400〜0.8600 for $^{207}Pb/^{206}Pb$). The lead isotope ratios indicate that the lead used for Group I, Group II and Group III originated from mines different from those that produced the lead used for bronze artefacts. This fact should be noted when we discuss the production areas of these lead-barium glasses, although there is a high possibility that the lead used for Group I, Group II and Group III also originated from China, since lead-barium glasses are considered to be unique in ancient China. We suppose that Group IV corresponds to Area B1.

It is unlikely that the lead isotope ratios correlate to the classification units of lead-barium glass artefacts based on the shapes and making techniques. On the other hand, there is the correlation on a certain level between the lead isotope ratios and the production age. First of all, Group I consists of only the oldest specimens of Tubular I. Most of the various kinds of lead-barium glass artefacts made mainly between first century BCE and first century CE belong to Group III, and a few of them also belong to Group II. Only the lead-barium glass artefacts made in the first to second century CE belong to Group IV. These facts correspond to the transition of origins of lead used for bronze mirrors made in ancient China. Such chronological transition of lead-barium glasses is synchronized with the transition of origins of lead used for bronze mirrors made in ancient China. So far, however, it is difficult to match each group to a certain mine and thus to narrow down the production area of lead-barium glasses.

14 Aoki 1989, 73-91; Brill et al 1979, 87-109; Fujimura and Hirao 2010, 138-153; Furihata 2012, 61-69; Hirao 2003, 346-368; Hirao et al 1995, 860-901; Koezuka 2000, 63-67; Koezuka 2004, 98-102; Mabuchi and Hirao 1985, 78-82; Mabuchi et al.1991, 206-215; Saito 1996, 221-226; Yamasaki 1987, 314-317.

Distribution Routes

Since little is as yet known about not only the production area of the lead-barium glass itself but also the area where the products were made from this lead-barium raw glass, the distribution routes of lead-barium glass to Japan need to be estimated mainly based on the distribution of products made from lead-barium glass.

Concerning Tubular I beads, the distribution area is limited to northern Kyūshū in Japan. The same kind of tubular beads have also been found in Jilin province in China and in the south-western part of the Korean peninsula, and they belong to almost the same age. These findings indicate the production area and the trade route of Tubular I beads. This route was also used to introduce advanced metal tool culture to Japan.

As for Tubular II beads, in the first century BCE they were distributed in northern Kyūshū. An important fact is that beads of the Tubular II type were accompanied by glass *bi* discs. The production of glass *bi* discs is considered to have developed around Hunan province near the midstream of the Yangtze River in China during the Warring States period. Glass *bi* discs belonging to the Western Han Dynasty were excavated in the coastal areas south of the Yangtze River. The most surprising detail is that the number of Tubular II beads and glass *bi* discs is relatively large in Japan, while such glass artefacts have not yet been found in Korea. Therefore it is supposed that Tubular II beads were directly brought to Japan from Jiangnan 江南 region in China[15].

In the first and second centuries CE, the distribution area of Tubular II beads shifted to the San'in 山陰 and Setouchi 瀬戸内 regions, and the northern part of Kinki 近畿 region. On the other hand, Tubular II beads became rare in northern Kyūshū at that time.[16] Although in the southern part of the Korean peninsula there are some examples of the Small III type, made by cutting of Tubular II ones, glass beads coloured by Chinese blue, which are common in Japan, have not been found yet. This evidence confirms that in this region, lead-barium glass artefacts had continued to be imported directly from the southern part of China since former periods.

It is inferred that Tubular III beads have been produced in China, but we have not found any evidence to narrow down the production area. The Tubular III type was distributed along the coastal areas of Japan Sea from northern

15 Oga 2010b, 231-254; Oga and Tamura 2013, 35-65.
16 Oga 2010a, 213-230.

Kyūshū to the northern part of Kinki region[17]. This kind of tubular bead is also rare in the Korean peninsula so far, and they also are supposed to be brought to Japan directly from China. In addition, the production areas of Tubular II and Tubular III beads are probably not the same because the Tubular II type differs from Tubular III in making technique, chemical compositions, and the distribution period in Japan.

Concerning Comma I and Small II, the production sites are found in Japan.[18] In Suku-gotanda 須玖五反田 site in northern Kyūshū, for example (second century CE), some moulds and glass cullet used for making Comma I beads were unearthed. Another example is the Naguoka site in the northern part of the Kinki region (first century BCE), where some glass cullet and unfinished products of the Small II New type were found.[19] Since Small II beads were found only in Japan, these glass beads are supposed to have been manufactured in Japan by using lead-barium glass cullet made in China.

Concerning Comma III and Small III examples, some have been found in the southern part of the Korean peninsula, and these kinds of beads were probably made in this region. We suppose that eye-beads and Small I beads were manufactured in China. However, they are rare in Japan, and it is difficult to clarify the distribution route.

Conclusions

In this study, we categorized lead-barium glass artefacts in ancient Japan and discussed each production area and trade route. Although there are many issues that need further examination, we clarified the basic situation. Lead-barium glasses in ancient Japan can be classified into more than five compositional groups, and we found chronological changes in chemical composition and in the relationship between chemical composition and bead-making technique.

Tubular beads are the most common lead-barium glass artefacts in Japan, and they can be divided into three types: Tubular I, Tubular II and Tubular III, based on the manufacturing methods and shapes. These three types of tubular beads are also different from each other in chemical composition and distribution period in Japan.

17 Oga 2010a, 213-230.
18 Oga 2010c, 27-35.
19 Oga et al, 2005, 1-12.

The production area of Tubular I beads is probably the northeastern region of China including Jinlin Province. They are considered to have been brought to northern Kyūshū via the southwestern part of the Korean peninsula in the third to second centuries BCE.

Concerning the production area of Tubular II beads, the region near the Yangtze River in China is one possibility. They were probably brought to Japan directly from the Jiangnan region in China. The production area of Tubular III, however, remains unclear.

In the future, it is necessary to develop a comparative study with the lead-barium glass artefacts found in the Korean peninsula and mainland China in order to narrow down the production area of each type of lead-barium glass artefact and its distribution route.

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