Composition analysis and identification of ancient glass products based on gray correlation

Shubo Chang [#,*], Yindong Yang [#], YangzhenHua Xu [#], Jisheng Chen [#]

Hainan University, Haikou, China, 570228

* Corresponding Author Email: chang011229@163.com

[#]These authors contributed equally.

Abstract. The study of ancient glass, as an important branch of scientific and technological archaeology, is an important physical material for exploring the economic, technological and cultural exchanges between China and foreign countries on the Silk Road. It is of great significance for the study of the development of ancient societies and cultural exchanges between China and foreign countries. This paper focuses on the composition analysis and identification of ancient glass products, based on data mining and software analysis, giving the weathering pattern of glass surface, predicting the chemical composition content before weathering, as well as cluster analysis and correlation analysis of different categories of glass and their chemical composition. We also concluded that the composition of silica, potassium oxide and barium oxide in high potassium glass artifacts changed to a great extent before and after weathering, while the composition of silica and lead oxide in lead-barium glass artifacts changed to a great extent, and finally, we constructed a multiple linear regression model for prediction by the data of each chemical composition at the weathering point, and conducted F-test on the model to prove that the prediction models of silica, potassium oxide, barium oxide and lead oxide had a good performance. The F-test of the model proved the reasonableness of the prediction models for silica, potassium oxide, barium oxide, and lead oxide. It can be concluded that the glass type has an influence on the chemical composition of glass, so the glass can be divided into two categories of glass: high potassium and lead-barium, and then the chemical composition of these two categories of glass were analyzed separately by gray correlation analysis to obtain the top five correlation ranking of the chemical composition of each category of glass, and then the differences in the chemical composition of these two categories of glass were compared.

Keywords: N Glass composition, multiple linear regression, F-test grey correlation analysis.

1. Introduction

The main raw material of glass is quartz sand, whose main chemical composition is silicon dioxide (SiO2) [1]. The main chemical composition differs depending on the flux added during calcination [2]. For example, lead-barium glass, which has a high content of lead oxide (PbO) and barium oxide (BaO) when lead ore is added as a flux during the firing process, is usually considered as our own invented glass variety, and potassium glass, which is made by firing substances with high potassium content such as grass ash as a flux, is mainly popular in Lingnan, China, and other regions in Southeast Asia and India [3]. The raw material formulations and the corresponding equipment and processes do not allow ancient glass to differ in performance, texture, color, and luster. To study the formula of ancient glass, it is necessary to observe the ancient glass objects, examine the documentary archives, and understand the conditions of its excavation or collection, as well as to take a comprehensive analysis of the laboratory formula. Among them, the most effective method is the laboratory analysis of ancient glass fragments. The purpose of this study is to address the surface weathering of glass artifacts and its relationship with the type of glass, decoration and color, as well as different types of glass artifacts samples with and without the weathering of the surface chemical content of the statistical law, predicting its chemical content before weathering. For different categories of glass artifact samples, the correlations between their chemical compositions were analyzed and the variability of chemical composition correlations between different categories was compared [4].

2. Data sources and methods

2.1. Data sources

The data used in this paper comes from the National Student Mathematical Modeling Contest 2022 C (http://www.mcm.edu.cn/html_cn/node/5267fe3e6a512bec793d71f2b2061497.html), which has been measured experimentally and accurately, and its authenticity, accuracy is maintained with high precision.

2.2. Method Introduction

Linear regression is a statistical analysis method that uses regression analysis in mathematical statistics to determine the quantitative interdependent relationship between two or more variables [5]. In linear regression analysis, only one independent variable and one dependent variable are included, and the relationship between them can be approximated by a straight line, and this regression analysis is called univariate linear regression analysis. If the regression analysis includes two or more independent variables and the relationship between the dependent variable and the independent variable is linear, it is called multiple linear regression analysis [6].

The equation of the multiple linear regression model is as follows

$$y_i = \beta_{0i} + \beta_1 H_{1i} + \beta_2 H_{2i} + \cdots + \beta_{13} H_{13i} + \beta_{14} H_{14i} + \xi_i \tag{1}$$

where $\beta_0 \ldots \beta_{14}$ is the linear regression coefficient, ξ_1_1 is the random error term, and y_i is the predicted result of a certain chemical component, (i=1,2...14).

In fact, a phenomenon is often associated with multiple factors, and the optimal combination of multiple independent variables together to predict or estimate the dependent variable is more effective and realistic than using only one independent variable for prediction or estimation. Therefore, multiple linear regression is more practical than one-dimensional linear regression. Using a regression model, a unique result can be calculated by standard statistical methods as long as the model and data used are the same.

Grey system theory was proposed by a famous scholar, Professor Deng Julong, in 1982 [7], which requires a small amount of data and is widely used in many scientific fields, where Grey Relation Analysis (GRA) is a multi-factor statistical analysis method [8]. To put it simply, we want to know the relative strength of a certain item in a gray system, which is influenced by other factors, and to put it more bluntly, we assume and know that a certain indicator may be related to some other factors, then we want to know which factor is relatively more related to the other factors, and which factor is relatively weaker. By analogy, ranking these factors and getting an analysis result, we can know which of the factors are more related to this indicator that we are concerned about. [9]

The formula of the gray correlation calculation model is as follows

$$\eta_i(j) = \frac{\min\limits_{1\le s\le 56}\min\limits_{1\le t\le 14}|h_0(t)-h_s(t)| + \rho \min\limits_{1\le s\le 56}\min\limits_{1\le t\le 14}|h_0(t)-h_s(t)|}{|h_0(j)-h_i(j)| + \rho \min\limits_{1\le s\le 56}\min\limits_{1\le t\le 14}|h_0(t)-h_s(t)|} \tag{2}$$

$$i = 1,2,\ldots,56, j = 1,2,\ldots,14$$

where $\eta_i(j)$ is the coefficient that compares the series h_i to determine the reference sequence h_0 on the jth indicator.,where $\rho\epsilon[0,1]$ is the resolution factor.,In this model, the resolution factor is chosen as 0.5,where $\min\limits_{1\le s\le 56}\min\limits_{1\le t\le 14}|h_0(t)-h_s(t)|$、$\rho \min\limits_{1\le s\le 56}\min\limits_{1\le t\le 14}|h_0(t)-h_s(t)|$ The two levels of minimum difference and two levels of maximum difference, respectively.

Solving for gray correlation values.

$$d_i = \sum_{k=1}^{14} w_k \eta_i(k) \tag{3}$$

Where $\boldsymbol{d_i}$ is the gray weighted correlation of the i-th chemical component to the ideal object, w_k is the weight corresponding to the j-th type of artifact, here we regard each chemical component as equal weight, so the weight w_k is 1/14.

Gray correlation analysis is done according to the development trend, so the sample size requirement is very low, and does not require a typical distribution law, and the calculation is relatively small, the results will be more consistent with the qualitative analysis results. Therefore, gray correlation analysis is a relatively simple and reliable analysis method in system analysis.

3. Model building and solving

3.1. Model Building

The large data prediction model for the user's electricity consumption is implemented in the Clementine software.

The relationship between surface weathering and glass type, ornamentation and color of glass artifacts was investigated because surface weathering and glass type, ornamentation and color are definite class variables, and because the chi-square test is a non-parametric test and is mainly used to compare the association analysis of two or more definite class variables, we used the chi-square test to derive the relationship between each indicator and the surface weathering of glass artifacts. Then, by classifying the surface weathering of the artifact samples and combining them with the glass type, we analyze the pattern of chemical composition content, and finally, based on the weathering point data, which is the percentage of chemical composition of each glass artifact, we establish a multiple linear regression model to predict the chemical composition content before weathering. The chi-square test is a statistical method proposed by Pearson to determine whether the actual probability matches the expected probability by comparing the chi-square statistic and the probability value of the chi-square distribution function at a certain confidence level and degree of freedom, and the correlation between two categorical variables can be tested by comparing the degree of agreement between the theoretical probability and the actual probability, so we separately conducted the chi-square test for the correlation between the weathering of the surface of glass artifacts and glass type, decoration, and color in order of chi-square test [10].

By using Excel to draw conclusions based on the comparative analysis of each chemical composition before and after weathering on the glass surface based on glass type, and then predict the pre-weathering chemical composition content based on the weathering point detection data, based on hypothesis 1 that the difference between each composition before weathering at each point on the surface of the artifact is not significant, so we can select the 14 chemical composition content of the artifact after weathering to build a multiple linear regression model and use it to predict the pre-weathering The data of one chemical composition before weathering were predicted, and so on, until all 14 chemical compositions were predicted. The data were then combined and divided into two groups: surface weathered and unweathered, and then the data were imported into SPSSPRO software to build a linear regression model using the weathered chemical composition data and the unweathered chemical composition data as the predicted object.

Dimensionless processing for the data, which are filled with zeros here because the given data are missing.

Determining the reference series h_0 and the comparison series h_i.

The reference sequence uses the Excel table function to find the sum of the percentages between the chemical components $h_0 = \{h_0(j)|j = 1,2,\dots,14\}$, Compare sequences.

The grey correlation coefficients between the characteristic series were determined, and the comparison series were the chemical composition of each glass artifact $h_i = \{h_i(j)|j = 1,2,\dots,14\}, i = 1,2,\dots,56$

$$\eta_i(j) = \frac{\min\limits_{1\le s\le 56}\min\limits_{1\le t\le 14}|h_0(t) - h_s(t)| + \rho \min\limits_{1\le s\le 56}\min\limits_{1\le t\le 14}|h_0(t) - h_s(t)|}{|h_0(j) - h_i(j)| + \rho \min\limits_{1\le s\le 56}\min\limits_{1\le t\le 14}|h_0(t) - h_s(t)|} \quad (4)$$

$$i = 1,2,\dots,56, j = 1,2,\dots,14$$

Where is $\eta_i(j)$ Comparing the series h_i to determine the coefficient of the reference series h_0 on the jth indicator, the $\rho\epsilon[0,1]$ is the resolution factor, In this model, the resolution factor is chosen

to be 0.5, $\min_{1\le s\le 56}\min_{1\le t\le 14}|h_0(t)-h_s(t)|$、$\rho\min_{1\le s\le 56}\min_{1\le t\le 14}|h_0(t)-h_s(t)|$ The two levels of minimum difference and two levels of maximum difference respectively.

Solving for gray correlation values.

$$d_i=\sum_{k=1}^{14}w_k\eta_i(k) \tag{5}$$

Where d_i is the gray weighted correlation of the ith chemical component to the ideal object, w_k is the weight corresponding to the jth type of artifact, here we regard each chemical component as equal weight, so the weight w_k is 1/14.

The gray correlation degree values are ranked to analyze the variability of the correlations of chemical components among different categories of glass artifacts.

3.2. Model solving and result analysis

In this paper, SPSSPRO software was used to complete the chi-square test, in the chi-square test, the default H0 holds, that is, there is no difference between the observed and actual frequencies, that is, the two groups of variables do not affect each other, the two groups of variables are not correlated, if the test P-value is high, the hypothesis test passes; if the test P-value is low, the test fails, there is a difference between the observed and actual frequencies, and the two groups of variables are correlated [10].

Table 1. Chi-square test results

topic	name	Surface weathering		sum	X²	Calibration X²	P
		Y	F				
Ornamentation	A	11	11	22	4.957	4.957	0.084
	B	0	6	6			
	C	13	17	30			
type	Lead Barium	12	28	40	6.880	5.452	0.009
	High Potassium	12	6	18			
	nan	4	0	4			
color	light green	1	2	3	9.432	9.432	0.307
	Pale Blue	12	8	20			
	Dark green	4	3	7			
	Dark blue	0	2	2			
	purple	2	2	4			
	green	0	1	1			
	Blue-Green	9	6	15			
	black	2	0	2			

According to the analysis of the data in Table 1, it can be seen that for glass ornamentation, the significance P value is 0.084, indicating that the 10% significance level P value is large, so the level does not present significance, and the original hypothesis is accepted, so there is no significant difference for the surface weathering and ornamentation data of glass artifacts. For glass type, the significance P-value is 0.009, indicating that the 1% significance level P-value is very small, so the level presents significance and the original hypothesis is rejected, so there is a significant difference for the surface weathering and type data. For glass color, the significance P-value of 0.307 indicates that the 10% significance level P-value is large, so the level does not present significance and the original hypothesis is accepted, so there is no significant difference for the surface weathering and color data. It can be seen that ornamentation and color have less influence on whether the surface of the glass artifacts are weathered, and on the contrary the glass type has more influence.

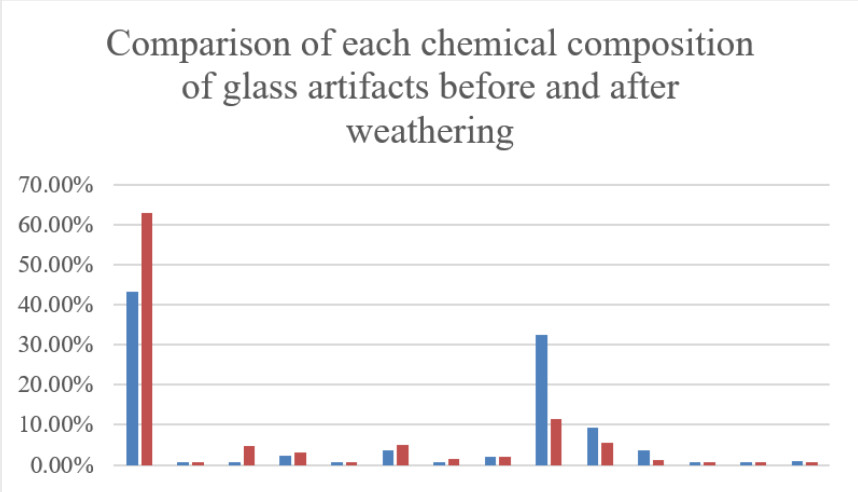


Figure 1. Comparison of each chemical composition of glass artifacts before and after weathering

Figure 1 Blue indicates differentiated data orange indicates unweathered data, from left to right, silica, nano oxide, potassium oxide, calcium oxide, magnesium oxide, aluminum oxide, iron oxide, copper oxide, lead oxide, barium oxide, phosphorus pentoxide, cesium oxide, tin oxide, and sulfurdioxide, respectively.

Through the comparison of the percentage of chemical composition of glass artifacts without weathering and weathering, it can be seen that the difference between the percentage of silica and lead oxide before and after weathering is the largest, indicating that these two chemical components may be more affected by the weathering of glass artifacts, while the difference between the percentage of barium oxide and potassium oxide before and after comparison is the second largest, and the other chemical components are less affected by the weathering of glass or the percentage of their own chemical components in the glass artifacts is The other chemical components are less affected by glass weathering or their own chemical composition in the glass artifacts is negligible. For the analysis of the chemical composition of glass artifacts of high potassium type before and after weathering, it can be seen that the chemical composition of silica increases about 25% before and after, potassium oxide loses about 10% before and after, and alumina loses about 8% before and after, and the chemical composition gaining and losing less than 5% before and after is regarded as not greatly affected by weathering.

For the comparative analysis of the chemical composition of glass relics of lead and barium types before and after weathering, it can be seen that the loss of silica is about 20% before and after, and the increase of lead oxide is about 15% before and after, and the loss of chemical composition before and after is less than 5%, which is considered to be not greatly affected by weathering.

In summary, for different types of glass artifacts before and after weathering the gain and loss of each chemical composition is different, among which silica is more affected by the weathering of glass artifacts, for high potassium glass weathering will lead to an increase in the content of silica, but for potassium oxide and barium oxide content will be reduced, while for lead-barium glass weathering will lead to a decrease in the content of silica, but will lead to an increase in the content of lead oxide.

The coefficients of the multiple linear regression models for predicting silica, potassium oxide, barium oxide, and lead oxide obtained by fitting the SPSSPRO software are shown in Tables 2, 3, 4, and 5 below.

Table 2. Coefficients of the multiple linear regression model for predicting silica

H_1	H_2	H_3	H_4	H_5	H_6	H_7	H_8	H_9	H_{10}	H_{11}	H_{12}	H_{13}	H_{14}	ξ
-0.319	0.89	5.948	0.555	-4.451	0.024	-3.461	-0.015	-0.378	-0.928	0.33	-2.143	4.155	-0.503	96.683

Table 3. Coefficients of the multiple linear regression model for predicting potassium oxide

H_1	H_2	H_3	H_4	H_5	H_6	H_7	H_8	H_9	H_{10}	H_{11}	H_{12}	H_{13}	H_{14}	ξ
-0.773	-1.127	3.977	-1.932	-2.202	-0.375	-2.197	0.556	-0.808	-1.185	-0.148	0.07	0.496	-0.236	81.903

Table 4. Coefficients of the multiple linear regression model for predicting barium oxide

H_1	H_2	H_3	H_4	H_5	H_6	H_7	H_8	H_9	H_{10}	H_{11}	H_{12}	H_{13}	H_{14}	ξ
0.513	0.455	-5.651	0.661	0.379	0.248	3.662	-0.051	0.545	0.952	0.023	1.148	-0.795	-0.068	-46.052

Table 5. Coefficients of the multiple linear regression model for predicting lead oxide

H_1	H_2	H_3	H_4	H_5	H_6	H_7	H_8	H_9	H_{10}	H_{11}	H_{12}	H_{13}	H_{14}	ξ
1.584	0.496	-6.267	4.019	7.715	0.99	1.935	-1.212	1.684	2.755	0.27	-1.2	-5.357	0.684	-150.191

The above multiple linear regression coefficients for the four chemical components are shown above, and the F-test for each of the above four chemical components can be obtained by, the P-value for silica significance is 0.046, which is significant at the level, and the original hypothesis that the regression coefficient is 0 is rejected, so the model basically meets the requirements. Potassium oxide significance P-value is 0.000, which is significant at the level of 0.000, rejecting the original hypothesis that the regression coefficient is 0, so the model basically meets the requirements. The P value of barium oxide is 0.016, which is significant at the level of 0.016, and the original hypothesis that the regression coefficient is 0 is rejected, so the model basically meets the requirements. The P value of lead oxide is 0.000, which is significant at the level of 0.000, and the original hypothesis that the regression coefficient is 0 is rejected, so the model basically satisfies the requirements. Since it is found that for different types of glass, the main chemical components affected by weathering are not the same, we only need to focus on the predicted results of silica, potassium oxide and barium oxide for the high potassium type of glass artifacts, and only silica and lead oxide for the lead-barium type of glass artifacts.

The gray correlation coefficients between the chemical compositions of the sample categories for the high potassium and lead-barium types of glass artifacts are shown in Table 6, which shows only the first five rows.

Table 6. Gray correlation coefficients for each chemical composition of high potassium type glass

SiO_2	SO_2	Al_2O_3	Fe_2O_3	SnO_2	MgO	SrO	CuO	P_2O_5	BaO	PbO	CaO	K_2O	Na_2O
0.99	0.89	0.98	0.89	0.89	0.89	0.89	0.93	0.96	0.89	0.99	0.95	0.98	0.89
0.98	0.89	0.99	0.94	0.89	0.95	0.77	0.86	0.96	0.58	0.67	0.94	0.90	0.89
0.99	0.66	0.97	0.94	0.90	0.89	0.90	1.00	0.98	0.90	0.90	0.91	0.94	0.90
0.98	0.59	0.94	0.90	0.90	0.87	0.88	0.94	0.99	0.90	0.90	0.90	0.92	0.90
0.99	0.89	0.88	0.92	0.89	0.85	0.74	0.98	0.74	0.78	0.97	0.89	0.98	0.89

Table 7. Gray correlation coefficients for each chemical composition of lead-barium type glass

SiO_2	Na_2O	CaO	Al_2O_3	Fe_2O_3	MgO	PbO	K_2O	BaO	CuO	SO_2	SnO_2	SrO	P_2O_5
0.99	0.91	0.99	0.95	0.86	0.93	0.96	0.68	0.91	0.92	0.91	0.91	0.96	0.99
0.98	0.91	0.96	1.00	0.91	0.99	0.98	0.91	0.96	0.93	0.91	0.91	0.96	0.87
1.00	0.92	0.92	0.95	0.89	0.92	0.94	0.77	0.89	0.87	0.92	0.92	0.92	0.93
0.97	0.58	0.94	0.95	0.92	0.99	0.96	0.92	0.99	0.95	0.92	0.92	1.00	0.92
0.94	0.91	0.97	0.98	0.97	0.95	0.96	0.96	0.94	0.93	0.91	0.79	0.94	0.94

Tables 6 and 7 show the evaluation of 14 chemical composition items for silicon dioxide (SiO2), sulfur dioxide (SO2), aluminum oxide (Al2O3), iron oxide (Fe2O3), tin oxide (SnO2), magnesium oxide (MgO), strontium oxide (SrO), copper oxide (CuO), phosphorus pentoxide (P2O5), barium oxide (BaO), lead oxide (PbO), calcium oxide (CaO), potassium oxide (K2O), sodium oxide (Na2O) and and 49 items of data respectively. The gray correlation analysis was performed on the data of calcium oxide (CaO), potassium oxide (K2O), sodium oxide (Na2O) and and 49 items, and the sum was used as the reference value sequence to study the 14 evaluation items (silicon dioxide (SiO2), sulfur dioxide (SO2), aluminum oxide (Al2O3), iron oxide (Fe2O3), tin oxide (SnO2), magnesium oxide (MgO), strontium oxide (SrO), and sodium oxide). Strontium oxide (SrO), copper oxide (CuO), phosphorus pentoxide (P2O5), barium oxide (BaO), lead oxide (PbO), calcium oxide (CaO), potassium oxide (K2O), sodium oxide (Na2O) and the total sum of correlation (correlation degree), and based on the correlation degree to provide analysis reference, using gray correlation analysis, the resolution coefficient is 0.5, combined with the correlation coefficient The correlation coefficient is 0.5, and the correlation coefficient is calculated by the formula, and then the correlation degree is calculated based on the correlation coefficient value for evaluation.

The gray correlation is shown in Table 8.

Table 8. Gray correlation between chemical compositions of high potassium type glasses

Evaluation items	Relevancy	Rank
SiO2	0.993	1
Al2O3	0.98	2
CuO	0.977	3
P2O5	0.971	4
K2O	0.971	5
MgO	0.97	6
CaO	0.969	7
Fe2O3	0.966	8
PbO	0.955	9
SrO	0.952	10
SnO2	0.95	11
BaO	0.947	12
Na2O	0.945	13
SO2	0.945	14

Table 9. Gray correlation between chemical composition of lead and barium type glass

Evaluation items	Relevancy	Rank
PbO	0.966	1
SiO2	0.964	2
Al2O3	0.954	3
BaO	0.954	4
SrO	0.951	5
CaO	0.942	6
MgO	0.932	7
CuO	0.925	8
P2O5	0.917	9
K2O	0.917	10
Fe2O3	0.913	11
Na2O	0.891	12
SO2	0.887	13
SnO2	0.883	14

As shown in Tables 8, 9, the interpreted correlation values ranged from 0 to 1. The larger the value represented the stronger correlation between it and the reference sequence. It also means that the higher its evaluation. The top five chemical composition correlations of high potassium glass are silicon dioxide (SiO2), aluminum oxide (Al2O3), copper oxide (CuO), phosphorus pentoxide (P2O5), and potassium oxide (K2O), indicating that the molding of high potassium type glass is mainly related to silicon dioxide, aluminum oxide, copper oxide, phosphorus pentoxide, and potassium oxide, while lead barium glass The top five chemical composition correlations are lead oxide (PbO), silicon dioxide (SiO2), aluminum oxide (Al2O3), barium oxide (BaO), and strontium oxide (SrO), indicating that lead-barium type glass molding is mainly related to lead oxide, silicon dioxide, aluminum oxide, barium oxide, and strontium oxide.

4. Conclusions

In this paper, we analyzed the relationship between glass type and weathering on the surface of glass artifacts by conducting frequency statistics of ornamentation, color, and type of glass artifacts in the form data and then using chi-square test according to whether the significance is less than 5%, and then analyzed the relationship between weathering and chemical composition on the surface of glass artifacts using frequency distribution histogram, and then judged the comparative relationship of each chemical composition of different glass types before and after weathering, and Finally, a multiple linear regression model was constructed to predict the chemical composition of the weathered glass and the F-test was conducted to prove the reasonableness of the prediction model for silica, potassium oxide, barium oxide, and lead oxide. The F-test of the model proved the reasonableness of the prediction model for silica, potassium oxide, barium oxide, and lead oxide. It is known that the glass type has influence on the chemical composition of glass, so the glass can be divided into two categories of glass: high potassium and lead-barium, and then the chemical composition of these two categories of glass were analyzed by gray correlation analysis, and the top five correlations of the chemical composition of each category of glass were obtained, and then the differences of the chemical composition of these two categories of glass were compared. The top five chemical composition correlations of high potassium glass are silicon dioxide (SiO2), aluminum oxide (Al2O3), copper oxide (CuO), phosphorus pentoxide (P2O5), and potassium oxide (K2O), indicating that the molding of high potassium glass is mainly related to silicon dioxide, aluminum oxide, copper oxide, phosphorus pentoxide, and potassium oxide, while the top five chemical composition correlations of lead barium glass are are lead oxide (PbO), silicon dioxide (SiO2), aluminum oxide (Al2O3), barium oxide (BaO), strontium oxide (SrO), indicating that lead-barium type glass molding is mainly related to lead oxide, silicon dioxide, aluminum oxide, barium oxide and strontium oxide.

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