Carbon contents in the manufacturing process of modern black pottery at San Nikolas, northern Luzon, the Philippines

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1. Research purpose

The AMS dating method has made it possible to directly date pottery samples. However carbonized residue on the exterior surface of the pottery could exhibit an "old wood effect", since this carbon might include fuel carbon from a large piece of wood. Furthermore, carbonized residue on the interior surface of the pottery is usually formed from carbonized food remains, possibly resulting in traces of the "marine reservoir effect" caused when marine food resources were cooked (Nakamura and Iwahana 1990; Nakamura et al. 1990; Kolic 1995; Nakamura et al. 2001; Oda and Yamamoto 2001).

However, organic carbons sealed within the pottery material will give the age when the pottery was manufactured. In the Philippines, black pottery, or pottery colored black through the adsorption of organic carbon, was excavated from Lal-lo shell midden sites, in Northeast Luzon (Ogawa, 2000, 2002). Philippino black pottery belonging to the Iron Age is traceable to the Chinese black pottery of the Longshan culture in BC 20c to BC 17c. Similar black pottery is distributed in, Thailand (Ban Chaing culture in BC 15c to AD 3c), Bo-hai (AD 7c to 9c) and Okinawa (Kamui-yaki in AD 12c to 15c) in Japan. Black pottery in the Philippines is still produced at San Nikolas, in the Ilocos Norte, northwest Luzon (Fig. 1).

The manufacturing process of the black pottery at San Nikolas was observed and documented by the author and the National Museum of the Philippines in 2004. Details of the manufacturing procedure are described in Appendix B-5 (Mihara and Koike, 2005). In this process sand is mixed with lumps of clay to increase strength. It is then squeezed and formed into pots on a lathe. The pot is patted to fix the shape, and polished after being dried. Red slip is applied to the surface of the pot and dried. The pot is then fired in the

ground under a reduced fire. While the pottery is being heated in red, it is taken out from the fire and immediately rice husk is covered on the pottery surface to produce a black carbon-adsorbed coloring (Fig. 2). If such black pottery retains enough carbon introduced from rice husk, it may be useful for the ^{14}C dating. As a preliminary research, the authors analyzed the carbon contents of the black pottery shards (Mihara et al, 2003), and suggested that well-preserved black pottery shards included enough amounts of carbon for the ^{14}C dating (Mihara et al, 2004).

To construct a detailed chronology using the AMS ^{14}C dating, I analyzed the carbon contents and stable carbon isotope ratios of carbons within the pottery according the manufacturing process (1) the carbon contents of the raw material such as clay, (2) the carbon contents of the first fired pottery, (3) the carbon contents of the carbon-adsorbed pottery. To examine the most suitable method of extracting carbon from the pottery, several samples were collected from the surface and inner part of the pottery, and drilling and grinding sampling for each pottery sample.

2. Materials

The manufactured pottery and the raw materials were collected during the research of the manufacturing process at San Nikolas in 2004.

Raw materials

Samples of the raw materials of the pottery were collected from a potter; a lump of clay (05IS01) which had been taken from a rice field, sand (05IS02), a mixed sample of clay and sand (05IS03) squeezed at the forming process, red clay (05IS04) which was obtained through trading, rice husk (05IS07) used for carbon adsorption of the pottery and charred rice husk (05IS08) remained after adsorbing carbon.

Pottery samples before carbon adsorption (Fig. 3)

The pottery before carbon adsorption, with only red-slipped surface, was made in the same process, but sampled before carbon adsorption. Two types of pottery, a pot and a plate were used for the samples. The pot was formed by patting and polishing, and had a thin, fine body. The plate was formed without patting and polishing, and had a coarse, thick body. A total of 4 pottery samples, 2 pots (05PO01 and

05PO02) and 2 plates (05PO03 and 05PO04) were analyzed.

Black pottery samples (Fig. 3)

The black pottery is adsorbed carbon from rice husk and colored black in manufacturing process. A total of 4 black pottery samples, 2 pots (05PO05 and 05PO06) and 2 plates (05PO07 and 05PO08) were analyzed.

3. Methods

Four sample categories were collected from each pottery; the surface (S) and inner part (I) of the pottery, drilling (D) and grinding (G) with a different sampling method. Powder samples were chemically treated by acid-alkali-acid (AAA) treatment, to remove acid and alkali soluble materials, such as humic acids and fulvic acids which may have been incorporated from burial soil. The AAA treatment consisted of successive cleaning by 0.1N HCl solution at 80 °C, 0.01N NaOH solution at 80 °C, and again by 0.1N HCl solution at 80 °C. The residues were rinsed to neutral and dried.

After being chemically treated, the charred materials were analyzed for Carbon contents (%C) and δ^{13}C values using an automated nitrogen and carbon analysis mass spectrometer (ANCA-mass, Europa Scientific Ltd). Analyses were done on 2 sets of 10-mg fractions from each pottery sample, and the average was calculated.

For AMS analysis, samples of the pretreated charred material containing about 2 mg of carbon were combusted at 850 °C for 4 hour in evacuated and sealed Vycor tubes together with CuO and Ag. The CO_2 was then purified cryogenically (using normal-pentane slush at -130 °C and ethanol slush at -90 °C) on a vacuum line. The purified CO_2 samples were then reduced to graphite (Kitagawa et al. 1993). The graphite samples were measured against a standard (NBS-II) using the Tandetron AMS (Model-4130 AMS, HVEE) at Nagoya University (Nakamura et al. 2000). We corrected for carbon isotopic fractionation using the δ^{13}C values provided by the AMS.

4. Results

The results of the %C and δ^{13}C analysis were shown in Table 1.

4-1. The %C and $\delta^{13}C$ values for raw materials

Carbon and nitrogen contents were obtained with a total amount of ^{12}C measured by ANCA-mass, shown as %C. For the $\delta^{13}C$ measurements, at least 0.1mg of carbon was necessary for $\delta^{13}C$ measurement from each sample. Samples which had a lower %C did not have enough reliability in $\delta^{13}C$, especially the pottery samples before carbon adsorption.

Raw materials

The weight of samples before and after the AAA treatment were shown in Fig. 4, and the recovery of samples after the AAA treatment were shown in Fig. 5. The %C values before and after the AAA treatment were shown in Fig. 6. The %C and $\delta^{13}C$ values were shown in Fig. 7.

Clay sample: The %C and $\delta^{13}C$ values of the clay sample before the AAA treatment were 0.50% and -12.7‰, and those after the AAA treatment were 0.10% and -15.7‰, respectively. It suggests that most of the carbon in the clay was removed by the AAA treatment.

Sand sample: The %C and $\delta^{13}C$ values of the Sand sample before the AAA treatment were 0.01% and -28.0‰, and those after the AAA treatment were 0.01% and -26.8‰, respectively. The sand had almost no carbon, being an ignorable factor on the measurement.

Mixed sample with clay and sand: The %C and $\delta^{13}C$ values of the mix sample before the AAA treatment were 0.45% and -14.4‰, and those after the AAA treatment were 0.04% and -22.6‰, respectively. This sample was composed of clay and sand as the pottery material, and the change of carbon was similar to the clay sample. After the AAA treatment more fine clay was washed out from the sample than sand, resulting in a high proportion of sand and a lower %C in the mixed sample.

Red clay sample: The %C and $\delta^{13}C$ values of the Red clay sample before the AAA treatment were 1.06% and -16.0‰, and those after the AAA treatment were 0.21% and -17.0‰, respectively. The red clay had a higher %C. However, this was used for decoration only on the surface, and fired directly. The carbon should be removed as shown on the results of pottery before carbon adsorption, which were described later, had only ignorable carbon to the measurement.

Rice husk sample: The rice husk is the resource of the adsorbing carbon for the black pottery. The %C and $\delta^{13}C$ values of the rice husk sample before the AAA treatment were 44.83%, and -27.8‰, and those after the AAA treatment were 54.5% and -28.3‰, respectively. The data showed no change in $\delta^{13}C$ value after the AAA treatment.

Charred rice husk sample: The %C and $\delta^{13}C$ values of the charred rice husk samples before the AAA treatment were 42.6%, and -27.8‰, and those after the AAA treatment were 41.33% and -27.8‰, respectively. These charred rice husk samples must be same as used for the adsorbing carbon. Consequently these results, showing that there was no difference between the rice husk sample and this charred rice husk sample, must be an important fact for this experiment.

The results of %C measurements, indicated that most of carbon in the raw materials must be removed by the AAA treatment.

4-2. The %C and $\delta^{13}C$ values for pottery samples

Since the AAA treatment was necessary to remove contaminated carbon for the AMS ^{14}C dating, the results of pottery samples were described only samples after the AAA treatment residue.

The recovery rates of samples after the AAA treatment were shown in Fig. 8. The recovery rates of Surface Grinding (SG) samples were 64 ± 5% for the pottery before carbon adsorption and 60 ± 6% for the black pottery. Those of Surface Drilling (SD) samples were 58 ± 7% for the pottery before carbon adsorption and 53 ± 3% for the black pottery. Those of Inner part Grinding (IG) samples were 68 ± 2% for the pottery before carbon adsorption and 67 ± 5% for the black pottery. Those of Inner part Drilling (ID) samples were 57 ± 5% for the pottery before carbon adsorption and 63 ± 8% for the black pottery.

The %C values before and after the AAA treatment were shown in Fig. 9. The %C of SG samples were 0.04 ± 0.03% for the pottery before carbon adsorption and 0.58 ± 0.28% for the black pottery. Those of SD samples were 0.06 ± 0.04% for the pottery before carbon adsorption and 0.78 ± 0.36% for the black pottery. Those of IG samples were 0.10 ± 0.03% for the pottery before carbon adsorption and 0.22 ± 0.18% for the black pottery. Those of ID samples were 0.09 ± 0.03% for the pottery before carbon adsorption and 0.27 ±

0.25% for the black pottery.

Considering the results of these examinations, the drilling sampling method produced a lower recovery rate but a higher %C value than the grinding sampling method in almost all pottery samples. The drilling sampling produces fine powder, inorganic fraction such as clay, might be removed more than the grinding sample in the AAA treatment. Hence, the drilling method which recovered relatively higher carbon from pottery is more suitable than the grinding method for extracting powder sample. After this, samples were described as drilling samples.

Pottery samples before carbon adsorption (Fig. 5)

The %C and δ^{13}C values were shown in Fig. 10.

The pot 05PO01: The %C and δ^{13}C values of SD sample were 0.02% and -31.5‰, those of ID sample were 0.04% and -19.1‰, respectively.

The pot 05PO02: The %C and δ^{13}C values of SD sample were 0.04% and -26.1‰, those of ID sample were 0.08% and -18.8‰, respectively.

The plate 05PO03: The %C and δ^{13}C values of SD sample were 0.05% and -24.7‰, those of ID sample were 0.10% and -16.8‰, respectively.

The plate 05PO04: The %C and δ^{13}C values of SD sample were 0.11% and -26.8‰, those of ID sample were 0.13% and 16.3‰, respectively.

Pottery before carbon adsorption had low %C values both inner part and on the surface. Although reliability was low, a difference in δ^{13}C was observed between the surface and the inner part, having a similar value to the clay sample.

Black pottery samples

The pot 05PO05: The %C and δ^{13}C values of SD sample were 0.36% and -30.2‰, those of ID sample were 0.04% and -31.7‰, respectively. This black pottery had a low %C value inner part and a high %C value on surface, suggesting carbon adsorption was much higher on the surface sample than the inner

part sample. The $\delta^{13}C$ value of the surface sample had a similar value to the rice husk sample, however, the $\delta^{13}C$ value of the inner part sub-samples had similar values to the clay sample. It suggests that carbon from rice husk adsorbed only near the surface in this pot.

The pot 05PO06: The %C and $\delta^{13}C$ values of SD sample were 0.61% and -29.7‰, those of ID sample were 0.62% and -29.8‰, respectively. This pot showed that the %C and $\delta^{13}C$ values of surface and inner part were the same, showed the same value of the rice husk. These values suggest that the carbon from clay was removed by the AAA treatment while adsorbed carbon from rice husk still remained inner part.

The plate 05PO07: The %C and $\delta^{13}C$ values of SD sample were 1.17% and -27.6‰, those of ID sample were 0.19% and -18.0‰, respectively.

The plate 05PO08: The %C and $\delta^{13}C$ values of SD sample were 0.78% and -27.1‰, those of ID sample were 0.22% and -18.3‰, respectively. These 2 black plates had a similar high %C value on the surface and a low %C value inner part. The $\delta^{13}C$ values of the surface sample had similar values to the rice husk sample and the inner part samples had similar values to the clay sample. The %C values of the surface samples were higher than those of the pots. This might be caused by the coarse surface, since carbon might be adsorbed easier than on polished pots.

The %C and $\delta^{13}C$ values of sub-samples from the surface and the inner part were different between in each pottery sample, this maybe due to the firing condition.

4-3. ^{14}C measurement

The results of the AMS measurement were shown in Table 2. ^{14}C concentration (ratio of ^{14}C and ^{12}C) of the charred rice husk sample (05IS08) was 1.0815 ± 0.0024. Those of the 2 black pots (05PO05 and 05PO06) were 0.9126 ± 0.0025 and 1.0120 ± 0.0024. Those of the 2 black plates (05PO07 and 05PO08) were 1.0024 ± 0.0024 and 1.0248 ± 0.0025.

5. Discussion

It is supposed that the %C and $\delta^{13}C$ measurements of the samples which were collected from through the process of pottery manufacturing shown in this chapter must be the first data.

Based on experiments using modern pottery, the raw clay had about 0.5% of %C, after firing the pottery, the %C value from the surface of pottery before carbon adsorption decreased to below 0.1%. After adsorbing the rice husk carbon, the %C value of the black pottery surface became higher, from 0.4% to 1.2%. The $\delta^{13}C$ values also showed a similar trend. The $\delta^{13}C$ values of raw clay were about -13‰. After firing, the $\delta^{13}C$ values of pottery before carbon adsorption shifted remarkably to around -21‰ to -27‰, although they had a low reliability for the $\delta^{13}C$ values due to low %C values. After adsorbing rice husk carbon, $\delta^{13}C$ values of the black pottery surface were fixed from -27‰ to -29‰, similar to the $\delta^{13}C$ values of the rice husk (-28‰). These results suggest that carbon from clay was removed after firing, and carbon from rice husk has adsorbed onto the surface of the black pottery.

Considering the black pottery, the differences of the %C and $\delta^{13}C$ values among the pottery samples seemed to have been caused by the amount of adsorbed carbon, affected by both the temperature of pottery when it was covered with rice husk and the duration that the pottery was covered with rice husk. The pottery which had a black color on both the surface and the inner part might be well-completed black pottery. Considering the black pottery, the pot 05PO06 was perfectly completed as the black pottery. The pottery with a thin body, which has a black color on both the surface and the inner part, may be suitable for dating. The plate with a thick body may have a difficulty for the movement of carbon; removal of carbon in firing and adsorbing carbon from rice husk.

[14]C concentrations by the AMS measurements varied among the 4 black pottery samples, and that for the rice husk sample was higher than that for the pottery samples. One reason for this must be caused that these pottery samples were made during the documentation of the manufacturing process, each pottery piece was not made under the same condition. The timing of firing, the temperature and the duration of adsorbing carbon might be different for each piece of pottery due to the process of documentation. However, based on the examination of ^{14}C concentrations for recent dates (Moriuchi, *et al.*, 2000), the [14]C

concentrations in 2004, when the pottery samples were manufactured, was about 0.98. The fluctuation of ^{14}C concentrations became larger among modern (post-bomb) samples, by the remarkable increase of ^{14}C concentrations caused by nuclear testing. The 3 pottery samples 05PO06, 05PO07 and 05PO08 were similar to estimated concentration, and their variations were within the limits of fluctuation. Another pottery 05PO05 had a lower ^{14}C concentration, and Charred rice husk had a higher ^{14}C concentration. This pottery had lower %C values, supposed that adsorption of the carbon from rice husk was not enough. It suggests that such a slight adsorption on the pottery might cause a low ^{14}C concentration, that is, older age. Charred rice husk sample was residue after used for adsorption process, there might be an isotopic fractionation and heavier ^{14}C might remain in the rice husk. It must be clarified by measurement of rice husk sample before the adsorption process. These results suggest that careful selection of well-fired black and adsorbed pottery is useful for the ^{14}C dating.

Acknowledgments

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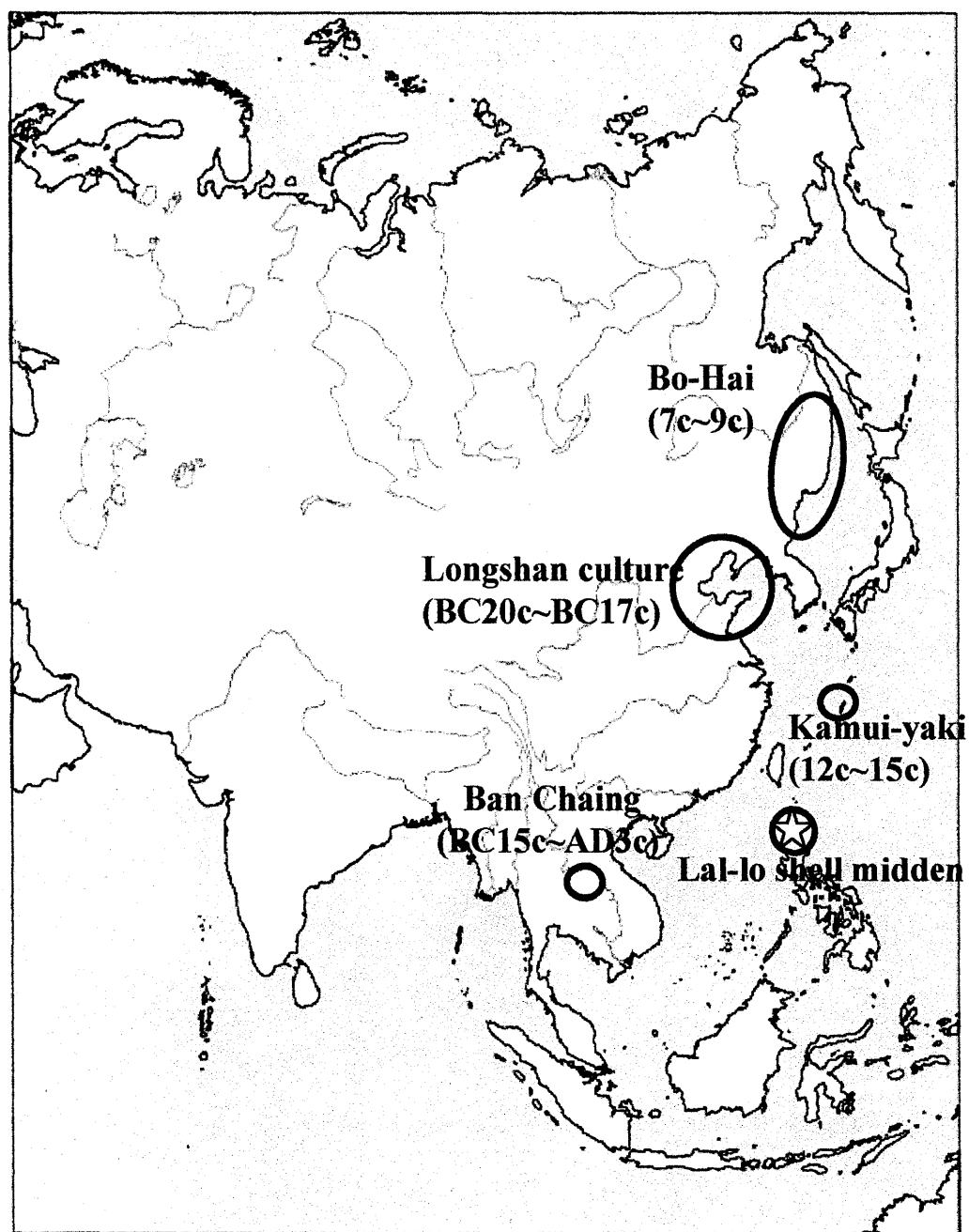
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Table 1 Result of carbon analysis for modern pottery and raw materials

lab No.	Name	before AAA				after AAA				recovery rate (%)
		weight	%C	Total C (mg)	$\delta^{13}C$	weight	%C	Total C (mg)	$\delta^{13}C$	
05PO01	SG	1274	0.03	0.34	-27.41	730	0.02	0.13	-30.12	57
	SD	1200	0.06	0.72	-25.47	684	0.02	0.15	-31.51	57
	IG	1550	0.05	0.78	-16.39	1046	0.07	0.78	-17.49	67
	ID	1096	0.09	0.95	-19.30	548	0.04	0.24	-19.13	50
05PO02	SG	1039	0.06	0.61	-24.34	670	0.02	0.16	-24.87	64
	SD	1054	0.08	0.85	-24.11	570	0.04	0.25	-26.10	54
	IG	1278	0.08	1.03	-17.14	862	0.09	0.81	-18.42	67
	ID	1148	0.09	1.05	-18.55	672	0.08	0.54	-18.85	59
05PO03	SG	1002	0.14	1.43	-23.59	696	0.08	0.55	-29.03	69
	SD	1148	0.09	0.98	-23.12	786	0.05	0.42	-24.69	68
	IG	1146	0.09	1.08	-14.83	771	0.10	0.77	-16.31	67
	ID	1057	0.09	0.92	-13.49	634	0.10	0.61	-16.83	60
05PO04	SG	1037	0.09	0.91	-21.31	683	0.04	0.31	-13.95	66
	SD	1020	0.13	1.28	-23.26	542	0.11	0.62	-26.82	53
	IG	1159	0.10	1.20	-12.46	818	0.14	1.12	-14.10	71
	ID	1025	0.14	1.45	-14.29	611	0.13	0.77	-16.29	60
05PO05	SG	1209	0.36	4.39	-29.31	763	0.29	2.20	-30.61	63
	SD	1009	0.50	5.09	-29.18	535	0.36	1.95	-30.25	53
	IG	1095	0.02	0.27	-17.29	674	0.03	0.17	-35.13	62
	ID	1186	0.06	0.77	-21.78	653	0.04	0.27	-31.71	55
05PO06	SG	1181	0.39	4.64	-27.30	795	0.39	3.09	-27.65	67
	SD	1094	0.55	6.01	-29.25	618	0.61	3.78	-29.75	56
	IG	1652	0.17	2.81	-17.14	1170	0.46	5.42	-27.69	71
	ID	1109	0.21	2.36	-18.41	637	0.62	3.98	-29.80	57
05PO07	SG	1071	0.64	6.88	-26.65	605	0.80	4.83	-27.50	56
	SD	1035	0.98	10.10	-27.66	504	1.17	5.87	-27.56	49
	IG	1022	0.14	1.44	-16.37	730	0.17	1.22	-16.88	71
	ID	1099	0.14	1.53	-15.37	763	0.19	1.43	-17.96	69
05PO08	SG	1008	0.66	6.64	-27.52	554	0.85	4.70	-28.17	55
	SD	1028	0.78	8.05	-27.15	538	1.00	5.36	-27.82	52
	IG	1318	0.19	2.45	-17.66	857	0.21	1.79	-17.22	65
	ID	1036	0.19	2.01	-19.22	714	0.22	1.58	-18.35	69
05IS01	clay	3060	0.50	15.26	-12.71	1502	0.10	1.52	-15.67	49
05IS02	sand	2019	0.01	0.28	-27.96	1801	0.01	0.09	-26.80	89
05IS03	mix	3032	0.45	13.62	-14.41	1750	0.04	0.77	-22.59	58
05IS04	red clay	494	1.06	5.22	-15.98	159	0.21	0.34	-16.97	32
05IS07	rice husk	298	44.83	133.60	-27.83	125	54.45	68.07	-28.28	42
05IS08	charred rice husk	500	42.59	212.97	-27.81	134	41.33	55.38	-27.79	27

Table 2 Result of ^{14}C concentration for modern black pottery and charred rice husk sample

lab No.		Name	before AAA				after AAA				recovery rate (%)	^{14}C concentration	analyze code (NUTA2-)
			weight	%C	Total C (mg)	$\delta^{13}C$	weight	%C	Total C (mg)	$\delta^{13}C$			
05PO01	SD	pottery before carbon adsorption	1200	0.06	0.72	-25.47	684	0.02	0.15	-31.51	57	-	-
05PO02	SD	pottery before carbon adsorption	1054	0.08	0.85	-24.11	570	0.04	0.25	-26.10	54	-	-
05PO03	SD	pottery before carbon adsorption	1148	0.09	0.98	-23.12	786	0.05	0.42	-24.69	68	-	-
05PO04	SD	pottery before carbon adsorption	1020	0.13	1.28	-23.26	542	0.11	0.62	-26.82	53	-	-
05PO05	SD	black pottery	1009	0.50	5.09	-29.18	535	0.36	1.95	-30.25	53	0.9126±0.0025	9872
05PO06	SD	black pottery	1094	0.55	6.01	-29.25	618	0.61	3.78	-29.75	56	1.0120±0.0024	9873
05PO07	SD	black pottery	1035	0.98	10.10	-27.66	504	1.17	5.87	-27.56	49	1.0024±0.0024	9874
05PO08	SD	black pottery	1028	0.78	8.05	-27.15	538	1.00	5.36	-27.82	52	1.0248±0.0025	9875
05IS01		clay	3060	0.50	15.26	-12.71	1502	0.10	1.52	-15.67	49	-	-
05IS02		sand	2019	0.01	0.28	-27.96	1801	0.01	0.09	-26.80	89	-	-
05IS03		mix	3032	0.45	13.62	-14.41	1750	0.04	0.77	-22.59	58	-	-
05IS04		red clay	494	1.06	5.22	-15.98	159	0.21	0.34	-16.97	32	-	-
05IS07		rice husk	298	44.83	133.60	-27.83	125	54.45	68.07	-28.28	42	-	-
05IS08		charred rice husk	500	42.59	212.97	-27.81	134	41.33	55.38	-27.79	27	1.0815±0.0024	9869

Fig. 1 Distribution of the black pottery in Asia

Putting red slip   Firing   Carbon adsorption   Completion

mix

clay

red clay

sand

Sand sample   Mix sample

Clay sample   Red clay sample

fuel

rice husk

Pottery before
carbon adsorption

Rice husk
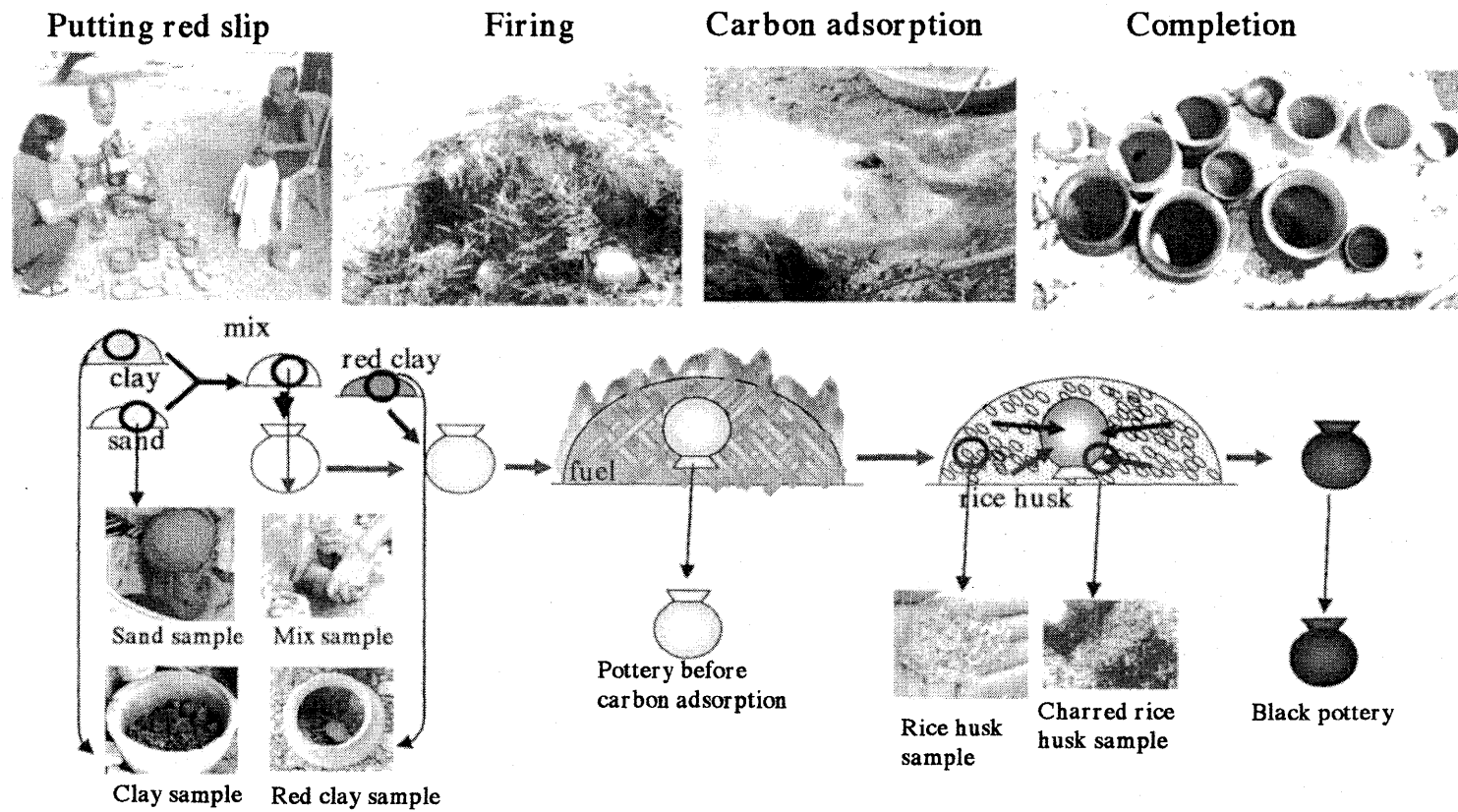sample

Charred rice
husk sample

Black pottery

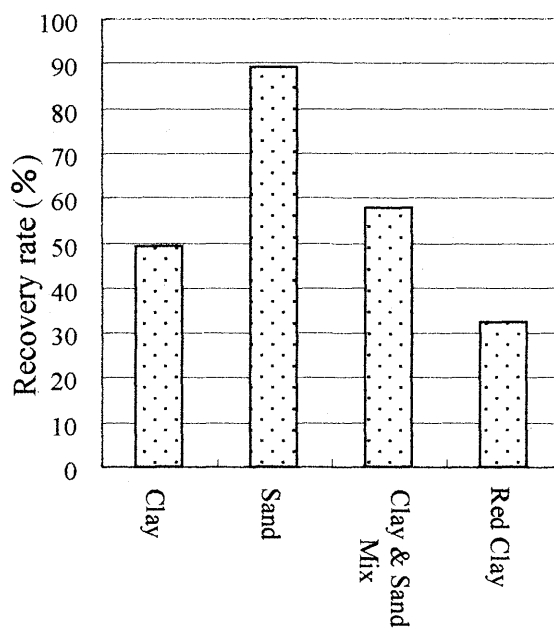Fig. 2 Manufacturing process of black pottery and samples for this study

05PO01 pot
Surface: red
Inner part: brown

05PO02 pot
Surface: red
Inner part : brown

05PO03 plate
Surface: red
Inner part : dark gray

05PO04 plate
Surface: red
Inner part : dark gray

05PO05 pot
Surface: black
Inner part : brown

05PO06 pot
Surface: black
Inner part : black

05PO07 plate
Surface: black
Inner part : dark gray

05PO08 plate
Surface: black
Inner part : dark gray
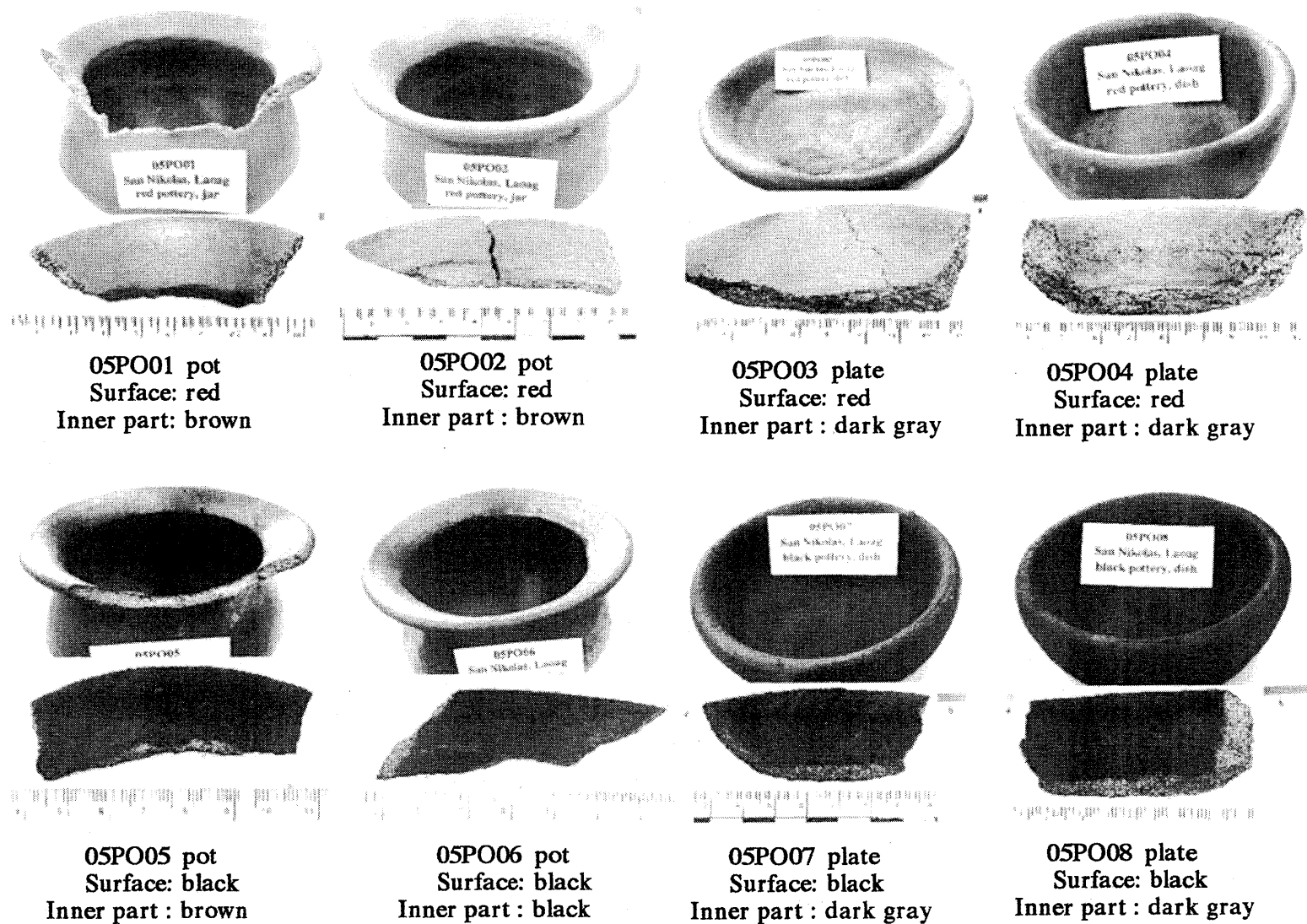
Fig. 3 pottery samples for analysis
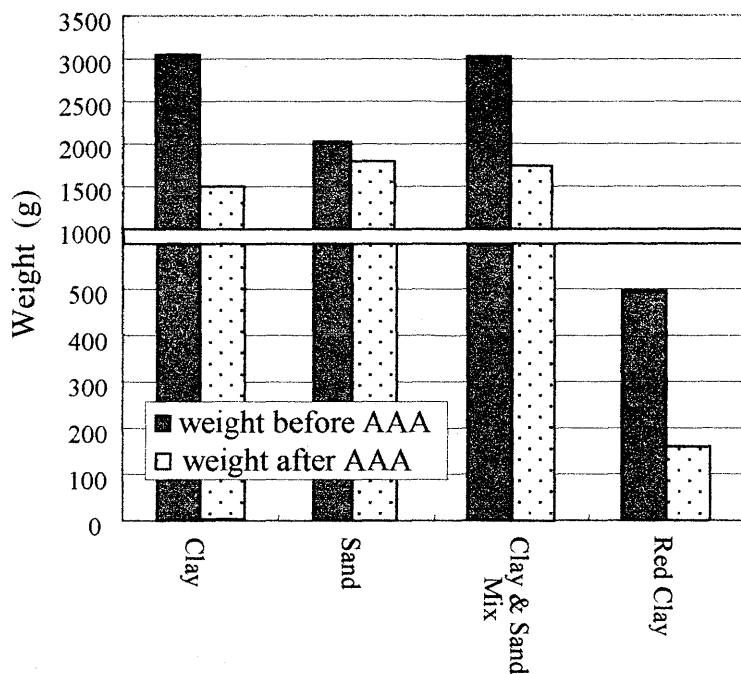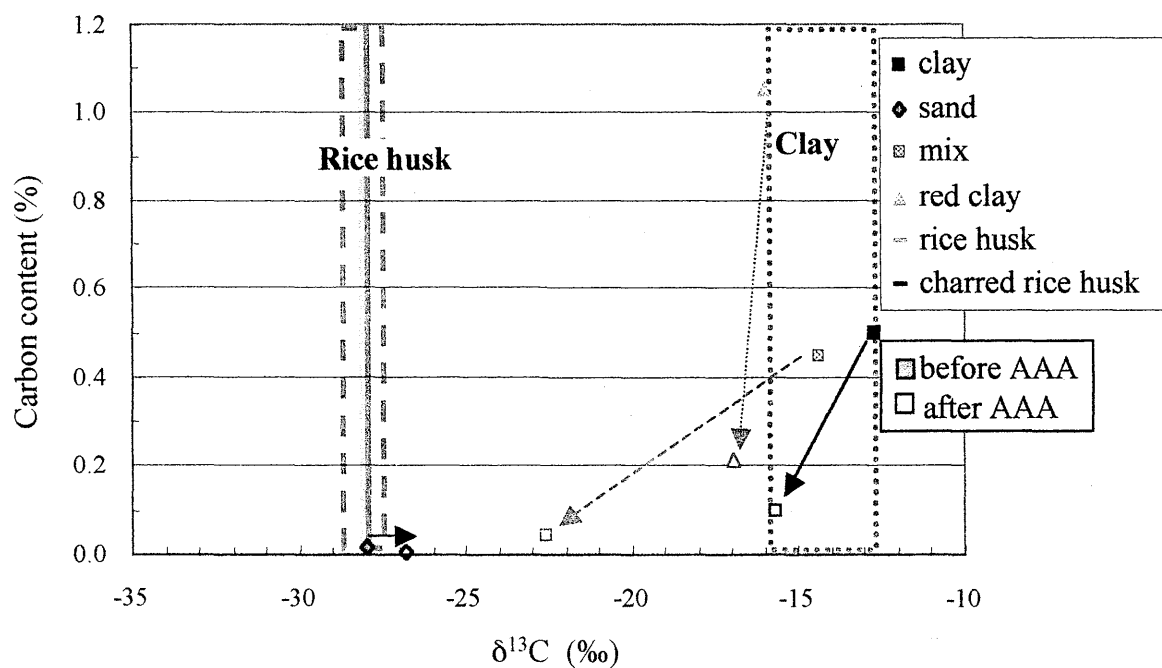
Fig. 4 Sample Weight of the raw materials

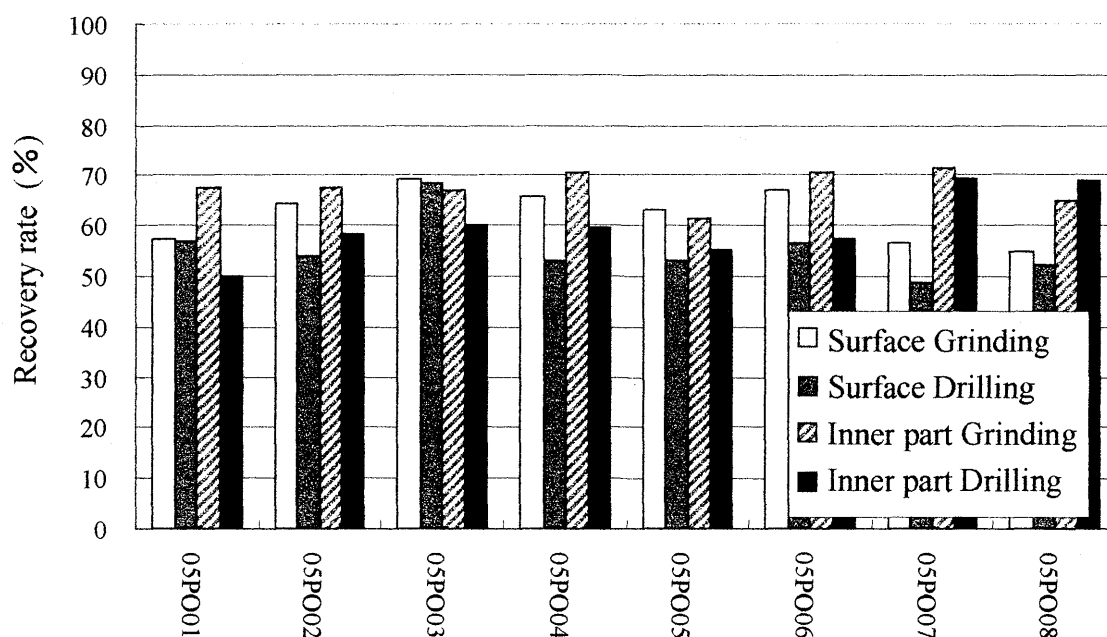**Fig. 5 recovery rate of the raw materials**
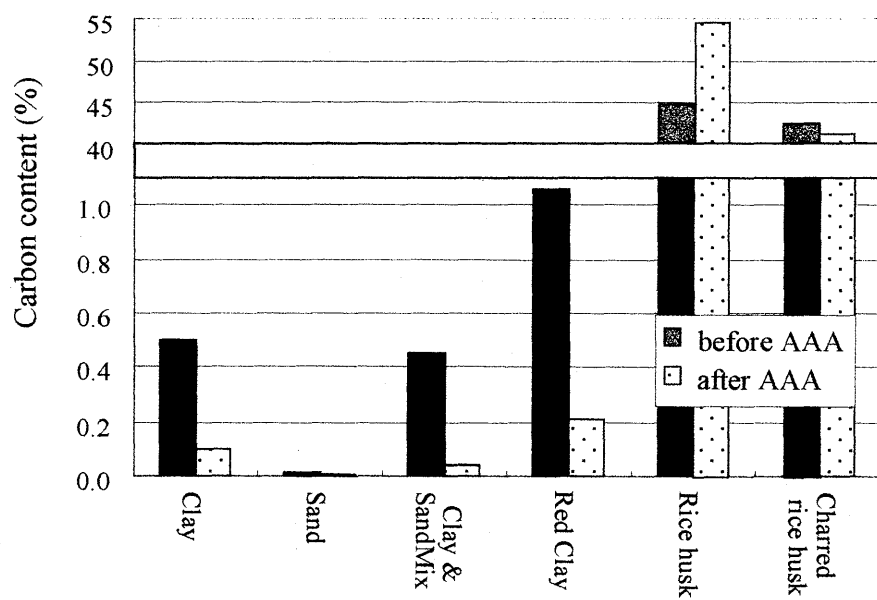
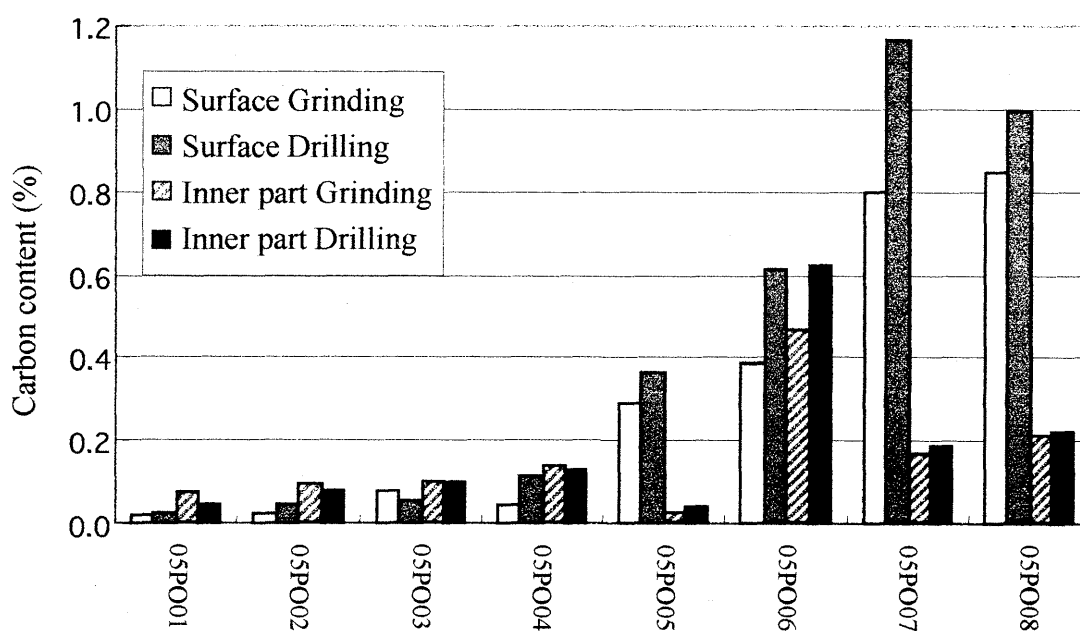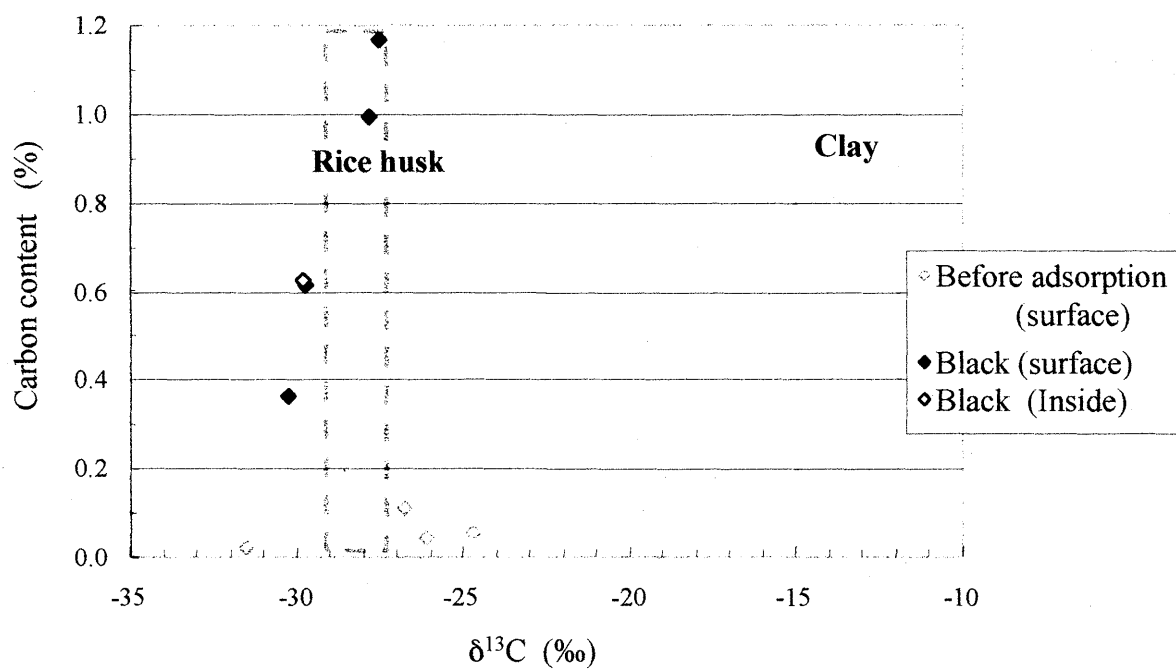**Fig. 6 Carbon content of the raw materials**

**Fig. 7 Carbon content and carbon isotope ratio
for raw material**

Fig. 8 Sample recovery of the pottery samples

Fig. 9 Carbon content for the pottery samples

Fig. 10 Carbon content and carbon isotope ratio for the pottery samples