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Colorimetric Analysis, Chemical Composition, and Mineralogical Characterization of Ceramics from the Tule'i Archaeological Site, Khuzestan: A Multidisciplinary Approach to the Study of Archaeological Materials

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Abstract

Tule'i Tepe, located in northern Khuzestan, is one of the key Neolithic sites on the Susiana Plain and has been dated to ca. 6200–5900 BCE based on previous studies. Although the site has been interpreted as a seasonal settlement or a mobile pastoralist camp with a relatively short occupation span, it is remarkably rich in material culture, particularly ceramics. Excavations at Tule'i have yielded a diverse ceramic assemblage, including both plain and painted Jaffar type pottery, as well as white and red wares decorated with various motifs, which show stylistic parallels with contemporary sites in Mesopotamia and southwestern Iran. Despite the significance of the site for studies of the Neolithic period and pastoralism, no comprehensive investigation has yet been conducted into the raw materials, production technology, or provenance of its pottery. The present study aims to address this gap by applying a multidisciplinary approach to the analysis of 41 ceramic samples from Tule'i Tepe. Three complementary analytical methods were employed: spectrophotometry for precise color measurement, portable X ray fluorescence (pXRF) for the determination of major and minor elements, and X ray diffraction (XRD) for mineralogical phase identification and the estimation of firing temperatures. Elemental analysis reveals that buff colored ceramic bodies contain the highest calcium (Ca) content, whereas red colored bodies show higher concentrations of iron (Fe) and aluminium (Al). Manganese (Mn) was identified as the primary coloring agent in the brownish decorations applied to the pottery. XRD analysis indicates the presence of minerals such as pyroxene and phlogopite, suggesting firing temperatures in the range of 600–700 °C. The presence of calcite in certain samples further indicates that firing temperatures remained below 800 °C, since calcite decomposes at higher temperatures. These findings suggest that Tule'i potters possessed considerable technical knowledge in controlling firing temperatures and selecting raw materials. A secondary aspect of this study compares the effectiveness of two color measurement methods: conventional visual comparison using the Munsell Soil Color Chart and instrumental analysis using reflectance spectrophotometry. The results indicate that the spectrophotometric method, owing to its higher resolution in distinguishing closely related colors (e.g., buff and light brown), can serve as an effective complement to non instrumental methods.

Keywords: Neolithic period, Tule'i pottery, spectrophotometry, mineralogy, chemical analysis, Munsell Soil Color Chart.

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Introduction

Tule'i Tepe, also referred to—sometimes erroneously—as Tuleiy or Tulai in the archaeological literature, is a small Neolithic site (ca. 6200–5900 BCE) located in northern Khuzestan (Fig. 1). It was excavated during a brief salvage operation in 1973 by Professor Frank Hole. The site takes its name from tuleh (tule'i), or wild mallow, a broad leaved plant that grows wild across much of southwestern Iran. A variety of ceramic types have been recovered from Tule'i Tepe. The pottery from this site has been classified using the same terminology applied to the Deh Luran region and includes the following categories: Jaffar painted ware, Jaffar plain ware, Khazineh red ware, white ware with red on buff decoration, white ware with black on buff decoration, painted white ware with black motifs, and white scratched ware. In addition, a grey ware—unknown in the Deh Luran region—has also been identified at Tule'i. The scratched painted wares from Tule'i resemble those of the Susiana A–B phases from Chogha Mish. Based on the ceramic assemblage, Frank Hole has suggested that the occupation of Tule'i Tepe falls chronologically between the Mohammad Jaffar and Sefid phases of Deh Luran (Malek Shahmirzadi, 1999: 232). The classification of pottery from this site, as outlined above, is based on macroscopic attributes such as color, texture, and decoration. Apart from clay preparation and processing, the quality of these ceramics—particularly their color—is largely determined by the early kilns used for firing (Rahimi and Mehran, 2003). These kilns, primarily fueled by plant materials, produced uneven temperatures within different parts of the firing chamber. Consequently, the quality of the pottery was influenced by its position in the kiln, the intensity of the heat, and the duration of firing (Ashurst and Ashurst, 2001: 47–48). Today, the attributes mentioned above can be examined and evaluated using scientific and instrumental methods, which allow for more precise data and refined classifications. This paper attempts to analyze and compare data from the various pottery types at this site through a multidisciplinary approach employing different instrumental techniques. The importance of colorimetric analysis, chemical composition, and mineralogical characterization in the study of ceramic samples highlights the necessity of employing such analytical methods (Stuart, 2007).

Research Background of Tule'i Tepe

In the spring of 1973, a small trench was excavated at Tule'i Tepe, revealing a temporary settlement interpreted as a campsite associated with a broader nomadic occupation. Chronologically, the site is comparable to the Sefid phase in the Deh Luran Plain. Tule'i may have been occupied for approximately 200 years, between 6200 and 5900 BCE. Excavations continued in several trenches and yielded dry stone wall foundations, pottery sherds, and T shaped figurines. Based on the Deh Luran chronology, Tule'i Tepe can be placed between the Mohammad Jaffar and Early Sefid phases. Although certain differences exist in the ceramics of the two regions (Fig. 2), other artifacts appear to

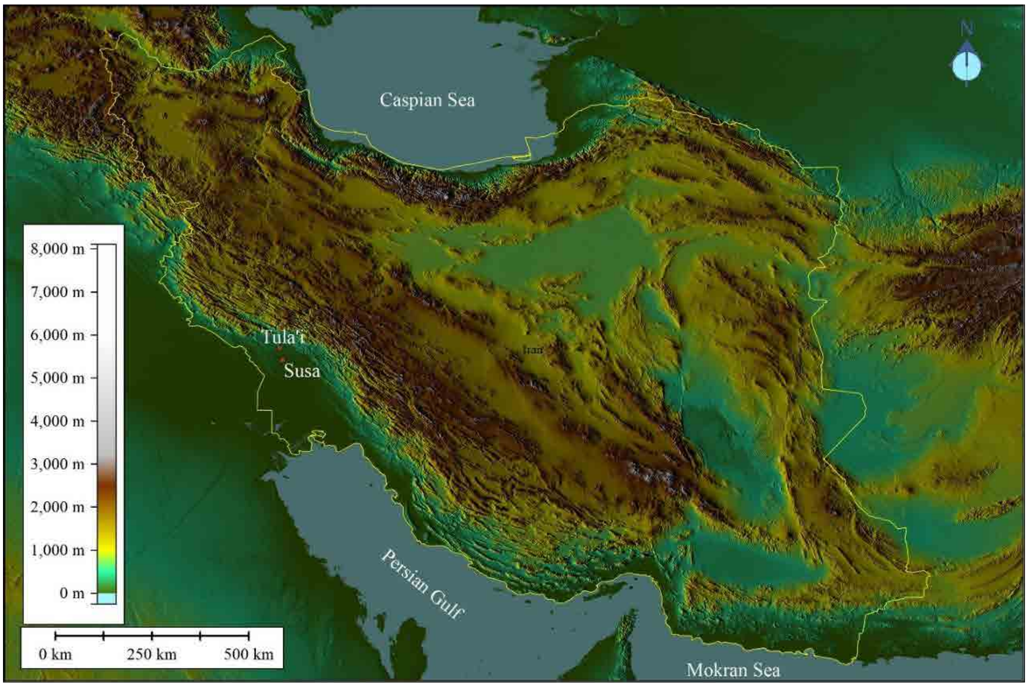

Fig. 1: Geographic location of the Tule'i site and its relative position to Susa (Author, 2025).

be identical. Changes in the two sequences—Deh Luran and Tule'i—follow the same general order; only the proportions and decorative designs of specific pottery types differ. At least part of Tule'i consists of a campsite whose layout is consistent with that of modern nomadic encampments. The arrangement and spacing of the stones leave little doubt regarding the nature of this portion of the site (Hole, 1974).

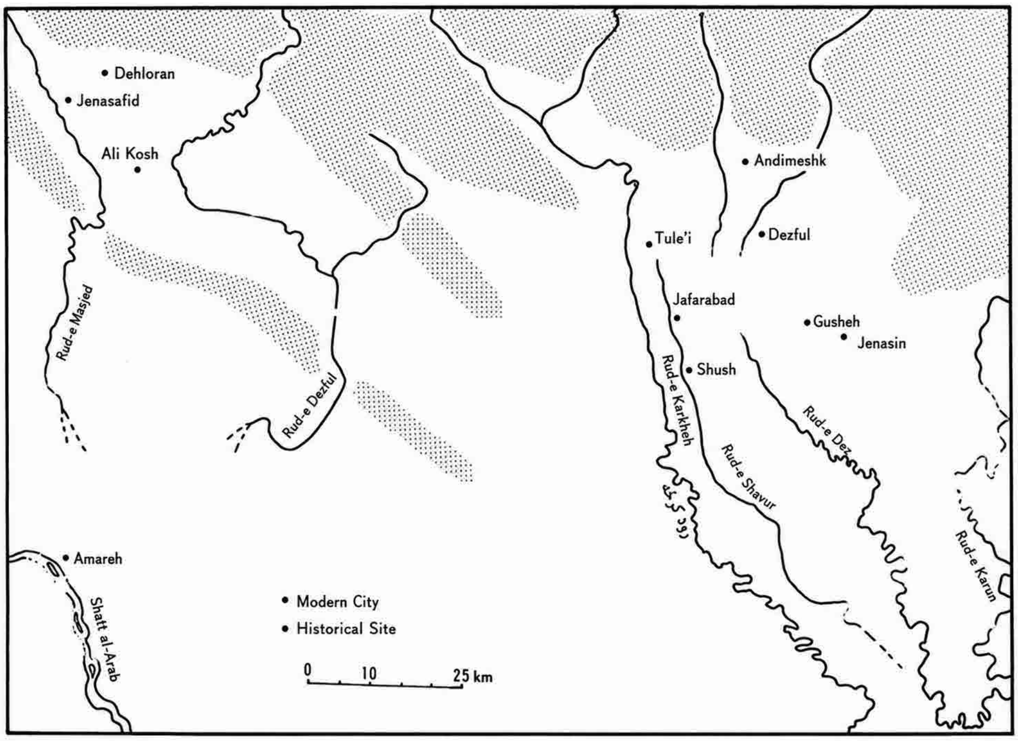

Fig. 2: Prehistoric sites of Deh Luran and Khuzestan (Hole, 1974: 220, Fig. 1).

Frank Hole's interpretations have been challenged by several scholars. Reinhard Bernbeck, in a paper entitled "Early Pastoral Patterns: A Reassessment of Tule'i Tepe," reanalysed the archaeological data derived from the material remains at Tule'i Tepe and argued that the site was occupied by village based herders who moved periodically along the Zagros foothills, exploiting favourable settlement locations. This type of mobility involved relatively long distance movements and differed considerably from the model proposed by Mortensen for earlier Neolithic sites (Mortensen, 1972). It also does not correspond to Henrikson's description of early Chalcolithic pastoral nomads in the Zagros Mountains (Henrickson, 1985: 27–33). It is possible that we are dealing with an intermediate type of subsistence pattern, one that lies between a "broad spectrum economy" and specialised mobile pastoralism combined with agriculture. Köhler Rollefson reached similar conclusions for the roughly contemporary "PPNC" village economy at 'Ain Ghazal in Jordan (Köhler Rollefson, 1988: 91). The Zagros region during the Late Neolithic may present a comparable example. At Qaleh Rostam, a village site, there is clear evidence for agriculture (Zagarell, 1989: 287). Risk reduction appears to have been one of the primary functions of the mobility described here. Pires Ferreira cites three reasons for maintaining fallow herds: (1) preventing overgrazing around the village; (2) enabling animals in fallow herds to benefit from better pastures; and (3) reducing the risk of losing a large portion of the herd to epidemic disease (Pires Ferreira, 1977: 279).

On the other hand, a pastoral economy based primarily on animal products is a high risk enterprise, involving periodic failures that could threaten the survival of entire groups (Barth, 1961: 108–109). Long term research shows that specialised nomadic groups are periodically compelled to purchase or obtain fodder from sedentary populations (Balland and Kieffer, 1979: 81). In some cases, raids by mobile pastoral communities on neighbouring sedentary populations may serve as a means of compensating for such shortages. In light of these perspectives, and according to the model proposed by Lees and Bates, specialised nomadic pastoralism is only feasible when combined with substantial surplus production by sedentary farmers (Lees and Bates, 1974). Hole proposed a slightly modified version of this modern nomadic model. According to this interpretation, the herders at Tule'i Tepe were able to live independently, relying on animal products and the surrounding natural environment. Such a subsistence strategy could theoretically have developed separately from agriculture. However, under this scenario it is difficult to explain the numerous connections between the inhabitants of Tule'i Tepe and the sedentary villages of the surrounding Deh Luran and Susiana plains. It has also been suggested that such herders might have reverted to hunting and gathering during periods of food shortage. Such periodic dietary shifts should be identifiable through variation in the animal remains consumed during lean periods (Colson, 1979: 22). In fact, there is no evidence at Tule'i Tepe for a diet different from that of its contemporary site, Chagha Sefid. At both sites, the faunal assemblages contain less than 10% wild animals, with gazelle representing the primary non domestic meat source. These similarities instead point to a shared economic foundation, as well as comparable strategies for protecting villages and settlement sites (Pires Ferreira, 1977, Table 3). Daniel Potts, in his book Nomadism in Iran: From Antiquity to the Modern Era, also criticized Frank Hole's theory of nomadism (Potts, 2014). The episodic or discontinuous nature of the occupation at Tule'i Tepe has never been questioned, but it remains worth asking whether the identification of nomads in Iran during the seventh millennium BCE is justified. It is very likely that herders maintaining fallow herds used this site seasonally. However, Hole's primary aim was not merely to argue that "the site was temporarily and seasonally occupied" (Hole, 2004). Rather, his objective was to identify nomads in the archaeological record thousands of years before most scholars—aside from Elizabeth Bacon, whose writings on the origins of nomadism in the 1970s had little influence on archaeologists—considered such a possibility. Until 2004, the "full range of village material culture" that Hole identified in the Tule'i assemblage had been interpreted not as evidence of permanent settlement, but rather as indicating that "a stable and complete life was maintained in tent sites with the presence of all members of the nuclear family" (Ibid: 80). This interpretation presents a view of Tule'i Tepe that differs markedly from those proposed during the preceding three decades. No explanation was provided as to why the large quantities of pottery sherds; grinding stones (including mortars, querns, stone blades, discs, pounders, and weights); fired clay animal figurines; and sling bullets should be interpreted as evidence for the presence of all members of clusters of nuclear nomadic families. Undoubtedly, this represents a highly selective interpretation of the site's material culture. Moreover, as Bernbeck demonstrated, the pottery types present do not support a strictly vertical nomadic interpretation, because their decoration lacks characteristics typical of sites located at higher elevations in the Zagros—which might represent the summer territories of groups wintering at Tule'i—but instead shows closer affinities with lowland sites such as Chogha Bonut, Chagha Sefid, and Chogha Mish (Bernbeck, 1992). Donna J. Sutliff, in a paper entitled "On Neolithic Nomadism at Tule'i, Iran: A Re evaluation of the Evidence," also critiqued Frank Hole's theory (Sutliff, 2015). Frank Hole later responded to this critique in a 2023 article entitled "Tule'i and the Question of Mobile Herders" (Hole, 2023).

Research Methodology

A total of 41 pottery sherds, including red, buff, and brown painted wares, were selected for analysis (Fig. 3).

Colorimetry

Colorimetric analysis of the samples was conducted using the CIELAB color space (L*, a*, b*) and reflectance spectra in the visible range (400–700 nm), measured with a Konica Minolta CM 2600d spectrophotometer. In addition, color assessment was

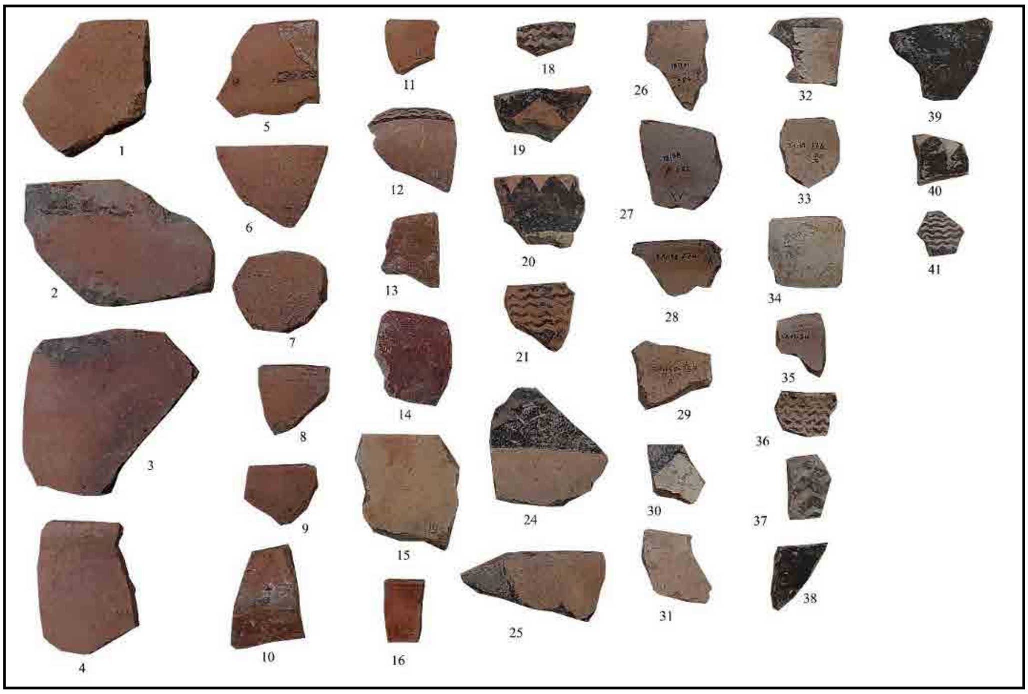

Fig. 3: The 41 ceramic sherds selected for analysis, including red, buff, and brown/black-painted wares (Author, 2025).

performed using the Munsell Soil Color Chart, based on the three attributes of hue, value, and chroma.

The CIELAB color model (L*, a*, b*) comprises the following parameters:

• L*: lightness (ranging from black to white)

• a*: the red–green chromatic axis

• b*: the yellow–blue chromatic axis

Chemical Analysis

Portable X ray fluorescence (pXRF) spectrometry was employed using a Thermo Scientific Niton XL3t GOLDD+ 950 instrument (USA). The technical specifications of the device are as follows: SDD detector; 6–50 kV / 0–200 µA; maximum power 2 W; Ag anode X ray tube.

Mineralogical Analysis

Mineral phase identification in the ceramic bodies was conducted using X ray diffraction (XRD) analysis with a Seifert 3000TT diffractometer. All of the above analyses were carried out at the Research Center for Conservation of Cultural Relics, Research Institute of Cultural Heritage and Tourism (RICHT), in collaboration with the Iranian Center for Archaeological Research (ICAR).

Results and Discussion

The CIELAB colorimetric data (L*, a*, b*) for the red-bodied ceramics are summarized in Table 1. As illustrated in Fig. 3, the L* (lightness) values for these samples range predominantly between 42 and 64. The lowest lightness value was recorded for the exterior surface of sample Touleie-32-R, while the highest was observed in sample Touleie-4-R.

Table 1. Color data (L*, a*, b*) for the red-colored ceramic group (Author, 2025).

Row	Color code	L*	a*	b*
1	Touleie-1-R	57.86	18.71	25.99
2	Touleie-2-R	60.14	17.94	25.02
3	Touleie-3-R	62.24	18.99	23.5
4	Touleie-4-R	63.75	17.82	24.57
5	Touleie-5-R	58.76	17.08	24.89
6	Touleie-6-R	56.32	15.75	22.98
7	Touleie-7-R	58.5	16.17	25.48
8	Touleie-8-R	59.07	16.88	26.5
9	Touleie-9-R	58.03	16.65	24.06
10	Touleie-10-R	50.14	18.89	19.7
11	Touleie-10-R-I	58.76	16.41	25.51
12	Touleie-10-R-O	58.02	14.01	21.48
13	Touleie-11-R	49.36	15.55	20.23
14	Touleie-12-R	47.58	18.73	17.81
15	Touleie-13-R	58	24.75	27.93
16	Touleie-14-R	59.33	17.14	23.94
17	Touleie-16-R	61.06	14.59	21.56
18	Touleie-30-R	59.26	13.61	21.75
19	Touleie-32-R	42.42	17.05	16.95

The a* (red–green) chromatic values predominantly range between 13 and 25. The highest a* value (24.8) corresponds to sample Touleie-13-R, whereas the lowest (13.6) was recorded for the interior surface of sample Touleie-30-R. The b* (yellow–blue) values range between 16 and 28. The highest b* value (27.9), indicating a more pronounced yellow hue, was observed in sample Touleie-13-R; conversely, the lowest (16.9) was found on the exterior surface of sample Touleie-32-R, suggesting a relative shift towards the blue spectrum.

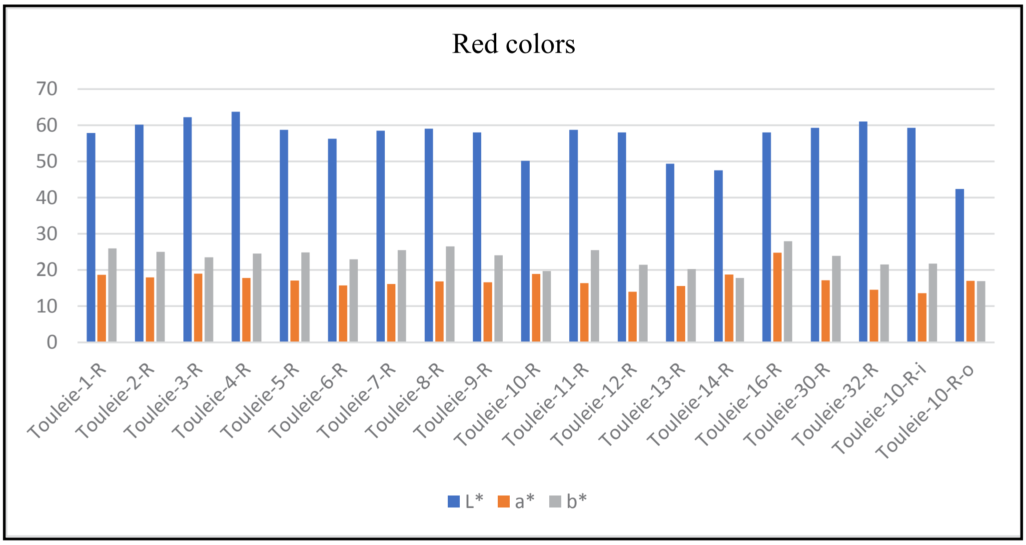

Fig. 3: Distribution of L*, a*, b* color data for the red ware group (Author, 2025).

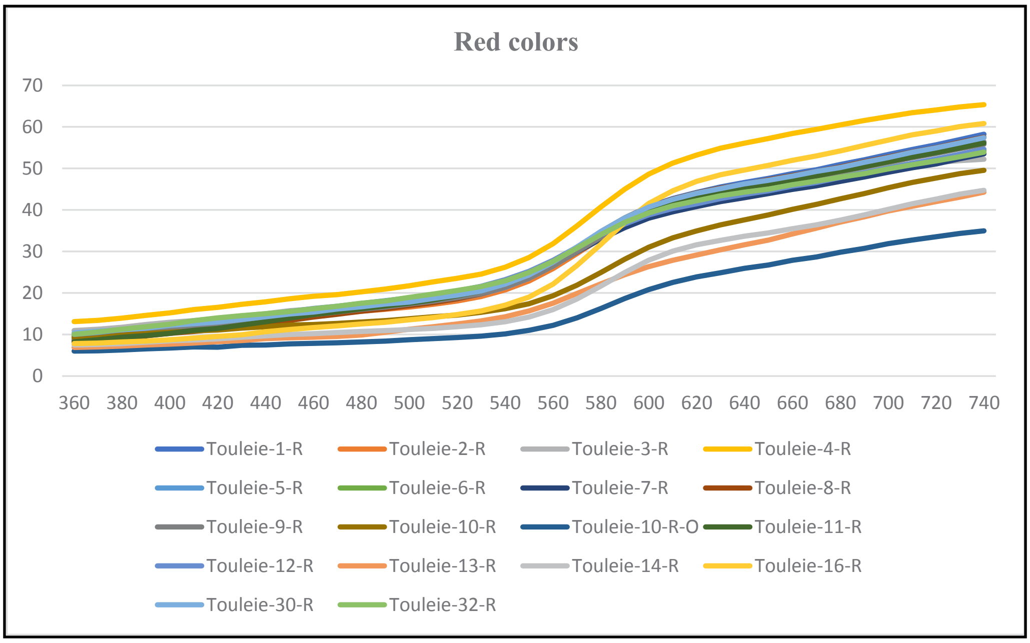

Fig. 4: Visible-range reflectance spectra of the red ware group (Author, 2025).

The CIELAB colorimetric data (L*, a*, b*) for the buff-bodied ceramics are summarized in Table 3. As illustrated by the quantitative values and Figure 8, the L* (lightness) values for these samples range between 59 and 75. The lowest lightness value (59.7), indicating the highest degree of darkness, is associated with sample Touleie-25-R, while the highest lightness (74.3) corresponds to sample Touleie-30-R. The a* (red–green) values for these samples range predominantly between 5.5 and 13.0. The highest a* value (13.1) belongs to sample Touleie-25-Y, and the lowest (5.7) was recorded for the interior surface of sample Touleie-30-Y. The b* (yellow–blue) values range between 19 and 27. The highest b* value (26.8) was observed in sample Touleie-15-Y, while the lowest (19.8) was found on the exterior surface of sample Touleie-35-Y.

Table 2. Color data (L*, a*, b*) for the buff-bodied or buff-surfaced ceramic group (Author, 2025).

Row	Color code	L*	a*	b*
1	Touleie-15-Y	65.06	11.07	26.83
2	Touleie-24-Y	60.4	10.78	23.93
3	Touleie-25-Y	59.68	13.12	23.7
4	Touleie-26-Y	61.65	10.19	23.01
5	Touleie-27-Y	61.75	10.54	23.13
6	Touleie-28-Y	65.35	9.84	21.84
7	Touleie-29-Y	61.79	12.4	23.9
8	Touleie-30-Y	74.29	5.69	20.97
9	Touleie-31-Y	66.91	10.5	21.73
10	Touleie-32-Y	67.35	7.53	21.02
11	Touleie-33-Y	70.03	6.8	21.87
12	Touleie-34-Y	63.57	8.67	21.44
13	Touleie-35-Y	66.25	11.7	19.75
14	Touleie-36-Y	60.36	11.76	23.33
15	Touleie-37-Y	66.55	8.16	23.27
16	Touleie-38-Y	63.49	9.61	22.17
17	Touleie-39-Y	61.1	9.48	21.66
18	Touleie-40-Y	69.67	8.42	22.13
19	Touleie-41-Y	66	12.26	28.55

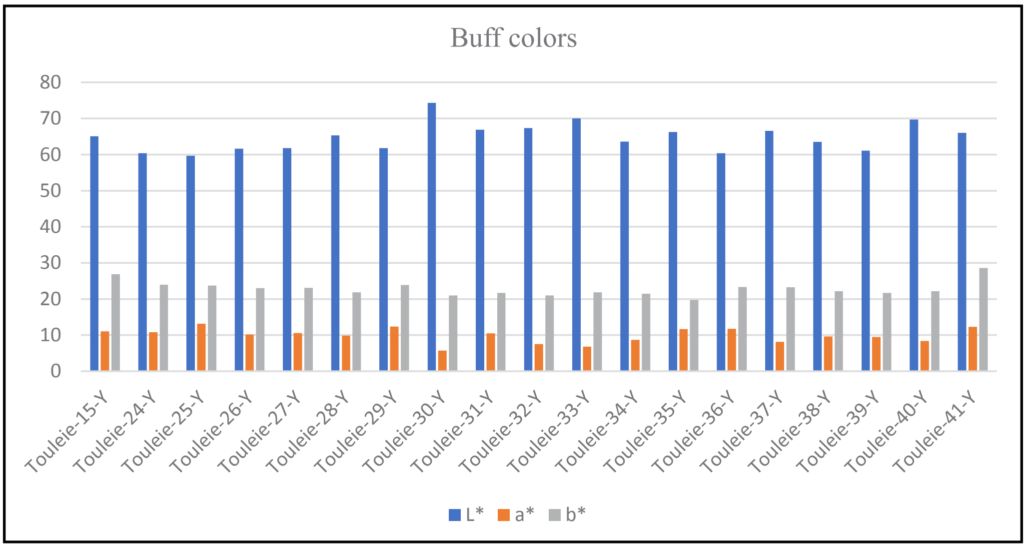

Fig. 5: Distribution of L*, a*, b* color data for the buff-colored ware group / buff-surfaced group (Author, 2025).

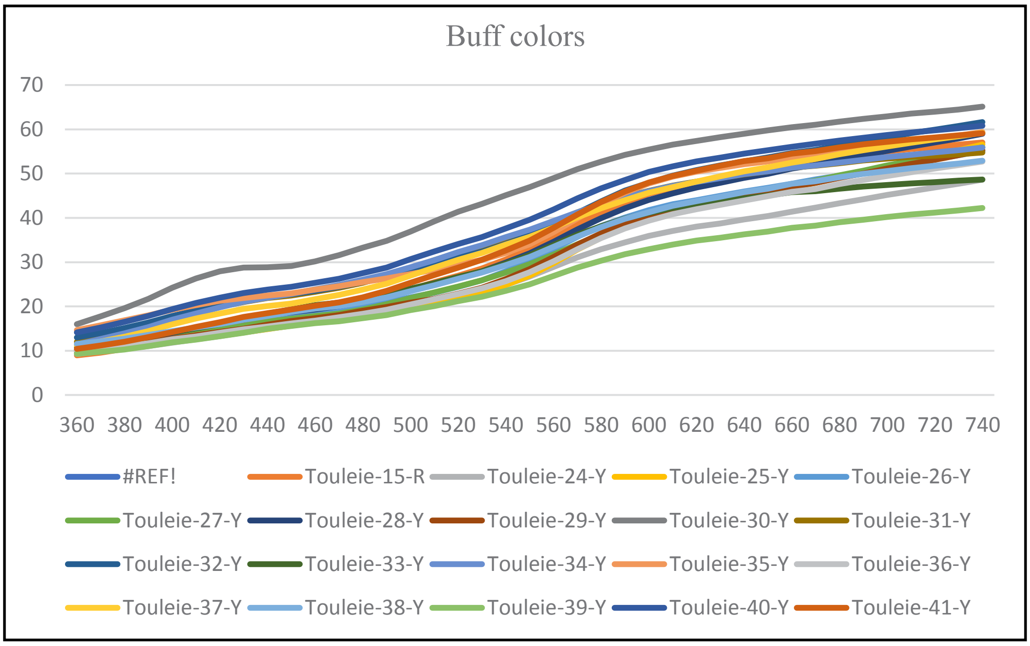

Fig. 6: Visible-range reflectance spectra of the buff ware group / buff-surfaced group (Author, 2025).

The CIELAB colorimetric data (L*, a*, b*) for the brown-surfaced ceramics are summarized in Table 4. As shown by the color parameters in this table and Figure 10, the L* (lightness) values for these samples range predominantly between 36 and 54. The lowest lightness value (36.9) corresponds to sample Touleie-19-B, while the highest (53.9) was recorded for sample Touleie-36-B. The a* (red–green) values for these samples range between 2.5 and 7.5. The highest a* value (7.3) belongs to sample Touleie-36-B, and the lowest (2.7) was observed in samples Touleie-20-B and Touleie-38-B. The b* (yellow–blue) values are predominantly between 8.0 and 16.5. The highest b* value (16.4), indicating a more pronounced yellowish hue, corresponds to sample Touleie-36-B, while the lowest (8.0), suggesting a shift towards the blue spectrum, was found on the exterior surfaces of samples Touleie-20-B and Touleie-38-B.

Table 3. Color data (L*, a*, b*) for the brown-painted decorations on the ceramics (Author, 2025).

Row	Color code	L*	b*	b*
1	Touleie-18-B	47.38	5.59	12.19
2	Touleie-19-B	36.9	4.05	9.3
3	Touleie-20-B	41.64	2.72	7.88
4	Touleie-21-B	44.31	6.89	11.55
5	Touleie-36-B	53.91	7.25	16.42
6	Touleie-37-B	52.34	5.01	13.75
7	Touleie-38-B	39.43	2.68	8.12
8	Touleie-39-B	43.93	3.78	9.47
9	Touleie-40-B	44.36	5.2	10.55
10	Touleie-41-B	50.38	3.27	8.5

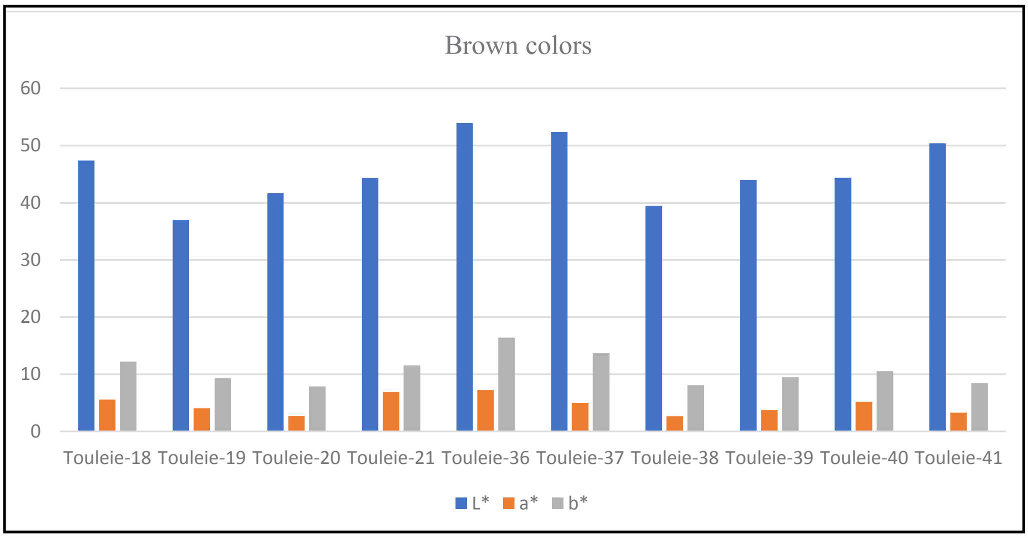

Fig. 7: Distribution of L*, a*, b* color data for the brown-painted ceramic decorations (Author, 2025).

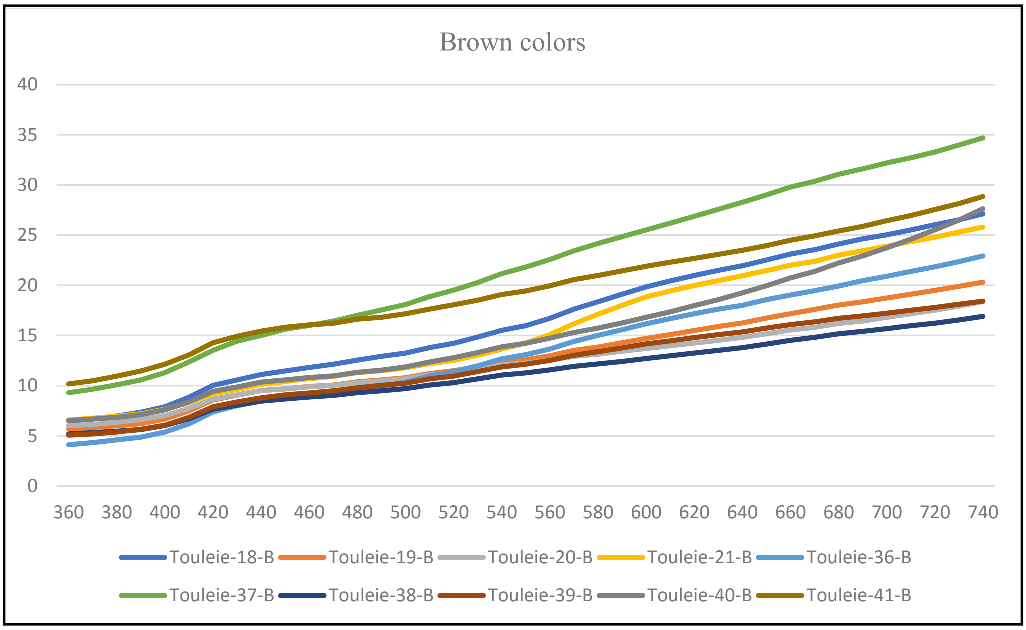

Fig. 8: Visible-range reflectance spectra of the brown-painted ceramic decorations (Author, 2025).

Munsell Color Chart

The ceramic samples were also examined using the Munsell Soil Color Chart. The results are summarised in Table 4.

Table 4. Results of colorimetric analysis of the ceramic color components using the Munsell Soil Color Chart (Author, 2025).

Row	Color code	Munsell color code	Row	Color code	Munsell color code
1	Touleie-1-R-M	2.5YR 6/6 light red	20	Touleie-15-Y-M	1.5YR 8/3 pink
2	Touleie-2-R-M	2.5YR 7/6 light red	21	Touleie-24-Y-M	5YR 7/3 pink
3	Touleie-3-R-M	2.5YR 7/6 light red	22	Touleie-25-Y-M	5YR 7/6 reddish yellow
4	Touleie-4-R-M	2.5YR 7/6 light red	23	Touleie-26-Y-M	5YR 6/2 pinkish gray
5	Touleie-5-R-M	2.5YR 6/6 light red	24	Touleie-27-Y-M	5YR 4/1 dark gray
6	Touleie-6-R-M	5YR 6/6 reddish yellow	25	Touleie-28-Y-M	5YR 5/1 gray
7	Touleie-7-R-M	5YR 7/6 reddish yellow	26	Touleie-29-Y-M	2.5YR 4/2 weak red
8	Touleie-8-R-M	5YR 7/6 reddish yellow	27	Touleie-30-Y-M	5YR 5/1 gray
9	Touleie-9-R-M	5YR 6/6 reddish yellow	28	Touleie-31-Y-M	7.5YR 8/3 pink
10	Touleie-10-R-M	2.5YR 6/6 light red	29	Touleie-32-Y-M	2.5YR 6/2 pale red
11	Touleie-10-R-I		30	Touleie-33-Y-M	5YR 6/2 pinkish gray
12	Touleie-10-R-O		31	Touleie-34-Y-M	7.5YR 8/3 pink
13	Touleie-11-R-M	2.5YR 6/6 light red	32	Touleie-35-Y-M	2.5YR 5/1 reddish gray
14	Touleie-12-R-M	2.5YR 6/6 light red	33	Touleie-36-Y-M	5YR 7/4 pink, 5YR 5/2 reddish gray
15	Touleie-13-R-M	2.5YR 6/4 light reddish brown	34	Touleie-37-Y-M	5YR 7/2 pinkish gray
16	Touleie-14-R-M	2.5YR 4/8 red	35	Touleie-38-Y-M	5YR 4/1 dark gray
17	Touleie-16-R-M		36	Touleie-39-Y-M	2.5YR 4/2 weak red
18	Touleie-30-R-M		37	Touleie-40-Y-M	7.5YR 8/2 pinkish white
19	Touleie-32-R-M		38	Touleie-41-Y-M	2.5YR 8/2 pinkish white

The color chips identified in Table 4 were also measured using the spectrophotometer. Figure 9 illustrates the colorimetric discrepancies between the visual and instrumental methods (calculated according to Equation 1) [21] (Johnston-Feller, 2001). As observed in the chart, the smallest differences between the two methods are associated with red tones, whereas the most significant discrepancies occur within the buff and brown hues.

$$\Delta E = \sqrt{(L2 - L1)^2 + (a2 - a1)^2 + (b2 - b1)^2}$$

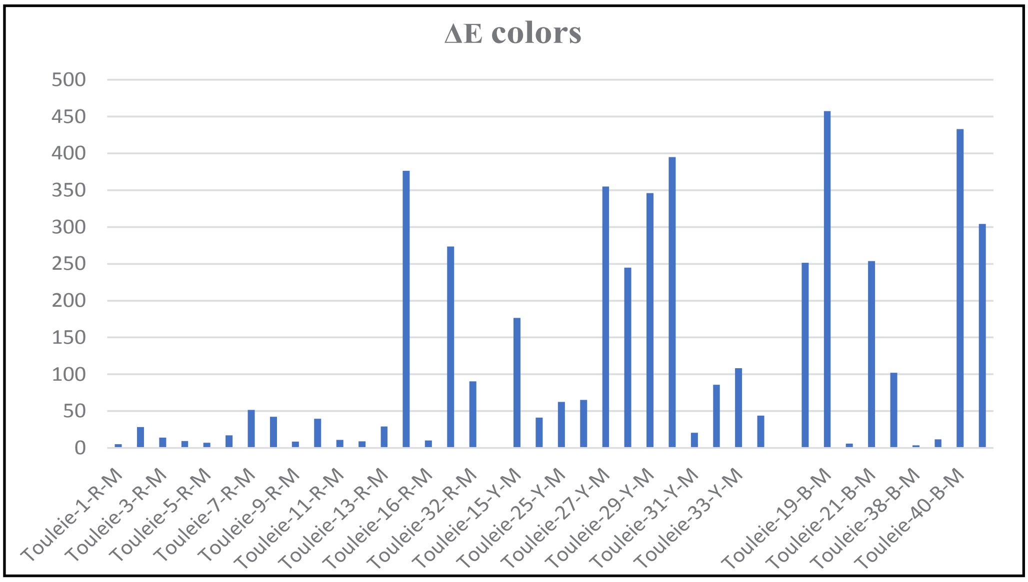

Fig. 9: ΔE color differences between the Munsell color chart and spectrophotometric measurements (Author, 2025).

Summary of Colorimetric Results

A comparison of the L*, a*, and b* color parameters across the three-color groups—red, buff, and brown—is summarized in Table 5. As shown in the table, the highest L* (lightness) values are observed in the buff group, whereas the lowest correspond to the brown group. In addition, the red group exhibits the highest a* values compared with the buff and brown groups. The brown group consistently shows the lowest values across all three parameters relative to the other two groups.

Table 5. Range of L, a, b color parameters for the three colors groups identified in the studied ceramics (Author, 2025).

Row	Color spectrum	*L range*	a range	b range
1	Red	42 – 64	13 – 25	16 – 28
2	Buff	59 - 75	5/5 – 13	19 – 27
3	Brown	36 - 54	2/5 – 7/5	8 – 16/5

The visible-range reflectance spectra (360–740 nm) of the three color groups reveal distinct patterns. The red ware group exhibits a shoulder in the region around 600 nm (Fig. 4). In contrast, the buff ware group shows a similar shoulder with a gentler slope at approximately 580 nm (Fig. 6). Unlike the first two groups, the brown ware group displays a shoulder in the region around 420 nm (Fig. 8). The slopes of these reflectance curves indicate that, at 740 nm, the red wares reach maximum reflectance values between 30% and 70%, while the buff wares range from 40% to 70%, and the brown wares fall within a narrower range of 15% to 35%. Furthermore, color naming and categorization are influenced by the semantic categories of the observer's native language [22] (Paramei et al., 2015: 159–170). In addition, visual comparison of colors with standard charts, such as the Munsell Soil Color Chart, may involve considerable discrepancies due to observer error. For these reasons, instrumental spectrophotometric methods can be considered more accurate for the colorimetric analysis of ceramic samples.

Chemical and Mineralogical Analysis of the Ceramics

Chemical Analysis

A total of 14 pigment samples taken from the surfaces of ceramics recovered from Tule'i Tepe in Khuzestan were analyzed non-destructively using portable X-ray fluorescence (pXRF). The resulting elemental data are summarized in Table 6.

Table 6. Elemental analysis of the ceramic bodies and surface color coatings from the Tule'i samples (Author, 2025).

Color	Element(%) Sample	Mg	Mn	Fe	K	Ca	Al	Si	S	Ti
Red	(body)R4	1.1	0.0	4.3	2.5	1.0	8.0	33.8	0.6	0.8
	(body)R5	2.1	0.1	5.3	2.5	3.7	7.7	36.9	0.3	0.6
	(body)R13	1.6	0.1	7.3	2.5	4.8	5.6	27.3	1.5	0.4
	(body)R14	1.5	0.1	6.4	2.0	5.0	4.2	31.2	0.8	0.4
	(body)R16	0.5	0.0	6.9	1.7	1.1	3.3	24.3	0.2	0.7
Buff	(body)Y15	2.2	0.1	5.9	1.3	4.8	5.5	34.4	0.4	0.6
	(body)Y25	1.2	0.1	4.6	2.0	9.3	5.1	26.3	0.3	0.4
	(body)Y28	1.5	0.1	5.0	1.6	11.0	4.0	22.8	0.2	0.5
	(body)R30	2.4	0.1	5.0	1.5	15.4	5.6	23.3	0.5	0.4
	(body)Y35	2.4	0.1	5.4	1.5	7.8	5.6	27.6	0.6	0.5
	(body)Y41	2.3	0.1	5.7	1.8	7.1	6.0	30.7	0.3	0.5
brown	(Motifs)B20	2.2	1.6	5.1	1.2	11.7	4.1	19.2	1.1	0.4
	Motifs) B38	1.0	4.4	5.2	1.0	8.5	4.1	17.2	1.5	0.3
	(Motifs)B41	2.3	0.8	5.6	1.1	8.4	4.8	28.9	0.9	0.5
	(Motifs)B41	1.2	1.1	5.5	1.1	8.0	4.4	27.2	1.0	0.5

The elemental analysis of the surface colors of the studied samples—both ceramic bodies and coatings—indicates that the concentrations of four elements (iron, manganese, calcium, and sulfur) show a significant correlation with the apparent color and the colorimetric results. Iron (Fe) was measured at 4–7% across all samples. Sulfur and manganese exhibit their highest concentrations in the brown samples, whereas the lowest calcium content is observed in the red samples. With the exception of sample R13, which has a reddish brown color and a sulfur content of 1.5%, the sulfur content in the red and buff samples remains below 1%, while the manganese content in these samples is approximately 0.1%. Manganese appears to be the primary element responsible for the dark and brown colors. The presence of high calcium compounds counteracts the red coloration produced by iron oxide, resulting in a lighter ceramic body (Ashurst and Ashurst, 2001: 48–49). Potassium, which acts as a fluxing agent in the brown coating of the ceramic samples, is present at approximately 1%, whereas in the buff and red samples this value ranges between 1.5% and 2.5%. Aluminium, the major constituent element of clay soils, is most abundant in the red bodies, followed by the buff bodies or coatings, and is lowest (approximately 4%) in the brown decorations.

Mineralogy

Mineralogical analyses were carried out on four ceramic samples (red and buff wares) using X ray diffraction (XRD). The results are summarized in Table 7 and the corresponding diffractograms (Fig. 10). Due to the limited quantity of black–brown pigments, XRD analysis of these decorations was not feasible.

Quartz and calcite were identified as the major phases in samples R4, R5, and Y28, whereas in sample R16 the principal phases were quartz and albite (a type of feldspar). The presence of pyroxene in samples R4 and Y28 suggests that these samples were fired at a higher temperature than the other two. However, the coexistence of pyroxene with calcite in the same samples indicates that the firing temperature could not have greatly exceeded the decomposition temperature of calcite, i.e., around 700 °C [24] (Khanzadi et al., 2021).

Phlogopite—a magnesium rich mica that ranges in color from brownish yellow to reddish brown and manganese bearing varieties, and is sometimes referred to as green mica—was identified in sample R4. This mineral forms at relatively high temperatures and occurs in dolomitic limestones; however, distinguishing it from biotite can be difficult [25] (Moezz Lesko, 2001: 131–132). Therefore, the biotite detected in sample R4 may in fact represent phlogopite. According to the literature, dolomitic compositions in the presence of silica form clino pyroxene like phases (Reaction 1), which transform into diopside (a pyroxene phase) at temperatures of approximately 500–600 °C (Reaction 2). Although pyroxenes are generally considered high temperature minerals, they may also form at lower temperatures in carbonate rich environments (Kharghani, 1988). The presence of non crystalline or amorphous silica, which is chemically more reactive than crystalline silica (quartz), plays a significant role in the complete sintering of dense ceramic bodies at temperatures around 600–700 °C. This phenomenon has also been documented in the bricks of Chogha Zanbil [27] (Hadian, 2015). Amorphous silica has been widely identified in riverine sediments such as those of the Dez River, and its presence in deposits from the Tule'i region is therefore plausible. As noted earlier, the high carbonate content in these ceramic bodies is responsible for the color shift from red to buff. In general, the studied ceramic bodies appear to have been well fired.

Equation 2: $CaCO3+SiO2 \longrightarrow CaSiO3 + CO2$

Equation 3: $CaMg(CO3)2+ SiO2 + CO2 \longrightarrow CaMgSi2O6$

Table 7. Mineralogical composition of the Tule'i ceramics determined by XRD analysis (Author, 2025).

Row	Sapmle	Detected phases
1	R4	Quartz, calcite, phlogopite, and diopside (pyroxene)
2	R5	Quartz, calcite, biotite
3	R16	Quartz and albite
4	Y28	Quartz, calcite, and pyroxene

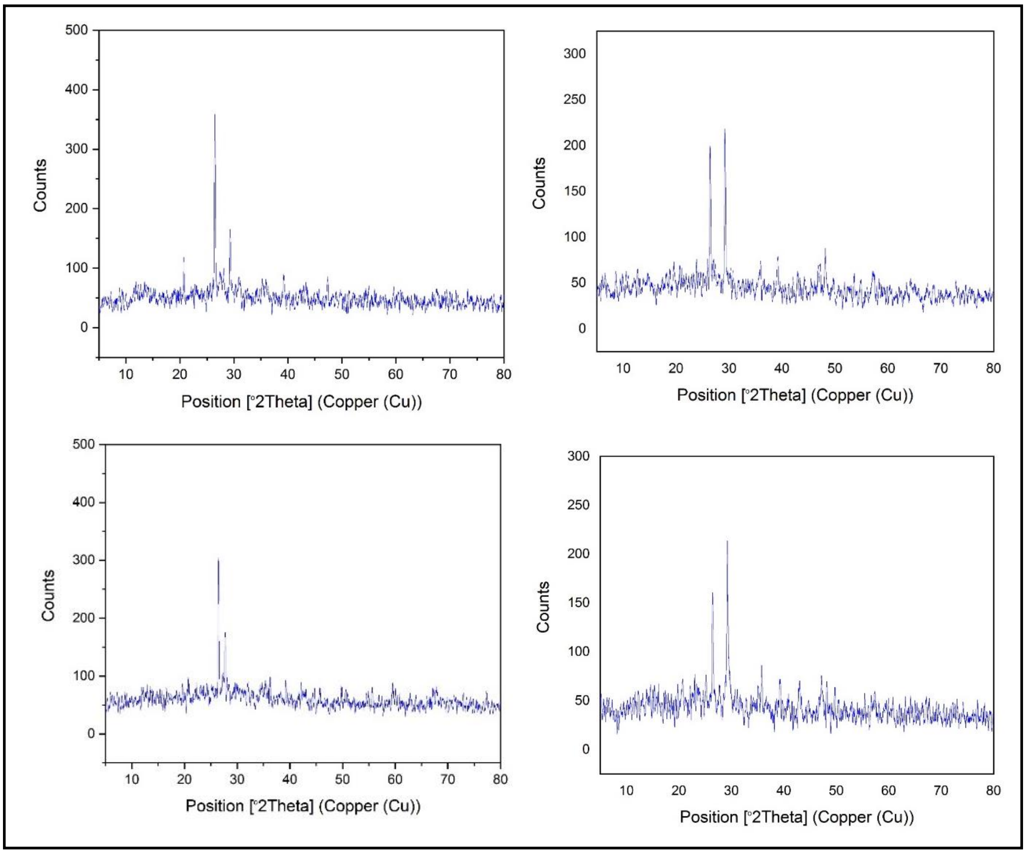

Fig. 10: X-ray diffraction (XRD) patterns of samples R4 (top-left), R5 (top-right), R16 (bottom-left), and R28 (bottom-right), (Author, 2025).

Conclusion

Based on the present investigation, a significant correlation is observed between the chemical composition and the apparent color of the ceramics. Manganese (Mn) plays the primary role in producing the brown coloration. Furthermore, sulfur (S) is more frequently detected in the brown coatings. The presence of high calcium compounds counteracts the red coloration produced by iron oxide, resulting in a lighter ceramic body. Accordingly, the highest Fe and Al contents occur in the red bodies, whereas the buff bodies exhibit the highest calcium concentrations. Differences in firing temperature among certain samples are inferred from the presence of specific minerals, such as diopside (a pyroxene mineral), which forms at higher temperatures. The presence of this phase suggests that the ceramic bodies were fired at temperatures around 600–700 °C. However, the persistence of calcite in the bodies indicates that the firing temperature did not exceed 800 °C. The occurrence of phlogopite, a high temperature mineral, is also consistent with dolomitic compositions that facilitate the formation of pyroxene.

The observed differences and similarities in the color parameters and reflectance spectra of the three ceramic groups can serve as criteria for the accurate comparison and identification of archaeological ceramic colors, complementing conventional non instrumental methods such as the Munsell Soil Color Chart. The color discrepancies between spectrophotometric and Munsell based assessments are illustrated in Figure 9. These differences are minimal for red hues but more pronounced for buff and brown colors, largely due to human visual error. Additionally, the reflectance spectra allow for a more refined classification of the ceramics. This technique can also serve as an auxiliary method for the relative comparison and provenance assessment of ceramics based on their colorimetric properties.

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Author Contribution

The contribution of all authors to the writing of the article has been equal.

Conflict of Interest

In adherence to ethical publication standards, the authors affirm that there are no conflicts of interest, either personal or financial, that could have influenced the content or conclusions presented in this research.

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تحلیل‌های رنگ‌سنجی، تجزیهٔ شیمیایی و کانی‌شناسی سفال‌های محوطهٔ باستانی توله‌ای خوزستان: رویکردی چندرشته‌ای در مطالعهٔ مواد باستان‌شناختی

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چکیده

تپه توله‌ای، واقع‌در شمال خوزستان، یکی از محوطه‌های مهم دورهٔ نوسنگی دشت شوشان است که براساس مطالعات پیشین در فاصلهٔ حدود ۶۲۰۰ تا ۵۹۰۰پ.م. تاریخ‌گذاری شده است. این محوطه به‌عنوان یک استقرار فصلی یا اردوگاه کوچ‌روان با گاهنگاری نسبتاً کوتاه شناخته می‌شود؛ اما از نظر تنوع مواد فرهنگی، به‌ویژه سفال، بسیار غنی است. مجموعه سفال‌های به‌دست‌آمده شامل سفال‌های گونهٔ جعفر منقوش و ساده، و نیز سفال‌های سفید و قرمز با نقوش متنوع است که ازنظر سبکی با محوطه‌های هم‌عصر در بین‌النهرین و جنوب‌غرب ایران شباهت دارند. باوجود اهمیت این محوطه در مطالعات نوسنگی و کوچ‌روی، تاکنون پژوهش جامعی دربارهٔ ترکیب مواد، فناوری ساخت و منشأ احتمالی سفال‌های آن انجام نشده بود. پژوهش حاضر با رویکردی میان‌رشته‌ای به مطالعهٔ ۴۱ نمونه سفال از تپه توله‌ای پرداخته و از سه روش تحلیلی مکمل شامل طیف‌سنجی نوری مرئی (Visible Spectrometry) برای سنجش رنگ، طیف‌سنجی فلورسانس اشعهٔ ایکس قابل‌حمل (pXRF) برای تعیین ترکیب عنصری و پراش اشعهٔ ایکس (XRD) برای شناسایی فازهای کانی‌شناسی و تخمین دمای پخت استفاده کرده است. نتایج نشان می‌دهد که بدنه‌های نخودی بیشترین میزان کلسیم (CaO) را دارند، درحالی‌که بدنه‌های قرمز از غلظت بالاتری از آهن (Fe_2O_3) و آلومینیم (Al_2O_3) برخوردارند؛ هم‌چنین منگنز (MnO) عامل اصلی ایجاد رنگ قهوه‌ای در نقوش سفال‌ها تشخیص داده شد. داده‌های XRD حضور کانی‌هایی مانند پیروکسن و فلوگوپیت را نشان می‌دهد که بیانگر دمای پخت حدود ۶۰۰ تا ۷۰۰ درجهٔ سانتی‌گراد است، درحالی‌که وجود کلسیت در برخی نمونه‌ها نشان می‌دهد دمای پخت از ۸۰۰ درجهٔ سانتی‌گراد فراتر نرفته است. این نتایج حاکی از دانش فنی قابل‌توجه سفالگران در کنترل دما و انتخاب مواد اولیه است. هم‌چنین مقایسهٔ دو روش رنگ‌سنجی نشان‌داد که طیف‌سنجی بازتابی در تفکیک طیف‌های رنگی نزدیک، به‌ویژه میان رنگ‌های نخودی و قهوه‌ای روشن، دقت بیشتری داشته و می‌تواند به‌عنوان روشی مکمل در مطالعات سفال‌شناسی به‌کار رود.

کلیدواژگان: دورهٔ نوسنگی، سفال توله‌ای، اسپکتروفوتومتر، کانی‌شناسی، تجزیه عنصری، جدول رنگ مانسل.

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