Firing Technologies and Raw Materials of Typical Early and Middle Bronze Age Pottery from Kaman-Kalehöyük: A Statistical and Chemical Analysis

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ABSTRACT

This work deals with the technological study of Early Bronze Age (EBA) and Middle Bronze Age (MBA) pottery from Kaman-Kalehöyük, where different pottery production techniques are represented in these two periods. The purpose of this paper is to suggest reasons for the significant change in raw materials used in MBA pottery as compared to EBA pottery, by elucidating the distinct firing techniques used. A multi-analytical approach of statistical, petrographic, chemical and diffractometric analyses was adopted. Petrographic groups based on mineralogical characterization by thin section were defined using a polarizing microscope. Chemical compositions of clay fractions were analyzed using microprobe analyses. Firing temperature and firing conditions were determined using X-ray diffraction, SEM and XANES. The results contribute to an understanding of the development of the firing technologies of EBA and MBA pottery at Kaman-Kalehöyük.

1. INTRODUCTION

Since pottery production techniques are behaviorally significant to a culture, pottery can serve as a tool for understanding cultural processes. In this article, we attempt to understand cultural processes in the EBA and MBA at Kaman-Kalehöyük through a study of pottery production techniques. A series of statistical and chemical analyses was employed to study the raw materials and firing techniques of the EBA and MBA pottery from the site.

The pottery types of the EBA (2100-1930 B.C.) and MBA (1930-1750 B.C.) at Kaman-Kalehöyük are very different from each other in their production techniques. The most striking difference is that the majority of EBA pottery was hand made while MBA pottery was wheel-turned. EBA of Kaman-Kalehöyük is subdivided into two strata, IVa and IVb, by the proportion of the hand-made and wheel-turned wares. The earthenwares unearthed from stratum IVb are mostly hand-made while stratum IVa has an equal mixture of both hand-made and wheel-turned (Omura 2002). Middle Bronze Age started from Strata IIIc in Kaman-Kalehöyük and instead of hand-made, the wares of this period are mostly wheel-turned. In addition, features of their pastes, such as temper, color and texture, are different in the two periods. Fundamental questions thus arise as to why there was such a difference in the raw materials used; the change may reflect the innovation of new firing techniques or the interruption or diffusion of different cultures. If we are to make any attempt at understanding cultural change, we must look at the various technologies being employed, for technology embraces a culture's knowledge and behavior as well as material culture.

Detailed statistical analysis was carried out on a series of securely stratified pottery sherds from

EBA to early MBA layers at Kaman-Kalehöyük (Strata IVb, IVa and IIIc) in order to distinguish their technological features. The analysis was based on distinct characteristics of the pottery paste. The result is of great importance because it can provide a precise quantitative typological study of these pottery types, which can refine the chronological information (Matsumura 2001). Based on the groups identified in the statistical analysis, typical EBA and MBA wares were then selected for chemical analysis in order to understand their production technology. A multi-analytical approach was applied to this study. Our goal is to suggest why there was a significant change in the raw materials used in the typical EBA and MBA pottery types, by elucidating their firing techniques.

2. BACKGROUND

Studies of the styles and motifs of pottery from archaeological sites in Western and Central Anatolia has led to the hypothesis that a sort of cultural renaissance took place in the last phase of the Early Bronze Age ñ that is, the last century of the third millennium B.C. and the first century of the second millennium B.C. (Mellaart 1971). Mellaart (1971) and Özgüç (1963) also proposed that the cultural transition from the Early Bronze Age (EBA) to the Middle Bronze Age (MBA) was continuous and gradual throughout, rather than sudden. To support this hypothesis, Özgüç (1963) shows that the monochrome and polychrome Hittite pottery at the site of Kültepe developed from the last phase of the EBA. As an explanation of this gradual cultural progress, Özgüç (1963) suggested that the native culture of Anatolia was deeply influenced by the influx of Assyrian merchants from northern Mesopotamia who established a sophisticated trading system inside Anatolia and possessed a highly developed civilization.

At Kaman-Kalehöyük, a gradual change from EBA to MBA has also been identified through studying the ceramics of Strata IVb, IVa and IIIc. Five building levels, Rooms 287, 240, R212, R104 and 98, belonging to the Strata IVb and IVa of EBA have been identified in north trench, Sector III (Fig. 1). The Alişar III type painted pottery excavated from Rooms 240, 212, 104 and 98 indicate that Stratum IVa of Kaman-Kalehöyük is contemporary with Strata III and IV of Kültepe-Karum and Stratum 5M of Alişar, which represent the so-called transition period of EBA to MBA (Omura 2000). Since Room 287 is dominated by hand made pottery, it is believed to belong to the true EBA cultural layer.

3. STRATIGRAPHICAL ANALYSIS

1286 rim sherds from six different building levels of Sector III, North trench, were selected for a series of classification and statistical analyses. These building

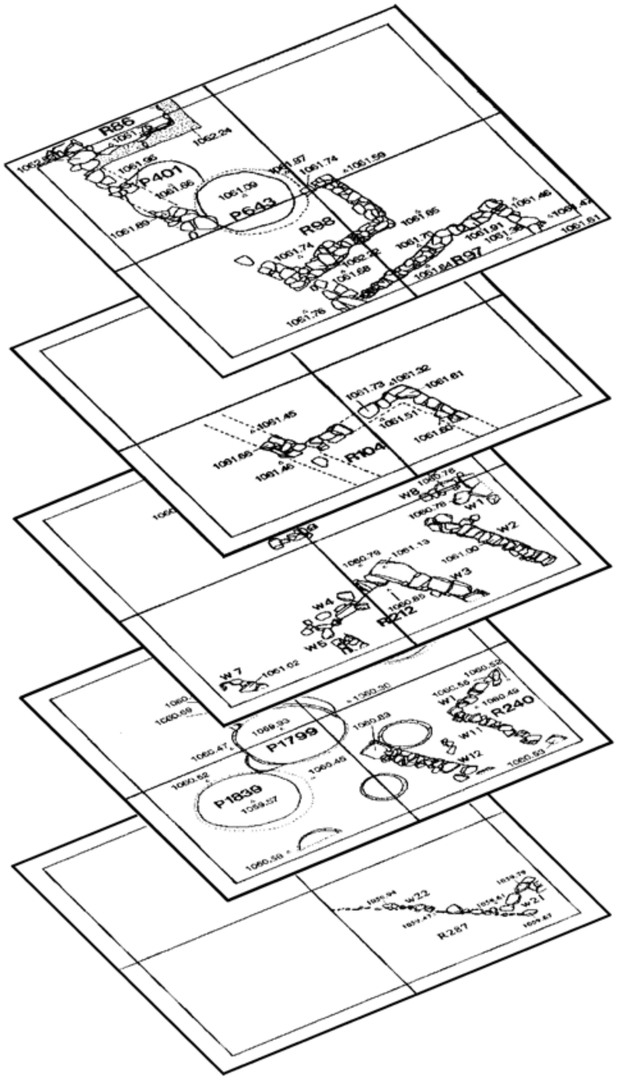

Fig. 1 Five building levels of EBA, North trench, Sector III. From top, R98, R104, R212, R240 and R287.

levels include the oldest EBA building level at Kaman-Kalehöyük (Stratum IVb), in Room 287, four building levels of the EBA-MBA transition period (Stratum IVa), in Rooms 240, 212, 104, and 98 and the oldest building level of the MBA (Stratum IIIc), in Room 86. In this discussion, the levels/periods are referred to as R287, R240, R212, R104, R98 and R86.

In the first step of classification, all of the sherds were divided into two groups, cooking pots and regular wares. There were 364 cooking pot sherds and 922 regular ware sherds (Tab. 1).

Table 1 Proportions of the cooking pot ware and regular ware from the six building levels.

Room Number	Cooking Pot	Regular Ware	Total sherds number
R.86 (IIIc)	70	149	219
R.98 (IVa)	44	140	184
R.104 (IVa)	86	222	308
R.212 (IVa)	36	95	131
R.240 (IVa)	68	137	205
R.287 (IVb)	60	179	239
Total	364	922	1286

Since the color of cooking pot sherds may be affected by secondary firing, such as by cooking fires, these sherd types usually do not provide much information in the study of firing techniques. Therefore, they were excluded from further investigation in this study. The remaining 922 regular wares were then classified into three categories based on differences in their temper, paste color and slip. This classification was developed to understand changes in the technological features over time.

Temper: There are mainly two kinds of inclusions observed in the studied sherds: straw and sand. Although we can confirm that straw must have been added deliberately to the clay as temper, we cannot judge through naked eye observation that sand was deliberately added as temper since it is common to find sand or rock fragments occurring naturally in some clays. Thus, instead of using the term 'sand-tempered' sherds, we prefer using 'sandy' sherds to describe those with sandy inclusions. Fig. 2 shows the proportions of the two types. The straw-tempered sherds are found in greater relative quantities in the lower (earlier) building levels, decreasing in the upper (later) building levels. In contrast, the sandy sherds increase in quantity over time. This indicates that EBA potters had the tradition of adding straw as temper to the paste, and this tradition did not continue into the MBA.

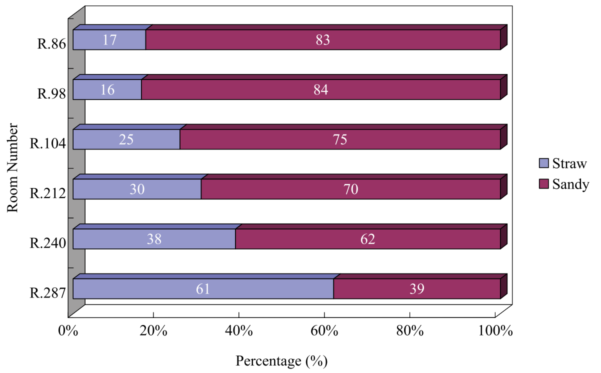

Fig. 2 Proportions of straw-tempered and sandy sherds in the six building levels.

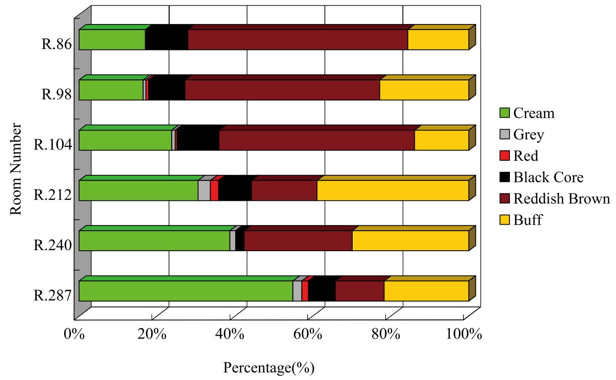

Fig. 3 Proportions of paste colors in the six building levels.

Paste color: The primary determinants of pottery color are clay composition and firing conditions -- atmosphere, temperature and duration of firing. Common clays usually consist of iron compounds, and an oxidation firing usually gives this type of clay a reddish color while a reduction firing gives gray or black. In some firing, the oxidation or reduction effects can be weak and at times may be nearly neutral; this usually gives a buff or yellowish color to the end product.

Three main colors are observed in most of the EBA and MBA sherds from Kaman-Kalehöyük: reddish brown, cream, and buff. Other colors such as black and gray also exist but in far fewer quantities (Fig. 3). This reveals that instead of reduction firing, oxidation firing techniques were generally used throughout the EBA and MBA periods. Fig. 3 shows clearly that from

R287 to R86, the proportion of the reddish brown sherds increased while the cream sherds decreased. This indicates that either different firing techniques or different raw materials were being used between these two periods. The proportion of the buff colored paste is lower in R86, R98 and R104 than in R212, R240 and R287. In this study, buff ware refers to the wares which were fired under a weak oxidizing atmosphere or neutrally fired. In other words, they might have been produced with a lack of control in the firing atmosphere. From this point of view, sophisticated firing techniques with good control of the firing atmosphere seem to have developed after the chronology sequence of R104.

Slip: As for surface decoration, a layer of red slip is usually applied to the EBA and MBA pottery. However, pottery with red slip is much more common in MBA pottery if compare with EBA pottery. Besides, the texture of the slip of MBA pottery is much better than those of the EBA pottery. The proportions of the slipped ware of both periods are shown in Fig. 4 and Fig. 5.

Combining the characteristics of temper, paste color, and slip, the statistical analysis shows that there are mainly nine different ware types defined in the six building levels. See Table 2.

Table 2 Types of ware (paste and color combinations) identified from the six building levels.

Sandy	Plant Tempered
1) Plain reddish brown	6) Plain cream body
2) Reddish brown body with red slip	7) Cream body with red slip
3) Reddish brown body with red slip and black core	8) Buff core with red slip
4) Buff body with red slip	9) Plain buff body
5) Plain buff body	10) Others

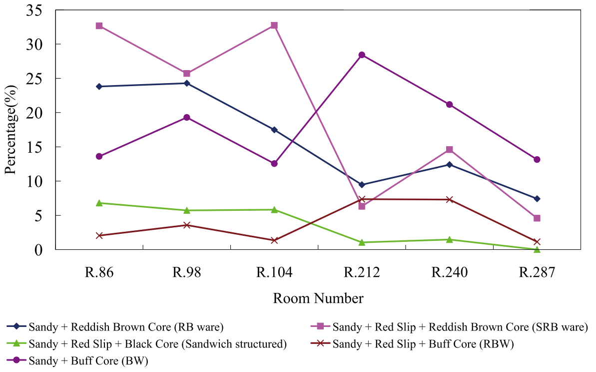

Fig. 4 Percentages of sandy wares in the six building levels.

Fig. 4 shows the proportions of all types of sandy ware. R86, R98 and R104 have the highest proportions of the Red Slipped Reddish Brown ware (SRB) and Plain Reddish Brown ware (PRB). These wares are wheel made and are typical MBA wares. The proportions of the SRB and PRB wares show a significant increase in R104 compared to R212. Some of the SRB wares display a sandwich structure with a black core and reddish brown margins. The proportion of this type of sandwich-structured SRB ware also increases between R212 and R104. Buff ware (BW) and Red Slipped Buff ware (RBW) show the opposite trend, becoming less dominant in R104 compared to R212. These distinct changes in the proportions of the different wares between R212 and R104 indicate that the MBA of Kaman-Kalehöyük might have started from the chronology sequence of R104.

Figure 5 shows the proportions of all straw-tempered wares. R287 has the highest proportion of PC ware which increases dramatically from R240. In addition, the straw-tempered Plain Cream (PC) ware and Red Slipped Cream ware (SC) tend to increase gradually from R240 to R212 and occupy a lower proportion from R104 to R86. Most of the SC and PC wares were hand made. These results reveal that the transition period from MBA to EBA might be somewhere between the chronology sequence of R240 to R212 and the real EBA culture at Kaman-Kalehöyük might have started from R287.

In conclusion, the statistical analysis indicates three phases of chronological change based on the variations in the late EBA and early MBA pottery from Kaman-Kalehöyük:

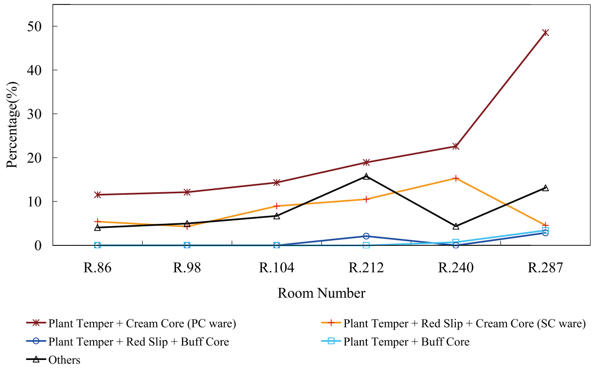

Fig. 5 Percentages of plant-tempered wares in the six building levels.

1st phase: R287, the period in which the PC ware is dominant (49%), and SRB and RB wares make up a very small proportion (<11%). This might be the starting of the real EBA cultural period at Kaman-Kalehöyük when the typical PC ware was widely in use.

2nd phase: From R240 to R212, a sharp increase in SRB and PRB wares (<22%) and a gradual decrease in PC and SC wares (>28%) was observed. This might be the EBA-MBA transition period.

3rd phase: From R104 to R86, the SRB and PRB wares occupy the highest proportions (>40%) among all kinds of wares. In contrast, PC and SC wares are very small in proportion (<17%). This is the period in which the MBA SRB and PRB wares first come into wide use at Kaman-Kalehöyük.

The results of the statistical analysis raise two questions. First, what caused MBA potters to change their raw materials and firing techniques from those of the EBA period. Second, how do these changes reflect the pottery production cultures of the EBA and MBA at Kaman-Kalehöyük. To answer these questions, further scientific examination which can provide a wide range of information and powerful evidence on the raw materials and firing techniques of pottery is needed.

4. CHEMICAL ANALYSIS

It is thought that ancient potters chose appropriate raw materials for specific types of firing techniques (Barlow and Idziak 1989). In order to suggest reasons for the significiant change in the raw materials used from the EBA to the MBA, we have to look into the firing properties of the raw materials and determine the firing techniques used. However, little is known about the production technologies and provenances of EBA

Table 3 List of the samples analyzed.
Notation: YNo.=artifact number; PL=provisional layer

No.	YNo.	Sector Area	Grid Name	PL	Stratum	Types
III-1	07000261	N-VII	XXXII-54	(78)	IIIc	SRB
III-2	92002879	N-III	XL-54	(52)	IIIc	SRB
III-3	92002866	N-III	XL-54	(58)	IIIc	SRB
III-4	07000275	N-VII	XXXII-54	(81)	IIIc	SRB
III-5	07000276	N-VII	XXXII-54	(81)	IIIc	SRB
III-6	97003405	N-III	XLI-55	(74) ⓑ	IVa	SRB
III-7	92002862	N-III	XLI-54	(59)	IVa	SRB
III-8	92002861	N-III	XLI-54	(59)	IVa	SRB
III-9	02000852	N-XXVI	XLV-52	(61)	IIIb	SRB
III-10	02000853	N-XXVI	XLV-52	(61)	IIIb	SRB
III-11	02000854	N-XXIX	XLVI-50	㊿	IIIb	SRB
III-12	92002860	N-III	XLI-54	(59)	IVa	SRB
IV-1	00001332	N-III	XLI-55	(90)	IVb	PC
IV-2	96002594	N-III	XLI-55	(61)	IVa	SC
IV-3	98001443	N-III	XLI-55	(75) ⓑ	IVa	SC
IV-4	99001640	N-III	XL-55	(75)	IVa	SC
IV-5	00001338	N-III	XL-55	(90)	IVb	PC
IV-6	99001639	N-III	XL-55	(76)	IVa	SC
IV-7	97003398	N-III	XLI-55	(72)	IVa	SC
IV-8	97003400	N-III	XLI-55	(73) ⓐ	IVa	SC
IV-9	02000792	N-VII	XXXIX 54	P2566	IVa	SC
IV-10	02000804	N-VII	XXXIX-54	(77)	IVa	SC
IV-11	02000786	N-VII	XXXVIII-55	(78)	IVa	SC
IV-12	02000787	N-VII	XXXVIII-55	(78)	IVa	SC

and MBA pottery except for the initial work of chemical analysis by Kanda and Nakai (1998; 1999), which clearly reveals that different kinds of raw materials were used in these two periods.

For this study, the typical EBA and MBA wares, PC, SC and SRB wares, were subjected to a combination of technological analyses in order to understand the relationship between the characteristics of the raw materials and their production technologies.

4.1 Samples and methods

Cross-sectional thin sections of 24 securely stratified PC, SC and SRB samples were first prepared for chemical composition analysis (Tab. 3). Of those, 6 SC, 2 PC and 6 SRB sherds were then selected for petrographic thin section analysis under a polarizing microscope. All of the thin sections were coated with a thin graphite layer before being examined with a JSM-7000F scanning electron microscope (SEM) using back-scattered electrons (BSE), with an acceleration voltage of 15kV. Micromorphology and vitrification microstructures of the fresh fractures were also examined by SEM using secondary electrons with an acceleration voltage of 5kV.

Chemical composition of the paste was examined under an energy-dispersive X-ray fluorescence spectrometer (EDS) attached to the SEM, using Si (Li) detector, for the determination of Si, Al, Fe, Ca, Mg, Na, K, Ti, P and S components. The ZAF procedure was used for correcting the matrix effects and the results were given as weight percentages of oxides normalized to 100% total.

Mineral phases of the samples were determined by X-ray powder diffraction (XRD) analysis on a RINT 2500 diffractometer using CuKα radiation (λ=1.54184), with an operating current of 100mA and an accelerating voltage of 50kV.

Finally, in order to evaluate the original firing atmosphere of the sherds, the chemical states of Fe in the samples were analyzed by synchrotron radiation-induced X-ray absorption near-edge structure (XANES) analysis. The Fe K-edge XANES spectra were measured in the fluorescence mode using the BL-12C of the Photon Factory, High Energy Accelerator Research Organization, Tsukuba, Japan. Monochromatized 2.5GeV SR X-rays, obtained from a Si (111) double-crystal monochromator, were used as an excitation source throughout the measurement. Two reference minerals with different oxidation states of iron, hematite ($Fe_2^{III}O_3$) and magnetite ($Fe_3^{II,III}O_4$), were measured. The intensity of the Fe K X-ray was measured with a Lytle-type fluorescence detector (Lytle et al. 1984).

4.2 Petrographic and chemical analysis

Petrographic examination of the samples shows primarily two different fabrics: a very calcareous paste with coarse-grained temper (the PC and SC sherds), and a fine-textured, low-calcareous paste (the SRB sherds).

The PC and SC sherds have cream colored calcareous paste which is coarser in texture (Fig. 6a). Organic temper (straw) is present in high proportion and the majority of the temper is fully oxidized, resulting in long, narrow, interconnecting pores. Occasional coarse quartz, feldspars (Fig. 7a, d), granitic rock fragments (Fig. 7b), argillaceous rock fragments (Fig. 7e) and micritic limestone (Fig. 7f) are present. Under BSE examination, these wares show the characteristics of very calcareous clay fabrics surrounded by clots of calcium carbonates with clear boundaries. Some of the calcium carbonates show signs of decomposition, leaving many small pores in the fabric (Fig. 6c).

The low-calcareous SRB ware shows different features. This ware has a reddish brown to orange paste which is finer in texture (Fig. 6b). Its body consists primarily of very fine to medium particles dispersed within silty particles. The inclusions in the paste consist of feldspar, abundant mica (Fig. 7c), amphiboles and quartz grains (Fig. 6d). The inclusions are mostly rounded and well sorted, indicating that they might have originated from a sedimentary source, such as a river bed. No organic temper is observed and the paste is solid with few pores.

Chemical compositions of the clay fractions of the pastes were analyzed using an electron microprobe. Their results are shown in Tab. 4. The use of microprobe analysis has advantages over other techniques in that large, coarse inclusions which affect the accuracy of quantitative analysis can be avoided since morphology observation can be done simultaneously with microprobe analysis (Freestone 1982).

One of the most peculiar features of PC and SC

Table 4 Results of SEM-EDS analyses on the clay matrix of the SC, PC and SRB wares.

Samples	Na_2O	MgO	Al_2O_3	SiO_2	P_2O_5	SO_2	KO_2	CaO	TiO_2	FeO
III-1	0.82	3.27	20.39	60.76	0.43	0.04	3.34	2.06	0.54	8.34
III-2	0.86	3.13	21.63	58.63	0.34	0.19	3.52	3.52	0.64	7.54
III-3	0.61	3.76	20.32	58.81	0.27	0.23	3.62	3.95	0.41	8.03
III-4	0.84	3.75	21.23	58.98	0.20	0.06	2.75	4.04	0.63	7.52
III-5	0.58	3.27	19.56	60.42	1.24	0.08	3.01	4.37	0.47	7.01
III-6	0.99	3.45	18.74	59.14	0.47	0.18	2.23	8.08	0.81	6.42
III-7	1.16	3.11	19.26	59.19	0.49	0.55	2.58	6.43	0.62	6.62
III-8	0.68	3.56	19.55	57.54	0.33	0.20	3.10	7.71	0.63	6.70
III-9	1.53	3.88	22.97	61.48	0.67	0.07	1.81	3.55	0.69	3.34
III-10	0.56	3.17	20.19	60.12	0.67	0.18	4.09	2.99	0.63	7.40
III-11	0.65	3.29	16.74	53.69	0.56	0.10	2.57	15.47	0.55	6.40
III-12	0.53	4.09	21.80	58.76	0.43	0.07	2.68	3.34	0.56	7.71
IV-1	0.59	3.07	15.63	49.87	0.55	0.90	3.52	20.28	0.47	4.94
IV-2	0.90	3.21	14.95	40.69	0.32	0.68	2.53	32.75	0.18	3.81
IV-3	0.53	2.18	15.12	47.84	0.89	0.42	3.87	24.04	0.62	4.50
IV-4	0.55	2.37	14.42	45.07	0.60	0.93	4.89	29.20	0.40	3.24
IV-5	0.78	2.25	12.99	40.32	0.57	0.36	1.91	36.05	0.35	4.43
IV-6	0.44	1.52	17.89	45.34	0.44	0.56	4.52	25.23	0.63	3.43
IV-7	1.06	2.70	14.04	43.12	0.44	0.55	2.79	29.10	0.63	5.10
IV-8	0.71	2.05	17.89	46.99	0.53	0.24	2.48	23.85	0.47	4.80
IV-9	1.21	4.15	17.55	50.56	1.33	0.74	2.52	20.44	0.24	1.25
IV-10	0.38	2.44	14.97	46.29	0.49	0.78	3.50	26.37	0.57	4.19
IV-11	0.85	2.82	14.07	45.08	0.41	0.32	3.27	29.36	0.49	3.33
IV-12	0.69	3.12	16.22	48.86	0.79	0.47	2.87	23.27	0.21	3.51

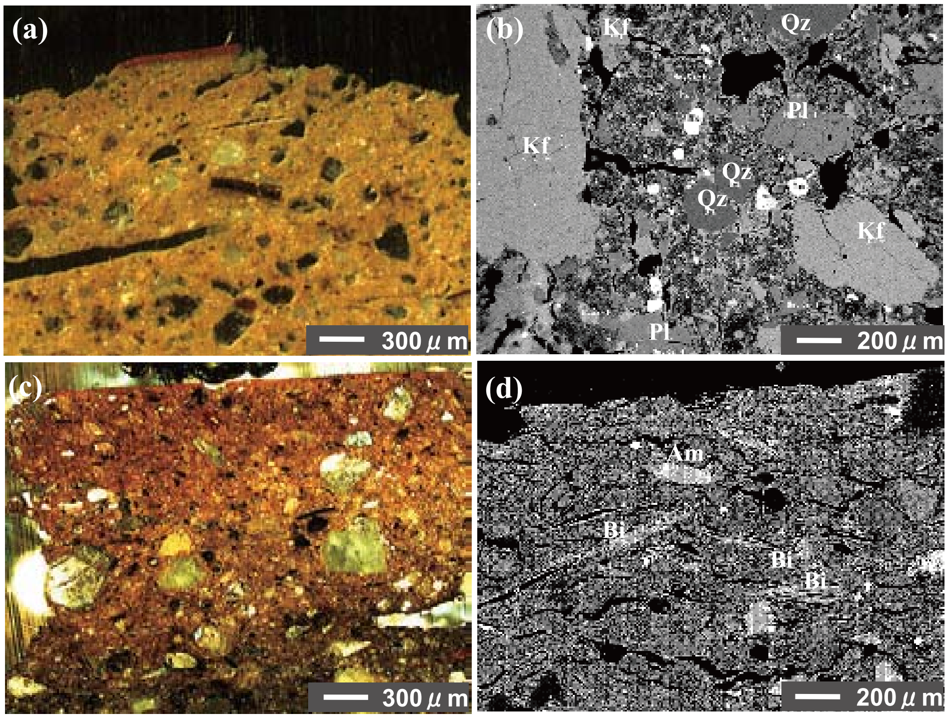

Fig. 6 (a),(c) Photomicrographs of SC and SRB ware; (b),(d) SEM-BSE micrographs of SC and SRB ware; Qz, quartz; Kf, alkali feldspar; Bi, biotite; Ca, calcium carbonate; Pl, plagioclase feldspar; Am, amphibole; He, hematite.

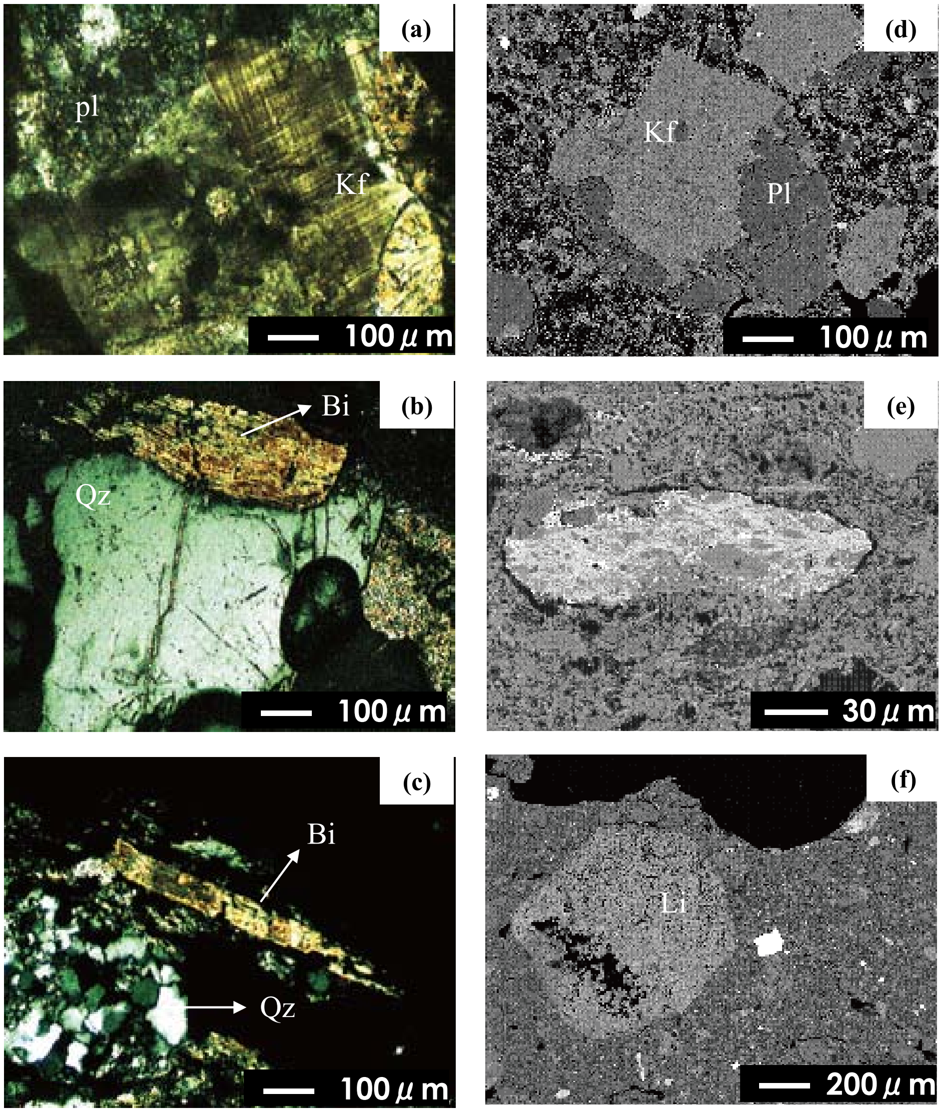

Fig. 7 Polarized micrograph of (a) Feldspatic rock fragment, tartan twinning of Alkali feldspar was shown on the left; (b) granite rock fragment, composed by of mega quartz, biotite and feldspar crystal (c) biotite with polycrystalline quartz; SEM-BSE image of (d) plagioclase + K-feldspar igneous rock fragments; (e) Ferric argillaceous rock fragment; (f) micritic limestone; Kf, alkali feldspar; Qz, quartz; Bi, biotite; Li, limestone.

ware is apparent from Fig. 8. Semi-quantitative EDS analysis showed that the CaO concentration in the EBA PC and SC ware is significantly higher than in the SRB ware; most of the PC and SC sherds showed CaO concentrations greater than 20%. This suggests that marl clay might have been used in the manufacture of EBA pottery at Kaman-Kalehöyük. In contrast, the CaO concentration in the MBA SRB ware is below 10%. This agrees well with the result of ICP-AES analysis carried out by Kanda and Nakai (1998).

Also seen in Fig. 8, the lime-poor SRB ware has a higher FeO concentration; this results in reddish brown color paste. In contrast, SC and PC wares are lower in FeO concentration, resulting in a creamy or whitish color.

The concentrations of SiO_2, Al_2O_3 and MgO in the SRB ware are higher than in the PC and SC wares (Tab. 4). Fig. 9 shows the concentrations of MgO and SiO_2 in both wares. A careful observation of thin sections under the polarizing microscope shows that high concentrations of these elements were reflected in the mineralogical composition of the paste of the SRB ware, which is rich in fine-grained quartz, amphibole, feldspar and biotite (Fig. 10b, 6d).

In contrast, PC and SC wares show lower concentrations of SiO_2, Al_2O_3 and MgO, and an extremely high concentration of CaO, suggesting that their origin might differ from SRB ware. Thin sections observed under the polarizing microscope also show the PC and SC clay fractions are composed of numerous fine crystals of micritic calcite with high birefringence (Fig. 10a). Although very coarse, sharp angled granitic rock fragments and alkaline feldspar minerals are present in the pastes of PC and SC ware, these inclusions are poorly sorted and they do not correlate well with the chemical composition of the clay fractions (Fig. 6b). Thus, we assume that they might be temper added deliberately into the marl clay by the potter.

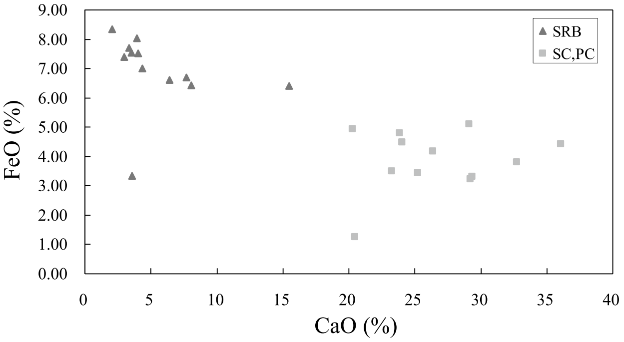

Fig. 8 SEM-EDS analysis, plot of FeO versus CaO.

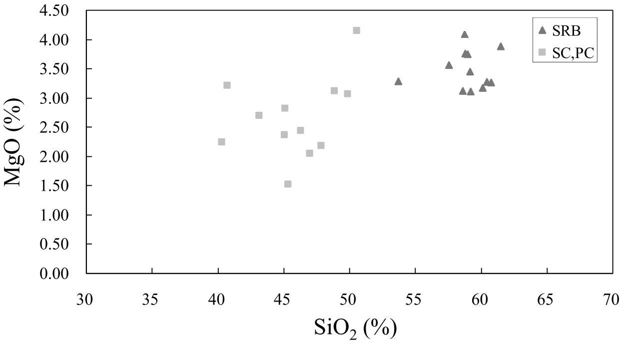

Fig. 9 SEM-EDS analysis, plot of MgO versus SiO_2.

With the combination of petrographic thin section analysis and EDS analysis, it is clear that the EBA and MBA wares show great distinctions in both the mineralogy and chemical composition of their pastes. This reveals that different kinds of raw materials, calcareous clay (CaO > 16%) and low-calcareous clay (CaO < 16 %), were selectively used by the potters of these two periods.

4.3 Estimation of original firing temperature

Firing temperatures of 19 samples were evaluated by two techniques: X-ray diffraction (XRD) (Maritan *et al*. 2006; Rice 1987) and the comparison of vitrification stages as seen in the fresh fracture under SEM (Maniatis and Tite 1981). By re-examining the pottery after refiring at known temperatures, we were able to follow the development of vitrification. Using the SEM data together with XRD data, it was possible to establish the approximate original firing temperatures of each sherd.

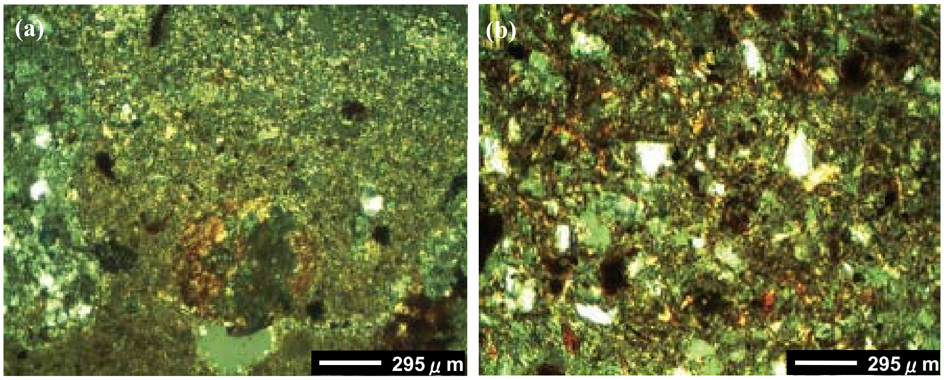

Fig. 10 Polarized micrographs of the clay matrix of (a) SC ware, which shows fine crystals of micritic calcite with high birefringence and (b) SRB ware, with fine-grained quartz, amphibole, feldspar and biotite.

The as-received SC and SRB sherds were first refired at known temperatures from 600 °C to 1050 °C, at 100 °C intervals, except the last interval, which was 150 °C, in an oxidation atmosphere in an electric kiln. The changes in their mineral phases with the increases in temperature were studied using XRD analysis. High-temperature mineral phases began to form in the clay at approximately 800 °C. These high-temperature phases then became the indicator for determining the maximum original firing temperatures of the wares.

One representative sample each of calcareous SC ware (IV-9) and low-calcareous SRB ware (III-11) are used here to illustrate the experiment and measurement procedures we used to determine their firing temperatures.

The as-received calcareous SC sherd, IV-9 (CaO = 20.44%), shows a great amount of calcite, with no high-temperature phases present (Fig. 11). This calcite decreased as the firing temperature was increased, and fully decomposed at the firing temperature of 800 °C (Fig. 11). Above 800 °C, high-temperature Ca-silicates such as gehlenite and anorthite were formed. Since high-temperature phases were absent in the as-received SC sherd, it must have been fired below 800 °C.

The phases present in the as-received low-calcareous SRB sherd, III-11 (CaO = 15.47%), include only small amounts of calcite (Fig. 12). Other phases, such as mica, feldspar mineral groups and amphibole are also present. During refiring, high-temperature phases such as diopside began to form at 900 °C. The formation of diopside instead of gehlenite is due to the low concentration of calcium and high concentration of magnesium in the paste (Assal *et al.* 1999). Meanwhile, the decomposition of mica phases between 900 °C and 1050 °C was also observed. These results suggest that the original firing temperature of the SRB sherd was between 800°C and 900°C, higher than that of the SC sherd.

A summary of the XRD results of 19 representative samples at their as-received state are presented in Tab. 5. Since EBA samples IV-2, IV-7 and IV-8 show high-temperature Ca-silicates (gehlenite), they were fired above 800 °C. Since the decomposition of calcium carbonates usually occur after 800 °C, other EBA samples which show a great amount of calcium carbonates were fired below 800 °C. Most of the EBA samples show similar mineral phases, with the present of mica and low amount or non calcium carbonates, except samples III -6 and III-8. This result reveals that a majority of EBA pottery were fired between 800°C and 900°C.

Fresh fractures of SC and SRB wares were examined by SEM to observe the internal morphologies that developed at different refiring temperatures. This allowed us to evaluate the degree of vitrification at each stage (Tite *et al.* 1982).

Figure 13 shows that the morphology of the refired SC sherd, IV-9, remained unchanged until the temperature reached 800 °C, where the initial vitrification stage began (Fig. 13b). At this stage, slight buckling and rounding of the edges of the clay was observed. Since the original SC ware shows no

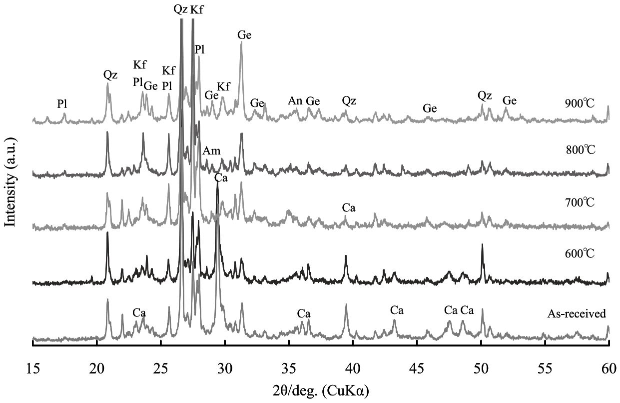

Fig. 11 XRD diffraction patterns of as-received SC ware (IV-9) and its refired sherds. Qz: quartz, Ca: calcite, Kf: alkali feldspar, Pl: plagioclase, Am: amphibole; Ge: gehlenite.

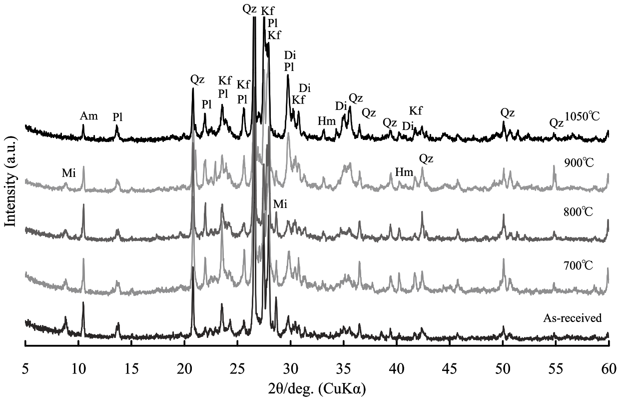

Fig. 12 XRD diffraction patterns of as-received SRB ware (III-11) and its refired sherds. Qz: quartz, Ca: calcite, Kf: alkali feldspar, Pl: plagioclase, Mi: mica, Am: amphibole; Di: diopside, Hm: hematite.

vitrification, the firing temperature of the SC ware must have been approximately between 700 °C and 800 °C (Fig. 13a). Above 800 °C, extensive vitrification occurred. The development of vitrification was inhibited by the formation of crystalline phases with high melting temperatures, that is, calcium alumino-silicates, gehlenite; a completely disturbed structure with essentially no smooth areas of glass was observed (Fig. 13d).

As for the low-calcareous SRB sherd, III-11, its

Table 5 Mineral phases of 19 samples, revealed by XRD.

Samples	Qz	Ca	Kf	Pl	Mi	Am	Ge	Px
III-1	xxx	x	x	xx	x	xx		x
III-2	xxx	x	x	x	x	x		x
III-3	xxx	x	x	x	x	x		
III-4	xxx		x	xx	x	x		
III-5	xxx		xx	x	x	x		x
III-6	xxx		x	x		xx		x
III-7	xxx	x	x	x	x	x		
III-8	xxx	x	x	x	x			
III-11	xxx		x	x	x	x		x
III-12	xxx	(x-)	xx	x	x	xx		x
IV-1	xxx	xxx	xx	xx		x		
IV-2	xx	xxx	xx	xx		x	(x-)	
IV-3	xx	xxx	xx	xx		xx		
IV-4	xx	xxx	x	x				
IV-5	xxx	xxx	x	x				
IV-6	xxx	xxx	x	x				
IV-7	xxx	x	x	x			xx	
IV-8	xxx	x	x	x	(x-)		xx	
IV-9	xx	xxx	xx	xx		x		

Notation: Qz: quartz, Ca: calcite, Kf: alkali feldspar, Pl: plagioclase, Mi: mica, Am: amphibole; Ge: gehlenite, Px: pyroxene; xxx=major phase, xx=present in large quantities, x=present, (x-)=trace.

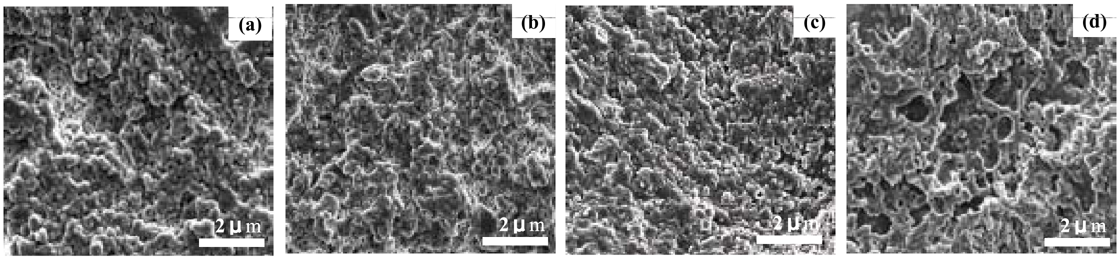

Fig. 13 SEM photomicrographs of the SC ware, IV-9, (a) as received; and its development of vitrification stages at (b) 700 °C, (c) 800 °C, and (d) 900 °C.

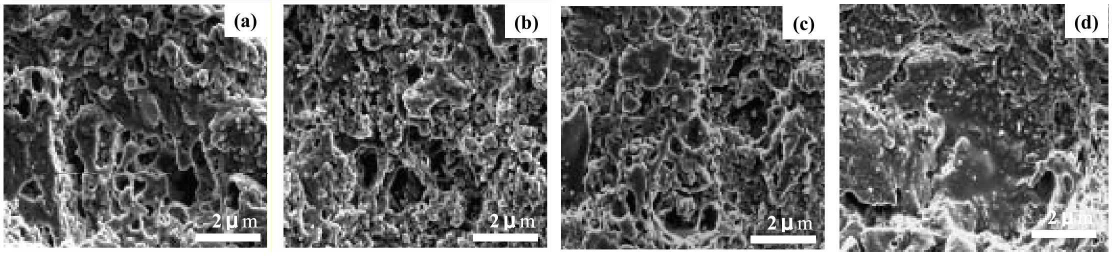

Fig. 14 SEM photomicrographs of the SRB ware, III-11, (a) as received; and its development of vitrification stages at (b) 800 °C, (c) 900 °C, and (d) 1050 °C.

morphology remain unchanged until the temperature reached 1050 °C, where a continuous vitrification stage began and a wide area of glassy phase was observed (Fig. 14). The as-received low-calcareous SRB sherd, III-11, shows extensive vitrification (Fig. 14a), indicating a firing temperature between 800 °C to 1050 °C. However, since the XRD result shows that mica is present originally in this sample, and the decomposition of mica phases occur between 900 °C and 1050 °C, the firing temperature of sample III-11 is thus estimated approximately between 800 °C and 900 °C.

There are two EBA SC samples, IV-7 and IV-8, which appear to have been originally fired over the temperature of 900 °C. SEM examination of the as-received sherds shows continuous vitrification of the fabric, accompanied by isolated bloating pores, as illustrated in Figure 15a. The SEM examination confirms the XRD results showing that the high-temperature phase gehlenite is present in each sample (Tab. 5).

A summary of the results of SEM examination of the 19 samples is presented in Tab. 6. The majority of SRB sherds displayed extensive vitrification (Fig.

Table 6 Results of firing temperatures determined by SEM on 19 samples.

Samples	CaO (wt%)	Body vitrification	Firing temperature (˚C)
III- 1	2.06	V	800- 900
III- 2	3.52	V	800- 900
III- 3	3.95	V	800- 900
III- 4	4.04	V	800- 900
III- 5	4.37	V	800- 900
III- 6	8.08	CV	900-1000
III- 7	6.43	V	800- 900
III- 8	7.71	V	800- 900
III-11	15.47	V	800- 900
III-12	3.34	V	800- 900
IV- 1	20.28	NV	700- 800
IV- 2	32.75	IV	800- 900
IV- 3	24.04	NV	700- 800
IV- 4	29.20	IV	800- 900
IV- 5	36.05	NV	700- 800
IV- 6	25.23	NV	700- 800
IV- 7	29.10	CV	900-1000
IV- 8	23.85	CV	900-1000
IV- 9	20.44	NV	700- 800

Notation: NV: non vitrification, IV: initial vitrification, V: extensive vitrification, CV: continuous vitrification.

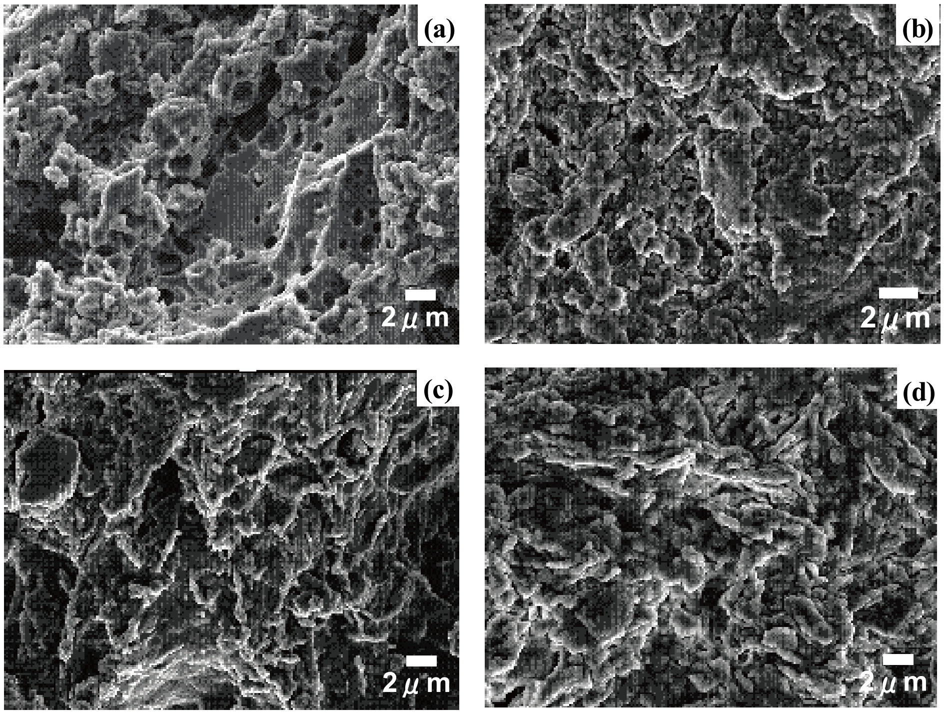

Fig. 15 SEM photomicrographs of (a) high temperature SC (IV-8) sherd, showing continuous vitrification with small bloating pores; (b) low temperature SC (IV-4) sherd, showing non-vitrification; (c) high temperature SRB (III-12) sherd, showing extensive vitrification; and (d) high temperature SRB (III-1) sherd, showing extensive vitrification.

15c, 15d) while SC sherds displayed varying stages of vitrification (Fig. 15a, 15b). This illustrates that in the MBA, uniform firing temperatures above 800 °C can be assumed, while in the EBA, the firing temperatures were not uniform. It seems probable that the lack of uniformity in the EBA pottery production might be due to chance positioning of the pottery during firing or the use of less sophisticated firing techniques such as open firing. Since firing temperatures are strongly associated with firing techniques, different kinds of firing techniques are assumed to have been used by EBA and MBA potters at Kaman-Kalehöyük.

4.4 Estimation of firing atmosphere

A firing atmosphere that gives color to pottery is attributed to the chemical form of iron compounds in the paste. Reddish color is usually given by an oxidation firing, which causes Fe to form trivalent iron (Fe^{3+}); a reduction firing will cause the formation of divalent iron (Fe^{2+}) and turn the color of the paste gray or black. However, in dealing with pottery which exhibits cream or buff paste, such as the PC and SC wares in this study, the causes of their color will be complex due to the fact that a cream color can be attributed to several different firing techniques (Matson 1971). In addition, the firing conditions that produced the sandwich-structured SRB ware displaying a black core and reddish brown margins remains unknown. According to Rye (1981), this type of structure is due to firing in reducing conditions with an oxidizing cooling stage, or to firing organic matter-rich clays in oxidizing conditions. In either of these cases, it is difficult to judge the firing condition by naked eye observation of the paste colors.

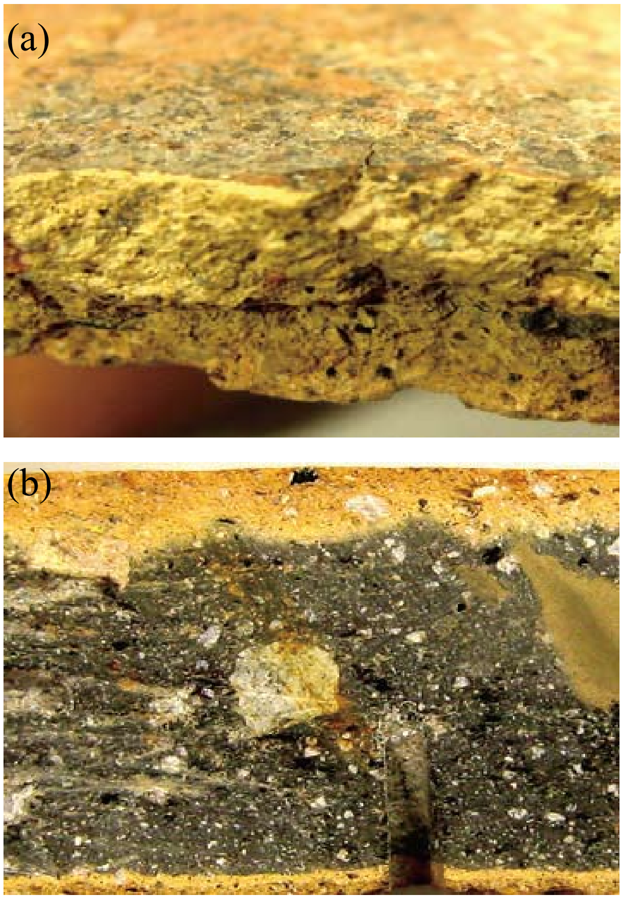

Fig. 16 Cross-sectional photographs of (a) SC and (b) sandwich-structured SRB ware for XANES analysis.

Through measuring the Fe K-XANES spectra, we are able to obtain basic information on the chemical form of iron in these samples and thus determine their firing atmosphere (Matsunaga and Nakai 2004). In the present study, one representative sample each of SC ware (IV-9) and sandwich-structured SRB ware (III-11) were analyzed, along with two reference samples with different Fe chemical forms, hematite (α-Fe_2O_3) and magnetite (Fe_3O_4). The sherd samples were measured at two spots, the surface and the core. Cross-sectional photographs of SC and black-cored SRB ware are shown in Fig. 16. It is known that the absorption edge shifts to a higher energy level as the valence state of the absorber atom in a sample increases (Calas, Manceau and Petiau 1988). Thus, since the valence of iron in hematite (Fe^{3+}) is higher than in magnetite (Fe^{2+}, Fe^{3+}), its absorption edge shifts to a higher energy level.

The Fe K-XANES spectra of the surface and core of the SC ware are similar to those of the hematite reference sample, all of which contain trivalent iron (Fig. 17). This reveals that the SC sample was fired in an oxidation atmosphere.

The Fe K-XANES spectra of the surface and core of the sandwich-structured SRB ware are different from each other. The spectrum of the surface is close to that of hematite, indicating an oxidation firing, while the black core's Fe K-edge shifts to much lower energy level than magnetite, indicating a reduction firing (Fig. 18). This suggests that the sandwich-structured SRB

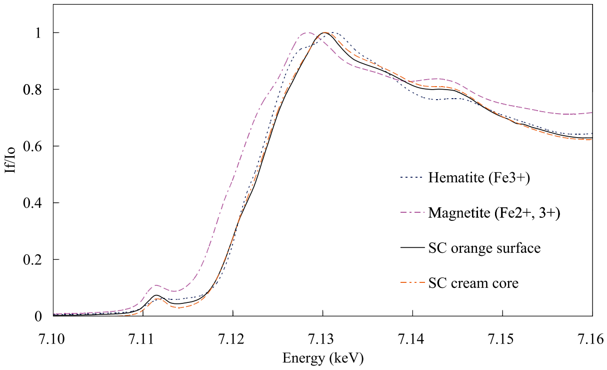

Fig. 17 Fe K-XANES spectra of the SC (IV-9) ware.

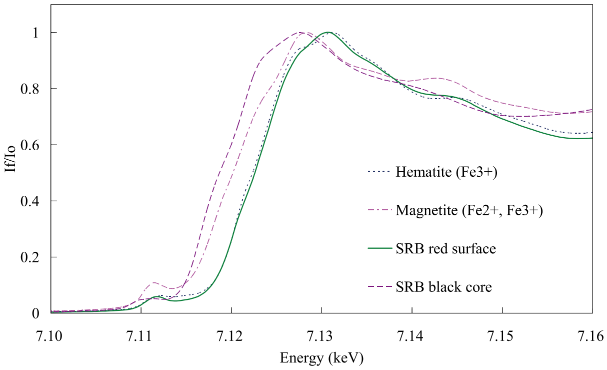

Fig. 18 Fe K-XANES spectra of the SRB (III-11) ware.

ware might have been fired under incomplete reduction conditions with an oxidizing cooling stage. Alternatively, it was also reported that this type of sandwich-structured ware can usually be obtained by organic-rich clay fired under kiln firing conditions with a low heating rate and long residence time (Maritan 2006). Further detailed mineralogical analysis of this type of sandwich-structured ware is needed to enrich our understanding of its firing technique.

In addition to XANES analysis, refiring the sherds in a full oxidation atmosphere at temperatures between 750 °C and 900 °C can also provide important insights on original firing atmosphere, through observation of any color changes. The SC sherd was refired at 900 °C in oxidation atmosphere. The color of the paste after firing remained the same, cream in color (Fig. 19a). From this result, we can thus understand that the EBA SC or PC wares are fired in a complete oxidation atmosphere.

After refiring the sandwich-structured SRB sherd at 900 °C, the color of its black core turned red (Fig. 19b). This reveals that the sherd was originally fired in an incomplete oxidation atmosphere, where the combination of temperature, time and draft was probably inadequate for full oxidation. The chance positioning of the pottery during firing and the thickness of the paste could be the causes of insufficient oxidation. There is also a possibility that this type of ware was fired in a reducing atmosphere followed by an oxidizing cooling stage. Since fully reduction-fired wares are not found much in MBA contexts at Kaman-Kalehöyük, it is possible that MBA potters did not produce reduction-fired wares intentionally, so the former hypothesis is thought to be more probable than the latter.

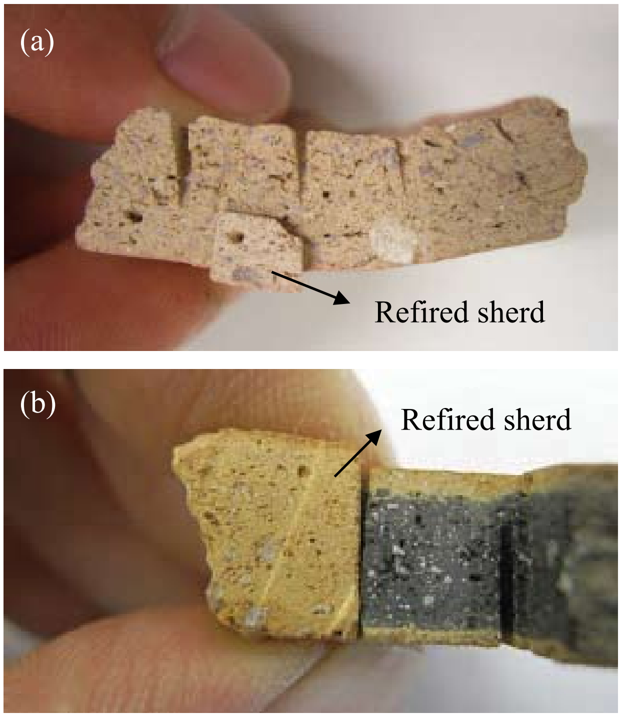

Fig. 19 Color changes in (a) SC and (b) SRB sherds, after refiring at 900 °C.

5. DISCUSSIONS

In order to understand the relationship between the selective use of raw materials and the firing techniques used, one must follow the basic concept that ceramic materials must be of good quality in order to assure a sturdy product that will not crack and break during firing. If we compare the firing techniques used by EBA and MBA potters, it indeed points out that a different kind of firing technique was adapted to the pottery production of both periods. This may have been one of the most important driving forces for the change in pottery raw materials since certain raw materials are appropriate for specific kinds of firing techniques.

The firing temperatures of most of the hand made calcareous SC and PC ware lack uniformity, with most of the sherds fired below 800 °C. Many of these sherds show black spots or fireclouds on their surfaces. This feature suggests that this ware might have been fired in open firing; this technique is always short and uneven, usually with a higher heating rate and generally with a lower temperature. A paste filled with coarse temper would be appropriate for this type of firing because it has the ability to resist thermal shock caused by rapid firing or cooling and this prevents the vessels from cracking (Arnold 1985). However, instead of resistance to thermal shock, coarse temper might have been added to the calcareous paste in order to improve its workability, since calcareous clay is usually sticky due to its hygroscopic properties. The concentration of Ca in the SC and PC wares is extremely high, especially compared to SRB or RB ware.

It is also seen that if the firing temperature is too high, over 800 °C, calcite in the calcareous clay decomposes on firing and forms lime (CaO). Lime is very hygroscopic; it will pick up moisture from the air, forming quicklime and releasing heat. This is accompanied by volume expansion, which sets up stresses in the surrounding body, causing vessels to crack and spall. This is most noticeable when the lime particles in the clay are comparatively large (Rye 1981). For these reasons, the use of calcareous clay seems appropriate to the low-temperature open firing technique. Nevertheless, the raw materials used by EBA potters would be deemed of poor quality by potters today.

With the development of the wheel thrown technique, a switch to more uniform-textured, low-calcareous, fine clays appears to have occurred in the MBA, when vessels were able to be fired at higher temperatures, mostly over 800 °C. The potters of this period appear to have known how to choose a better quality of clay for ceramic production. From the uniformity of the firing technique and the achievement of a higher firing temperature, the kiln firing technique is assumed to have been developed in the MBA. However, it is also important to note that so far, no kilns have been found at Kaman-Kalehöyük.

6. CONCLUSION

We have shown that the differences in the raw materials and the firing techniques of the typical Early Bronze Age and Middle Bronze Age pottery at Kaman-Kalehöyük can be quantitatively revealed by a series of statistical and chemical analyses. We focused our study on the changes seen in the pottery types excavated from the six building levels of the late EBA to the early MBA.

The statistical data show that cultural processes could be identified through studying the relationship between the materials and technological features of the pottery, since technology is a unique system incorporating material resources, tools, and techniques, related to social behaviors and meanings. Three chronological phases were identified: 1st phase, the Early Bronze Age culture, with PC ware in highest proportion; 2nd phase, the transition period from EBA to MBA, with PC and SC ware gradually decreased while RB and SRB increased; and 3rd phase, the Middle Bronze Age culture, with SRB and RB ware in highest proportion. This result indicates that the cultural transition from EBA to MBA was gradual at Kaman-Kalehöyük.

The petrographic and microprobe analysis showed that there were two different kinds of raw materials used for pottery production in the EBA and MBA. The reconstruction of firing temperatures and atmosphere also show that different kinds of firing techniques were employed for pottery production in these two different periods. These changes in the ceramic production technology are of great importance as they illustrate that from EBA to MBA, there was an evolution of pottery production technology occurring between these two periods at Kaman-Kalehöyük. In this case, evolution in technical terms obviously led to improvements in raw materials in the ceramic production process. Cultural aspects, such as the inflow of Assyrian traders, however, could have played a role by introducing new pottery production techniques to the Anatolian inhabitants. Both technical and sociocultural factors, simultaneously, maybe the driving forces for change in EBA and MBA ceramic production techniques at Kaman-Kalehöyük.

Further analysis of other elements, such as the environmental context of the relationship of EBA and MBA potters to their environment, where they collected the raw materials and a more detailed stylistic study of forms should be done in the future in order to understand more about the historical and social changes and material cultural evolution in the EBA and MBA periods of Kaman-Kalehöyük. These studies will enrich our discussion of the underlying reasons for the changes or choices in raw materials.

ACKNOWLEDGEMENTS

We would like to thank Dr. S. Omura for providing us the samples and for his support for this study. We are also grateful to Ms. C. Kumazaki for her help during the laboratory experiment.

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