Research on the white pottery, stamped hard pottery, and proto-porcelains unearthed at Erlitou

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Abstract

The earliest proto-porcelains in northern China known to date have been unearthed at Erlitou Site in Yanshi City, Henan Province. This archaeological evidence is very important for the exploration of the origin of protoporcelain in China. As research subjects, the white pottery, stamped hard pottery and proto-porcelain samples unearthed at Erlitou Site are analyzed by systematical experimental methods to determine their chemical compositions, baking temperatures, phase compositions and microstructures, and then the raw materials and baking technology of these samples are discussed. In addition, the characteristics and origin of proto-porcelain from Erlitou Site were also studied.

Keywords: Erlitou Site; firing technology; multivariate statistical analysis; pottery raw materials

Brief introduction of the issue

Erlitou Site in Yanshi City, Henan Province is an



Figure 1 Two dimensional correspondence analysis diagram of the main chemical compositions of the pottery body samples from Erlitou.

important site in Chinese archaeology. A large number of ceramic artifacts have been excavated from Erlitou, most of which were for daily use. They mainly include gray pottery and gray black pottery. Meanwhile, artifacts made of white pottery, stamped hard pottery and proto-porcelain also have been found at Erlitou. Importantly, these protoporcelain samples date to the Phase II of Erlitou Culture and are the earliest proto-porcelain wares known to date in northern China, making them invaluable materials for the research on the origin of proto-porcelains. Under the support of the "the Origin of Chinese Civilization Project," we carried out systematic scientific studies on the chemical composition, firing temperatures, phase composition, and microstructures of white pottery, stamped hard pottery and proto-porcelain samples from Erlitou and obtained some significant results.

The samples

24 samples were selected and provided by the Erlitou Archaeological Team of the Institute of Archaeology, Chinese Academy of Social Sciences. The contextual information for these samples is listed in Table 1.

Experimental results and discussion

1. Energy dispersive X-ray fluoresce analysis (EDXRF).

The chemical compositions of the samples were determined by EDXRF (EDAX Eagle-III μ Probe, USA). The maximum power of the X-Ray tube was 40W, the diameter of X-ray focus was 300 μ m and the Si (Li) detector was used to determine the characteristic X-ray. The EDXRF analyzer has the advantages of being non-destructive, high-precision, and capable of multi-element analysis, making it especially suitable for nondestructive analysis of rare samples. The test results are listed in Table 2 and Table 3. Table 2 contains EDXRF results for the bodies of the clay matrix. Table 3 contains EDXRF results from exposure to the exterior glazes still present on four samples. Note that Table 2 does not contain the body composition of sample ELT-77.

2. Chemical composition characteristics of the raw materials.

The main chemical composition data in Table 2 were analyzed using multivariate statistical analysis. Meanwhile, the chemical composition data of some common pottery types including gray and black pottery from Erlitou (Wu et al. 2007) were also included to show the unique characteristics of the white pottery, stamped hard pottery, and proto-porcelain. Figure 1 shows the two dimensional correspondence analysis diagram of the main chemical compositions of the pottery samples. The total

| No. | Code | Vessel shape | Ceramic type | Period | Excavation unit | Description |
|-----|--------|---------------------------|----------------------|---|-----------------|---|
| 1 | ELT-56 | Gui-tureen | White pottery | Between Erlitou Phase I and Phase II | 2002YLVT12 | Yellow |
| 2 | ELT-57 | Gui-tureen | White pottery | Erlitou Phase II | 2003YLVT32 | Yellow |
| 3 | ELT-58 | Zun-jar | White pottery | Erlitou Phase II | 2003YLVT32 | Reddish outside and gray inside |
| 4 | ELT-60 | He-pitcher | White pottery | Erlitou Phase II | 2001YLVT6 | Yellow |
| 5 | ELT-61 | Jue-cup | White pottery | Erlitou Phase II | 2001YLVT6 | Gray |
| 6 | ELT-62 | Zun-jar | White pottery | Erlitou Phase III | 2001YLVT6 | Reddish |
| 7 | ELT-63 | He-pitcher | White pottery | Erlitou Phase IV | 2004YLVT84 | Gray |
| 8 | ELT-65 | He-pitcher | White pottery | Erlitou Phase II | 2003YLVT32 | White |
| 9 | ELT-66 | Zun-jar | White pottery | Erlitou Phase II | 2003YLVT32 | Reddish |
| 10 | ELT-67 | He-pitcher | White pottery | Erlitou Phase III | 2001YLVT5 | Gray |
| 11 | ELT-68 | Jue-cup | White pottery | Erlitou Phase III | 2000YLIIIT2 | Gray |
| 12 | ELT-70 | Jue-cup | White pottery | Erlitou Phase IV | 2004YLVT85 | Gray |
| 13 | ELT-72 | _ | Proto-porcelain | Erlitou Phase II | 2002YLVT20 | Green glazed outside, stamped, thin gray body |
| 14 | ELT-73 | Jue-cup | White pottery | Erlitou Phase III | 2000YLIIIT2 | Gray |
| 15 | ELT-74 | Zun-jar | White pottery | Erlitou Phase II | 2003YLVT32 | Reddish |
| 16 | ELT-75 | Zun-jar | White pottery | Erlitou Phase II | 2001YLVT5 | Reddish |
| 17 | ELT-76 | He-pitcher | White pottery | Erlitou Phase II | 2003YLVT32 | White |
| 18 | ELT-77 | Conical object | White pottery | Erlitou Phase II | 2002YLVT15 | White and red pigment on surface |
| 19 | ELT-78 | He-pitcher | Proto-porcelain | Erlitou Phase IV | 2002YLVT23 | Brown glazed outside, stamped, thin gray body |
| 20 | ELT-79 | He-pitcher | Proto-porcelain | Erlitou Phase II | 2003YLVT35 | Brown glazed outside and thick body |
| 21 | ELT-80 | He-pitcher | White pottery | Erlitou Phase II | 2001YLVT4 | Gray |
| 22 | ELT-81 | <i>He</i> -pitcher or pot | Stamped hard pottery | Erlitou Phase II | 2001YLVT5 | Thin gray body, stamped |
| 23 | ELT-82 | He-pitcher | Proto-porcelain | Erlitou Phase II | 2002YLVT14 | Green glazed outside and thick body |
| 24 | ELT-83 | Zun-jar | White pottery | Erlitou Phase II | 2003YLVT32 | White and hard |

| Table 1 | Experimental | samples | from | Erlitou. |
|---------|--------------|---------|------|----------|
|---------|--------------|---------|------|----------|

cumulative variance of two factors was 80.5%.

Figure 1 reveals that the body compositions of the white pottery, stamped hard pottery and proto-porcelain were obviously different from common pottery. One main difference is that the flux contents (Na₂O, MgO, K₂O, CaO and Fe₂O₃) in common pottery bodies were higher than in white pottery, stamped hard pottery and proto-porcelain.

In Table 2, the total flux contents in white pottery, stamped hard pottery, and proto-porcelain bodies were all less than 10%, which was the main reason that they were made of materials classified as porcelain clay whereas common pottery used fusible clay. Porcelain clay can bear higher firing temperatures. The chemical composition data in Table 2 also show that the white pottery bodies at Erlitou

| No. | Code | Na ₂ O | MgO | Al ₂ O ₃ | SiO ₂ | K ₂ O | CaO | TiO ₂ | Fe ₂ O ₃ |
|-----|----------|-------------------|------|--------------------------------|------------------|------------------|------|------------------|--------------------------------|
| 1 | ELT-56-b | 0.59 | 1.08 | 21.25 | 66.35 | 2.71 | 4.02 | 0.52 | 2.48 |
| 2 | ELT-57-b | 0.30 | 0.43 | 24.66 | 68.97 | 0.90 | 0.58 | 0.86 | 2.30 |
| 3 | ELT-58-b | 0.27 | 0.91 | 26.68 | 63.55 | 2.53 | 0.70 | 0.72 | 3.65 |
| 4 | ELT-60-b | 0.33 | 0.67 | 27.86 | 63.61 | 1.71 | 0.98 | 0.71 | 3.13 |
| 5 | ELT-61-b | 0.40 | 0.92 | 24.33 | 65.61 | 2.82 | 0.57 | 0.59 | 3.75 |
| 6 | ELT-62-b | 0.61 | 1.04 | 29.16 | 60.16 | 2.02 | 1.37 | 0.70 | 3.95 |
| 7 | ELT-63-b | 0.77 | 0.94 | 23.62 | 64.81 | 2.72 | 0.96 | 0.71 | 4.48 |
| 8 | ELT-65-b | 0.49 | 0.95 | 30.11 | 60.29 | 2.27 | 2.29 | 0.62 | 1.97 |
| 9 | ELT-66-b | 0.21 | 1.08 | 24.35 | 65.93 | 2.69 | 1.03 | 0.56 | 3.14 |
| 10 | ELT-67-b | 0.18 | 1.02 | 21.92 | 68.85 | 2.39 | 0.84 | 0.52 | 3.28 |
| 11 | ELT-68-b | 0.26 | 1.22 | 20.78 | 67.07 | 2.65 | 0.95 | 0.56 | 5.52 |
| 12 | ELT-70-b | 0.07 | 0.95 | 21.80 | 70.27 | 1.81 | 1.10 | 0.53 | 2.48 |
| 13 | ELT-72-b | 0.21 | 2.13 | 16.38 | 75.32 | 1.64 | 0.27 | 0.60 | 2.45 |
| 14 | ELT-73-b | 0.26 | 1.24 | 20.77 | 67.42 | 2.46 | 1.08 | 0.58 | 5.20 |
| 15 | ELT-74-b | 0.37 | 1.17 | 25.21 | 65.09 | 2.95 | 0.87 | 0.63 | 2.71 |
| 16 | ELT-75-b | 0.23 | 0.94 | 29.91 | 59.89 | 2.17 | 1.19 | 0.76 | 3.91 |
| 17 | ELT-76-b | 0.07 | 1.09 | 23.74 | 67.65 | 2.48 | 0.72 | 0.51 | 2.73 |
| 19 | ELT-78-b | 0.74 | 0.82 | 17.69 | 72.53 | 2.12 | 0.22 | 0.61 | 4.28 |
| 20 | ELT-79-b | 0.07 | 1.36 | 22.04 | 67.92 | 2.76 | 0.68 | 0.65 | 3.52 |
| 21 | ELT-80-b | 0.27 | 0.76 | 24.52 | 64.88 | 2.70 | 1.17 | 0.59 | 4.11 |
| 22 | ELT-81-b | 0.46 | 1.21 | 18.59 | 72.50 | 2.04 | 0.36 | 0.89 | 2.95 |
| 23 | ELT-82-b | 0.22 | 1.17 | 22.30 | 67.93 | 2.40 | 0.19 | 0.78 | 4.03 |
| 24 | ELT-83-b | 0.31 | 1.18 | 26.06 | 65.20 | 2.54 | 0.47 | 0.61 | 2.62 |

 Table 2
 The main chemical composition of the white pottery, stamped hard pottery and proto-porcelain body samples from Erlitou by EDXRF (wt %).

Table 3 The main chemical composition of the proto-porcelain glaze samples from Erlitou by EDXRF (wt %).

| Code | Na ₂ O | MgO | Al ₂ O ₃ | SiO ₂ | K ₂ O | CaO | TiO ₂ | Fe ₂ O ₃ | MnO | P_2O_5 |
|----------|-------------------|------|--------------------------------|------------------|------------------|------|------------------|--------------------------------|------|----------|
| ELT-72-g | 0.34 | 3.63 | 12.32 | 66.87 | 2.98 | 9.26 | 0.48 | 3.11 | 0.40 | 0.25 |
| ELT-78-g | 0.31 | 2.16 | 15.48 | 66.30 | 3.75 | 5.17 | 0.56 | 5.28 | 0.77 | 0.22 |
| ELT-79-g | 0.68 | 2.74 | 17.18 | 62.40 | 5.18 | 6.65 | 0.41 | 3.22 | 0.13 | 0.21 |
| ELT-82-g | 0.51 | 2.48 | 14.98 | 64.29 | 3.27 | 8.65 | 0.47 | 4.36 | 0.14 | 0.99 |

had high Al₂O₃ content ranging from 20% to 30%, while the Fe₂O₃ content also had large variability. The colors of white pottery from Erlitou include white, yellow, red, and gray, with the average Fe₂O₃ content increasing with the change of color (white 2.44% \rightarrow yellow 2.63% \rightarrow red 3.47% \rightarrow gray 4.12%), although this rule is not absolute. It should be pointed out that although red or gray colored white pottery are slightly different from the general white pottery, they nevertheless should be distinguished from common pottery of a red or gray color because of their distinct clay chemical composition.

The body materials of four proto-porcelain samples

and one stamped hard pottery sample can be divided into two kinds based on the statistical analysis shown in Figure 1. The first kind consists of two proto-porcelain samples (ELT-79 and ELT-82), which were thick and had high Al₂O₃ content (about 22%) similar to white pottery. The second kind consists of three samples (proto-porcelain samples ELT-72, ELT-78 and stamped hard pottery sample ELT-81), which were thin with stamps on the surface and had high SiO₂ content (about 73%). Additionally, during the testing of proto-porcelain glazes on a few samples, we found that the chemical compositions of the glazes were very uneven across the surface of the sample, especially in the CaO content. The data in Table 3 is the average of multiple points of testing. From Table 3, the CaO content in the proto-porcelain glaze is between 5% and 9% and can be classified as a calcium glaze. Sample ELT-78 has high Fe₂O₃ content and can be classified as an iron-rich glaze with a certain amount of MnO and P₂O₅ as well, belonging to ash glaze type. Therefore the proto-porcelain glazes at Erlitou include both a green glaze with high calcium content and a brown glaze with high iron content. This is consistent with the characteristics of other protoporcelain glazes discovered so far.

Sample ELT-77, a white pottery sample, was in a conical shape and is the first example of this type ever found at Erlitou. In the same tomb, archaeologists also found a large-sized dragon-shaped object with inlayed turquoise designs and jades of non-local style, indicating that the grave's occupant might have had an unusual social position. The white pottery sample from this tomb had fine and smooth texture with bright red pigment on the surface (Figure 2). The results of non-destructive analysis of the clay matrix and the red pigment by EDXRF are listed in Table 4.

From the results shown in Table 4, the chemical composition of the red pigment on the surface of the white pottery sample suggests that the pigment was primarily cinnabar. Cinnabar was a very important mineral pigment in ancient times. Sample ELT-77 is the earliest example of white pottery decorated with cinnabar discovered so far. Additionally, the white clay body contains sulfur. Because the body chemical composition data was obtained only from surface testing, the sulfur might be contamination from the cinnabar. Other than the sulfur, the body material is similar to that of kaolin clay, although the Al₂O₃ content is higher and the Fe₂O₃ is lower than other white pottery samples shown in Table 2. This particular sample was very white and might have used pure kaolin with only

few impurities. The selection of such a fine clay material and the application of the cinnabar may indicate that the occupant of the tomb was a noble. Finally, we identified some strontium in the clay used for this sample, but strontium is not found in other white pottery, further indicating that sample ELT-77 had a special source.

3. Analysis of firing technology.

Thermal dilatometers (German, DIL402C) were used to determine the firing temperature of the samples and also observe color changes after sample re-firing. The results are shown in Table 5.

The firing temperatures of the white pottery from Erlitou were between 900° C to 1000° C. From the color changes after re-firing in an unprotected atmosphere, the color of yellow and reddish samples did not change, but gray samples changed to yellow or brown. We can infer that the gray samples were fired in a reducing atmosphere and the yellow and reddish samples were fired in an oxidizing atmosphere. Sample ELT-58 is reddish on the outside and gray on the inside, so this sample may have been fired with insufficient oxidation. Because samples of differently colored white pottery can have either similar or different Fe₂O₃ content, the various colors of the white pottery may likely be relate to both chemical composition and firing atmosphere.

The firing temperature of the proto-porcelain wasn't determined because of the limitation of sample size and rarity of this type of ceramic. Instead, we used X-Ray diffraction analysis to determine the phase composition of proto-porcelain sample ELT-72 (Erlitou Phase II). The result is shown in Figure 3.

From Figure 3 we can see that there are both



Figure 2 Sample ELT-77.

Table 4 Chemical compositions of the white clay matrix body and red pigment of ELT-77 (wt %).

| | Al ₂ O ₃ | SiO ₂ | SO ₃ | K ₂ O | CaO | Fe ₂ O ₃ | SrO |
|-------------|--------------------------------|------------------|-----------------|------------------|------|--------------------------------|-------|
| white body | 41.57 | 37.69 | 15.87 | 3.15 | 0.48 | 0.65 | 0.59 |
| Dedeisment | Al | Si | S | K | Ca | Fe | Hg |
| Keu pigment | 2.93 | 8.23 | 9.15 | 1.04 | 0.51 | 1.28 | 76.77 |

| | ELT-56 | ELT-57 | ELT-58 | ELT-61 | ELT-63 |
|--------------------|--------|--------|---------|--------|--------|
| Before re-firing | yellow | yellow | reddish | gray | gray |
| After re-firing | yellow | yellow | reddish | yellow | brown |
| Firing temperature | 980°C | 1000°C | 925°C | 950°C | 1000°C |

Table 5 Firing temperatures of some white pottery and the color changes after re-firing in unprotected atmosphere.



Figure 3 XRD spectrum of the body of sample ELT-72.

cristobalite and mullite in the crystal phase beside α -quartz in the clay matrix, which is consistent with the characteristics of porcelain. We can infer that the firing temperature of this sample should be above 1000° C, which is higher than the firing temperature of white pottery and also indicates that the high temperature kiln technology had already been improved by Erlitou Phase II. This sample was green glazed and stamped, with the glaze appearing very even on the surface and associating well with the clay body. From the microstructure observed in a thin cross section of this sample (shown in Figure 4), we can see that the glaze was 130μ m thick. There are a few anorthite crystals in the glaze and there is no interlayer between the body and glaze, which reflects the primitiveness of the glazing and firing technology at that time. Chen Yaocheng (2002) studied several hard potteries with vitreous layers at Erlitou and found that the firing temperature was about 1160° C. Chen also noted that the vitreous layer was not from the application of the glaze itself, but rather was a natural glaze of ash fell or "kiln sweat" dripped down from the roof of the kiln because the thickness of the layer was very thin $(10\mu m - 50\mu m)$ and discontinuous. Our experiment result for sample ELT-72 is not consistent with Chen's opinion. The glaze thickness exceeds 100μ m and is very even, which should be characteristic of proto-porcelain glazes. The contradictory findings may be related to the different samples we used, but we believe that sample ELT-72 is a good one. Chen



 SICCA SEI
 20.0kV
 ×1,500
 10µm
 WD11mm

 Figure 4
 Microstructure of a cross section of sample ELT-72.

 Top.
 Microstructures of glaze and body layer;

 bottom.
 Anorthite crystals in the glaze.

and others also studied the samples from Maqiao in Shanghai (1999), which dates to a similar time period as Erlitou. They concluded that there were both real protoporcelain and hard pottery with vitreous layers present at Maqiao, which is similar to the Erlitou case. Therefore we believe that the Erlitou Phase II may mark the invention of proto-porcelain in ancient China.

4. The relationship between proto-porcelains from Erlitou and other sites.

Figure 1 shows that the experimental samples of proto-porcelain and stamped hard pottery from Erlitou site can be divided into two types: one with high Al₂O₃ content, and the other with high SiO₂ content similar to the porcelain in southern China. The origin of protoporcelain is still an unsolved mystery. Luo Hongjie and others (1996) argued that the proto-porcelain unearthed in northern China originated from the south because the clays of northern proto-porcelains were similar to those made in southern China. The source of the earliest protoporcelain samples in northern China has become a focus of research among Chinese archaeologists. We carried out statistical analysis on the chemical composition of samples from Erlitou and other sites to further explore the potential source of the proto-porcelain. The results are shown in Figures 5 and 6. It should be point out that the chemical composition data of samples from Erlitou also include Chen Yaocheng's study as well as our own work. The data of other sites are cited from Li Jiazhi's work (1998), which are the sites of Yuangu in Shanxi Province (Shang Dynasty), Zhengzhou in Henan Province (Shang Dynasty), Magiao in Shanghai (Xia-Shang Dynasties), Wucheng in Zhangshu and Jiaoshan in Yingtan (both of the Shang Dynasty in Jiangxi Province), and so on. The data were collected by different researchers using different methods, so there may be some odds and errors.

In Chen Yaocheng's work, they inferred that the protoporcelains from Erlitou should originate from southern China because the clay composition materials (SiO₂ content 69.6%-73.5%, Al₂O₃ content 17.5%-22.9%) were similar to the proto-porcelain from Zhejiang and Jiangsu Provinces. In Figure 5, we can see that some of the protoporcelains from Erlitou with high SiO₂ content overlap with other sites. There is a transition region between



Figure 5 Two-dimensional correspondence analysis diagram of the main chemical compositions of white pottery, stamped hard pottery and proto-porcelain body samples from Erlitou and proto-porcelain from other sites.

the white pottery and proto-porcelains, which includes four white pottery samples from Erlitou, three protoporcelain samples from Erlitou (ELT-72, ELT-82, and a sample of 22.9% Al₂O₃ content in Chen Yaocheng's work), and one proto-porcelain sample from Wucheng. The above results are similar to those shown in Figure 1, which indicates that although these data come from different testing methods, the general trends are basically the same. Because there was no white pottery sample in Chen Yaocheng's work, the high Al₂O₃ content is not as obvious as it would be. The main characteristic of the samples in the transition region is that their Al₂O₃ content (20.5%-23%) is lower than that in most white pottery and higher than in most proto-porcelain samples. Additionally, the white pottery samples in the transition region are all gray colored, so they may be closely related to the other three proto-porcelains. So far, the clay body materials of all white pottery unearthed in northern China have high Al₂O₃ content, which is similar to the porcelain used in northern China during later periods. The relationship between white pottery and porcelain in northern China should receive more attention and further research. From Figure 6 we can see that the proto-porcelain glaze can be divided into two types: one is an iron-rich glaze (Region I) and the other is a calcium-rich glaze (Region II). The proto-porcelain glaze samples from Erlitou fall in the middle of these two types of glaze. In other words, the CaO and Fe₂O₃ contents in Erlitou glazes do not fall at the extreme high-end of calcium glaze and iron glaze types, but do indicate that the basic aspects of these two kinds of glazes had already appeared by the Erlitou period.

According to the above analysis, although the number of proto-porcelain samples from Erlitou in our study is small, we are still able to classify their clay raw materials into two kinds, which may indicate that the proto-



Figure 6 Two-dimensional correspondence analysis diagram of the main chemical compositions of protoporcelain glaze samples from Erlitou and other sites.

porcelains at Erlitou Site had diverse origins. The protoporcelain samples with bodies similar to that of gray white pottery cannot regard white pottery as progenitor at present, but their relationship was nevertheless very close. Although the area where proto-porcelain originated is not yet clear, our discussion on when it originated is still meaningful. In the future, researchers need to carry out comprehensive analysis on many more samples to resolve the origin of proto-porcelain in ancient China.

Conclusions

1. The clay body materials of white pottery, stamped hard pottery and proto-porcelain at Erlitou Site are different from those of the common daily-use pottery. The clay bodies have low flux content and belong to porcelain clay, which indicates that the ancient potters of the Erlitou Culture could select raw materials for specific use. This kind of raw materials can be fired at a higher temperature, a trait that became the foundation of proto-porcelain and porcelain production in later periods.

2. The range of raw materials used to make white pottery is quite variable, with increasing Fe₂O₃ content causing color changes from white, to yellow, to red, to gray $(2.44\% \rightarrow 2.63\% \rightarrow 3.47\% \rightarrow 4.12\%)$. One particular sample was the conical-shaped white pottery object unearthed from a noble tomb made of pure kaolin and decorated with cinnabar.

3. The firing temperature of the white pottery at Erlitou is around 900-1000° C. The firing atmosphere of white pottery varied and produced different colors. The crystal phase in the clay matrix body of proto-porcelain sample ELT-72 is similar to common porcelain and its firing temperature likely exceeded 1100° C. There are a few anorthite crystals in the glaze of this sample and no interlayer between the body and glaze, which indicates the primitiveness of the firing and glazing technology. However, it is clear that the glaze did not form unintentionally by ash and kiln sweat falling on the surface.

4. There are two kinds of clay materials of protoporcelain and stamped hard pottery at Erlitou. One kind has high silicon and low aluminum content and is similar to porcelain clay in southern China. The other kind is closely related to other gray white pottery at Erlitou. Further research is needed on the potential sources of these proto-porcelain ceramics at Erlitou Site.

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Postscript

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