Composition Analysis of Glass Based on Weighted Mean and Correlation Analysis

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Abstract. Due to weathering and other external influences, the surface and chemical composition of ancient cultural relics will be corroded to a certain extent. This paper analyzes the chemical composition of two main kinds of ancient glass, high potassium glass and lead barium glass, classifies the cultural relic samples according to the type and weathering degree, and studies the chemical composition content rule of weathered samples respectively through descriptive statistics. The weighted average was used to predict the chemical constituents before weathering, and the correlation analysis of the main constituents was carried out. The composition analysis of ancient cultural relics in this paper lays a foundation for further effective measures to protect and restore cultural relics.

Keywords: Ancient glass, weathering, weighted average, correlation analysis.

1. Introduction

Glassware first appeared in the Spring and Autumn Period and the Warring States Period. It was an important commodity and link for trade along the Silk Road [1]. The identification of the composition of ancient glass products is not only the important historical materials in the study of Chinese ancient handicraft manufacturing history, but also beneficial to take effective protection measures of cultural relics. Based on the chemical composition data of ancient glass, this paper analyzes whether the cultural relics have weathered chemical composition content law, predicts the chemical composition content before weathering according to the data of detection points, and analyzes the correlation between the components.

2. Methods and models

2.1. Data classification and statistical analysis

As glass cultural relics composed of different chemical elements are affected by weathering to different degrees and chemical compositions, the data are first arranged according to category and weathering degree and divided into four categories: before weathering of high-potassium glass, after weathering of high-potassium glass, before weathering of lead barium glass, and after weathering of lead barium glass. The average proportion of each component and the data difference before and after weathering were analyzed [2].

2.2. Normal distribution function was used to predict the composition content before weathering

Due to the different effects of weathering on different types of glass, the prediction of chemical composition contents of weathering points of different types before weathering is not the same, which should be divided into high potassium glass and lead barium glass to solve respectively. According to the analysis of the data, there is only the composition before or after weathering of a certain weathering point in the data, but there is no specific sample composition change in time series [3].

On the basis of data preprocessing, weighted average and Gaussian process regression fitting were used to analyze and predict, and the results were compared [4].

Firstly, weighted average is used for preliminary judgment and solution: 14 chemical component variables are defined as $x_{1j}, x_{2j}, x_{3j}, \ldots\ldots x_{14j}$, taking x_{1j} as an example, represents the proportion of the *j*th data of a chemical component. The variables of weight of each chemical component are $w_{1j}, w_{2j}, w_{3j}, \ldots\ldots w_{14j}$, taking w_{1j} as an example, represents the weight of the *j*th data in a chemical component. Because the data need to be divided into four groups of high potassium weathering, high potassium weathering without weathering, lead barium weathering and lead barium weathering without weathering, the value of j is different for different groups. For the high potassium weathering group, $= 1, 2,3, \ldots\ldots 7$. For the high potassium non-weathering group, $j = 1,2,3, \ldots\ldots 11$. For lead-barium weathering group, $j = 1,2,3, \ldots\ldots 28$; For the high potassium weathering group, $j = 1,2,3, \ldots\ldots 21$. The $x_{1j}, x_{2j}, x_{3j}, \ldots\ldots x_{14j}$ is substituted into the normal distribution function respectively to calculate the weight of each chemical component. The calculation formula is as Eq. (1) follows:

$$f(x) = \frac{1}{\sqrt{2\pi}\sigma} e^{-\frac{(x-\mu)^2}{2\sigma^2}} \tag{1}$$

The calculated weights $w_{1j}, w_{2j}, w_{3j}, \ldots\ldots w_{14j}$ and $x_{1j}, x_{2j}, x_{3j}, \ldots\ldots x_{14j}$ can be multiplied separately to obtain the weighted data, so that the weighted average of each column of data can be calculated. Taking silica, the first chemical component of the high potassium weathering group, as an example, the calculation results are shown in Formula 2. The weighted average of all components is obtained, and the ratio between the measured value and the actual value after weathering should be equal to the ratio between the weighted average without weathering and the weighted average after weathering, so as to obtain the prediction of the composition before weathering (Eq. (2)).

$$\bar{x}_1 = \frac{x_{1,1}w_{1,1} + x_{1,2}w_{1,2} + x_{1,3}w_{1,3} + \cdots + x_{1,7}w_{1,7}}{w_{1,1} + w_{1,2} + w_{1,3} + \cdots + w_{1,7}} \tag{2}$$

2.3. Correlation analysis

2.3.1 Pearson correlation analysis

The content data of each component are continuous variables and can be compared in pairs. The normal value of data after processing; There was a linear correlation between the data. It follows normal distribution, so Pearson correlation coefficient can be used to analyze the correlation between 14 chemical components, such as silica and sodium oxide. The method is efficient and the quantitative analysis is objective and practical [5].

Pearson correlation coefficient describes the linear correlation between two variables. Its value is between -1 and 1, and is generally expressed by r. When r is greater than 0, it indicates that there is a positive correlation between two variables. The closer it is to 1, the more linearly correlated the two variables are. The formula is as Eq. (3) follows:

$$r = \frac{\sum_{i=1}^{n}(X_i - \bar{X})(Y_i - \bar{Y})}{\sqrt{\sum_{i=1}^{n}(X_i - \bar{X})^2}\sqrt{\sum_{i=1}^{n}(Y_i - \bar{Y})^2}} \tag{3}$$

2.3.2 Grey correlation analysis

Due to the grey characteristics of each chemical component, grey correlation analysis was used to find out the most closely related components [6].

3. Result analysis

3.1. Differences of components before and after weathering

SiO_2 in high potassium glass is at a high level both in weathering and without weathering, which has the greatest influence on the composition of high potassium glass. After weathering, the proportion increases, indicating that weathering has little effect on silica composition in high-potassium glass or is not easy to be weathered. In Figure 1B, it can be seen that SiO_2, PbO and BaO have the greatest influence on the chemical composition of lead-barium glass, while silica has the greatest reduction degree, indicating that it is most affected by the degree of weathering. However, the proportion of PbO and BaO increases after weathering, indicating that weathering has little effect on PbO and BaO components in lead-barium glass.

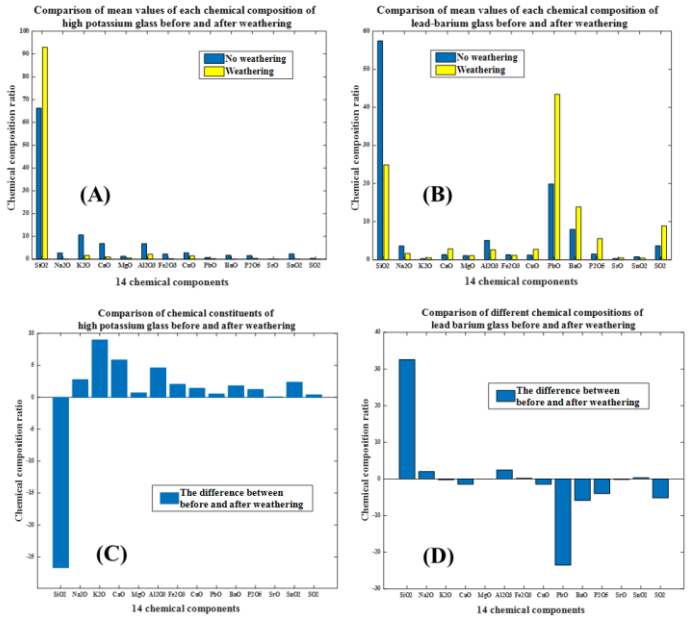


Fig. 1 Differences in chemical compositions of different types of glass before and after weathering. (A) Average values of chemical compositions of high-potassium glass and (B) lead-barium glass before and after weathering. (C) Differences of components of high-potassium glass and (D) Lead-barium glass before and after weathering

3.2. Components of glass before weathering

Due to the different effects of weathering on different types of glass, the prediction of the chemical composition content of weathering points of different types before weathering is different. In this paper, the glass is divided into high potassium glass and lead barium glass for prediction respectively.

The results of weighted averages are shown in Table 1. Furthermore, taking the weathered samples of high-potassium glass as an example, the prediction results obtained by weighted average and Gaussian regression are shown in Table 2 and 3 (the case of no detected components after weathering is 0) [7].

Table. 1 The result of a weighted average

Glass type	Weathering type	SiO_2	Na_2O	K_2O	CaO	MgO	Al_2O_3	Fe_2O_3	CuO	PbO	BaO	P_2O_5	SrO	SnO_2	SO_2
High potassium	N	65.2107	0.4755	9.7175	5.6792	1.1559	6.5839	1.9515	2.4857	0.3015	0.4408	1.2314	0.0383	0.0965	0.0711
	W	92.7805	0.0000	0.8548	0.9345	0.1017	2.0314	0.2221	1.2940	0.0161	0.0000	0.3020	0.0000	0.0000	0.0000
Lead barium	N	55.4783	1.5033	0.1893	1.1332	0.6813	4.3829	0.3216	1.0219	21.1154	8.0052	0.5067	0.2465	0.0457	0.0782
	W	24.9244	0.0877	0.1030	2.6464	0.5270	2.5099	0.5437	1.9401	42.8353	11.2130	4.5813	0.4283	0.0244	0.6408

Note: N represents no weathering, W represents weathering.

Table. 2 Composition prediction of high potassium glass before weathering

Site	SiO_2	Na_2O	K_2O	CaO	MgO	Al_2O_3	Fe_2O_3	CuO	PbO	BaO	P_2O_5	SrO	SnO_2	SO_2
03 No. 1	61.1830	0.0000	58.9982	12.2157	0.0000	13.1589	0.0000	1.4983	4.6739	0.0000	2.6909	0.0000	0.0000	0.0000
07	65.1049	0.0000	0.0000	6.5029	0.0000	6.4174	1.4939	6.2239	0.0000	0.0000	2.4870	0.0000	0.0000	0.0000
09	66.7848	0.0000	6.7069	3.7680	0.0000	4.2783	2.8121	2.9775	0.0000	0.0000	1.4270	0.0000	0.0000	0.0000
10	68.0147	0.0000	10.4582	1.2763	0.0000	2.6253	2.2848	1.6136	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
12	66.2717	0.0000	11.4813	4.3758	0.0000	4.7320	2.5484	3.1696	0.0000	0.0000	0.6116	0.0000	0.0000	0.0000
22	64.9082	0.0000	8.4121	10.0886	7.2717	11.3439	3.0757	1.0565	0.0000	0.0000	0.8562	0.0000	0.0000	0.0000
27	65.1682	0.0000	0.0000	5.7128	6.1355	8.1352	1.7575	2.9583	0.0000	0.0000	1.4678	0.0000	0.0000	0.0000

Table. 3 Composition prediction of lead-barium glass before weathering

Site	SiO_2	Na_2O	K_2O	CaO	MgO	Al_2O_3	Fe_2O_3	CuO	PbO	BaO	P_2O_5	SrO	SnO_2	SO_2
24	71.0941	0.0000	0.0000	0.2013	0.0000	2.7766	0.0000	4.4562	14.3644	18.7263	0.0155	0.5238	0.0000	0.0000
30 No.1	76.4362	0.0000	2.5929	1.9226	1.2670	7.5964	1.2540	0.0000	19.3332	7.3463	0.0000	0.2014	0.7495	0.0000
30No. 2	82.2012	0.0000	0.0000	1.8156	0.6594	6.7407	1.6207	0.0000	18.6037	7.3892	0.1560	0.2763	0.8245	0.0000
02	80.7543	0.0000	1.9309	1.0020	1.5256	10.0062	1.1002	0.1370	23.3803	0.0000	0.3949	0.1094	0.0000	0.0000

08	44.8289	0.0000	0.0000	0.6337	0.0000	2.3400	0.0000	5.4834	14.1376	22.2960	0.3971	0.2130	0.0000	0.3149
08	10.2612	0.0000	0.0000	1.3660	0.0000	1.9384	0.0000	1.6540	15.9960	21.8605	0.8362	0.3050	0.0000	1.8347
11	74.7668	0.0000	0.3862	1.5030	0.9179	4.6975	0.0000	2.5968	12.5158	10.4305	1.0375	0.2130	0.0000	0.0000
19	65.9746	0.0000	0.0000	1.2546	0.7628	6.2342	0.7867	1.8489	21.1078	3.8195	0.9766	0.1094	0.0000	0.0000
26	44.0498	0.0000	0.0000	0.6166	0.0000	1.2224	0.0000	5.5676	14.5566	23.0242	0.3462	0.2590	0.0000	0.2393
26	8.2802	0.0000	0.7356	1.2889	0.0000	2.0606	0.0000	1.8963	14.7489	25.3087	0.6680	0.3568	0.0000	1.9470
34	79.6414	0.0000	0.4597	0.3340	0.0000	2.8290	0.2780	0.7954	22.9465	7.1393	0.0376	0.1266	0.0000	0.0000
36	88.0774	38.0365	0.2575	0.1584	0.0000	2.7941	0.1893	0.3582	20.5114	7.7318	0.0077	0.1266	0.0000	0.0000
38	73.2977	23.6443	0.0000	0.2912	0.0000	4.4880	0.1715	0.3845	24.3070	6.9894	0.0531	0.2360	0.0000	0.0000
39	58.4289	0.0000	0.0000	0.4753	0.0000	0.8731	0.0000	0.4635	30.0843	5.1546	0.1283	0.3511	0.0000	0.0000
40	37.1942	0.0000	0.0000	0.8007	0.0000	0.7858	0.1124	0.0000	34.6095	4.7762	0.1958	0.3914	0.0000	0.0000
41	41.0894	0.0000	0.8091	2.1239	3.5296	5.8151	1.0588	0.1001	21.7487	6.9679	0.8251	0.2705	0.0000	0.0000
43 No.1	27.6230	0.0000	0.0000	2.2438	1.1507	3.9291	0.4496	2.8181	29.5027	5.2045	0.0000	0.3684	0.0000	0.0000
43 No. 2	48.3012	0.0000	0.0000	2.7405	1.2282	5.9548	0.8222	0.7954	22.0592	2.3274	1.4190	0.2705	0.0000	0.0000
49	64.0826	0.0000	0.0000	1.9612	1.9005	9.3950	1.6207	0.3687	16.8488	4.3550	1.2277	0.2648	0.0000	0.0000
50	40.0210	0.0000	0.0000	1.3660	0.6077	3.2656	0.1952	0.5952	21.6895	10.1378	0.7012	0.3799	0.0000	0.0000
51 No.1	54.7785	0.0000	0.0000	1.5330	1.5385	9.1680	0.7039	0.7216	19.8360	6.3825	0.8959	0.2245	0.8807	0.0000
51 No.2	47.5222	0.0000	0.0000	2.1967	1.8747	4.3832	0.2484	0.3951	25.3077	0.0000	0.9678	0.0000	0.0000	0.0000
52	57.2937	20.9030	0.0000	0.9720	0.7111	2.0257	0.1360	0.3687	23.3754	6.1683	0.6315	0.2532	0.0000	0.0000
54	49.5922	0.0000	0.5885	1.3660	1.6549	7.2471	0.0000	0.4372	27.3386	5.0261	0.4690	0.5065	0.0000	0.0000
54	38.0845	0.0000	0.0000	0.0000	1.4351	6.3740	0.0000	0.7058	28.8175	0.0000	1.5628	0.6446	0.0000	0.0000
56	64.8839	0.0000	0.0000	0.5181	0.0000	3.2306	0.0000	0.4161	20.3339	11.0302	0.2809	0.0000	0.0000	0.0000
57	56.5815	0.0000	0.0000	0.5609	0.0000	3.8069	0.0000	0.6110	22.2317	12.3509	0.0000	0.0000	0.0000	0.0000
58	67.6440	0.0000	0.6252	1.4944	1.0214	6.1469	0.5087	1.6487	19.3973	5.4687	0.9943	0.1381	0.0000	0.0000

3.3. Correlation between chemical components

As shown in Figure 2, the correlation heat map of different glass components shows that after weathering of high-potassium glass, silica has strong correlation with six components such as ferric oxide, and it has strong negative correlation with K_2O, CaO, Al_2O_3, PbO and P_2O_5, and strong positive correlation with Fe_2O_3. K_2O has a strong positive correlation with PbO [8].

Before weathering of high potassium glass, SiO has a strong negative correlation with K_2O and CaO, and CaO has a strong negative correlation with P_2O_5, SrO and SnO_2.

After weathering of lead-barium glass, SiO_2 is mainly negatively correlated with BaO, SrO and SO_2. After weathering, the content of SO_2 increased, and the original detection point detected little SO_2, which led to a large increase in the proportion of SO_2, which confirmed the analysis in question 1 and could explain the result of the correlation between weathering of lead barium glass and SO_2. PbO is mainly negatively correlated with CuO and BaO. In addition, BaO has a strong negative correlation with MgO, Al_2O_3 and Fe_2O_3.

Before the weathering of lead-barium glass, SiO_2 is mainly negatively correlated with CuO, BaO and P_2O_5. PbO mainly has a strong negative correlation with Al_2O_3 and K_2O, BaO has a strong negative correlation with CaO and a strong positive correlation with CuO.

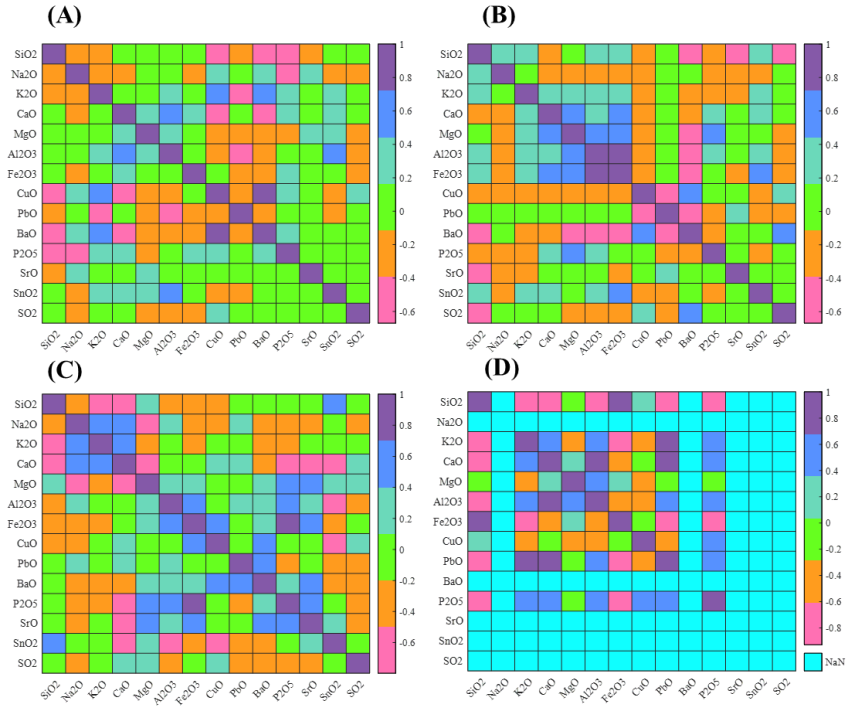


Fig. 2 Correlation heat map of different types of glass composition. A-D represent lead barium weathering type glass, lead barium unweathered type of glass, high potassium weathered type glass, and High potassium unweathered type of glass, respectively.

Since SiO_2 is the main component of both types of glass, SiO_2 is used as a reference sequence. Firstly, the averaging method was used to de-dimensionalize[9], SiO_2 was used as the parent sequence, other chemical components were used as the sub-sequence evaluation item, the resolution coefficient was set at 0.50, and the correlation degree ranking with SiO_2 was obtained as shown in Table 4.

After weathering of high potassium glass, SiO_2 is strongly correlated with Fe_2O_3. It is basically consistent with the results of Pearson correlation coefficient, and makes a certain supplement to its correlation size. When high potassium glass is not weathered, SiO_2 is mainly related to 6 kinds of oxides such as Al_2O_3. The relationship between SiO_2 and PbO, Al_2O_3 and CaO is strong after weathering of lead-barium glass. AiO was strongly associated with PbO and BaO before weathering of lead-barium glass. Comprehensive analysis shows that SiO_2 has extensive correlation with other chemical components.

Table. 4 Correlation degree ranking with SiO2 of different components

High potassium	A	Component	Fe_2O_3	CuO	Al_2O_3	CaO	P_2O_5	K_2O	MgO	PbO	Na_2O	SO_2	SnO_2	SrO	BaO
		Correlation degree	0.903	0.896	0.883	0.88	0.861	0.804	0.7	0.696	0	0	0	0	0
	B	Component	Al_2O_3	K_2O	MgO	CaO	CuO	Fe_2O_3	P_2O_5	SrO	PbO	BaO	SnO_2	SO_2	Na_2O
		Correlation degree	0.941	0.941	0.926	0.918	0.91	0.91	0.882	0.846	0.819	0.804	0.788	0.781	0.779
Lead barium	A	Component	PbO	Al_2O_3	CaO	SrO	BaO	MgO	P_2O_5	Fe_2O_3	CuO	K_2O	Na_2O	SnO_2	SO_2
		Correlation degree	0.944	0.933	0.912	0.904	0.901	0.883	0.882	0.872	0.868	0.846	0.824	0.823	0.818
	B	Component	PbO	BaO	Al_2O_3	SrO	MgO	CaO	K_2O	CuO	Na_2O	Fe_2O_3	P_2O_5	SO_2	SnO_2
		Correlation degree	0.968	0.956	0.948	0.943	0.94	0.939	0.937	0.92	0.905	0.904	0.901	0.882	0.881

Note: A means after weathering, B means before weathering.

The correlation difference between silica and different components in different types of glass is shown in Figure 3. Before and after the weathering of high potassium glass, the relationship between the chemical constituents of Na_2O and BaO and SiO_2 is significantly different, and the relationship between the weathering of high potassium glass and the weathering of lead barium glass is significantly different. It can be seen that the relationship between the constituents of high-potassium glass after weathering is weaker than that of lead-barium glass [10].

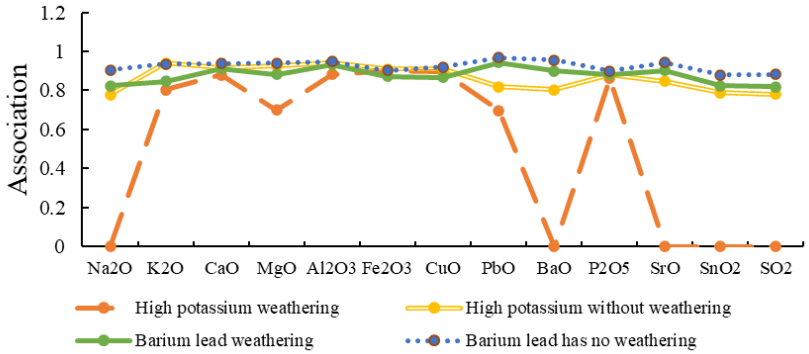


Fig. 3 Correlation difference between SiO_2 and different components of different types of glass

4. Conclusion

In this paper, the chemical composition of two kinds of ancient glass high potassium glass and lead barium glass was analyzed, and the chemical composition content rule of weathered samples was studied respectively through descriptive statistics. The weighted average was used to predict the chemical constituents before weathering, and the correlation analysis of the main constituents was carried out. The composition analysis of ancient cultural relics in this paper lays a foundation for further effective measures to protect and restore cultural relics.

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