

# Study on the ancient glass composition based on the multivariate statistical analysis method

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**Abstract.** The Silk Road was a channel of cultural exchange between China and the West, and glass was the precious material evidence of early trade. The study of the relationship between the weathering of ancient glass products and glass types and chemical composition contributes to the preservation of ancient cultural relics and promotes the further study of the history of the Silk Road. Based on this, this paper analyzed the surface weathering of cultural relics, and the type, ornamentation, color, and chemical composition of cultural relics, and it was finally found that there is a significant correlation between glass type and surface weathering. Then, discriminant analysis and systematic cluster analysis were applied to establish the classification function for the glass type variables and the chemical composition variables. Finally, gray correlation analysis was used to explore the correlation between each chemical component, and found that  $\text{SiO}_2\text{-Al}_2\text{O}_3$  was the most associated with each other in lead-barium glass, and  $\text{K}_2\text{O-CaO}$  was the highest in high potassium glass.

**Keywords:** correlation Analysis, discriminant analysis, systematic clustering, gray association analysis

## 1. Introduction

Being a route for cultural exchange between China and the West in its early years, the Silk Road exchanged a range of glass goods. Lead barium glass and high potassium glass were the two principal types of glass goods manufactured in ancient China. As ancient glass has been kept, it is susceptible to dryness and weathering brought on by environmental factors, as well as the extensive exchange of environmental components, which will change the color and decorative features of cultural artifacts. Therefore, it is an important topic to study the relationship between the weathering and chemical composition of ancient glass products and the relationship between glass types for the preservation of ancient glass relics and the study of the history of the Silk Road. In this paper, we study the significant relationship between the surface weathering of the glass relics types and chemical composition, and then the classification function is established by discriminant analysis and systematic clustering to divide the glass relics in detail. At the same time, the method of gray correlation analysis is used to study the correlation between the chemical components in cultural relics, so as to compare the differences between the chemical components of different categories of ancient glass products.

## 2. Literature review

Among the cultural relics excavated around the world, there are a large number of ancient glass products, which provide valuable material data for the study of the origin and development of ancient glass technology. It has become an important topic to study the chemical composition of ancient glass products and identify their types. In previous studies, proton excitation X-ray fluorescence analysis and X-ray diffraction and laser Raman spectroscopy techniques were usually used to obtain the chemical composition data of each glass, and then the glass was classified according to the content of the main chemical components, and the production technology and characteristics of various places were inferred according to the unearthed sites of various cultural relics [1, 2]. However, in the previous research directions, only the content of the main chemical components of cultural relics is considered, and the influence of other trace components on glass types is not considered, so the glass types cannot be judged comprehensively. Meanwhile, the influence of glass weathering on the chemical composition content of glass was not considered in previous studies. In the subsequent studies, all the chemical components of glass relics can be investigated, and the correlation between weathering and each chemical component can be studied.

## 3. Data source and pre-processing

In this paper, we used the data from the 2022 China Undergraduate Mathematical Contest in Modelling [3]. We deleted literature 19,40,48 and 58 in the data, deleted the component data of cultural relic sampling point 15 and cultural relic sampling point 17 in the data, and defined all the missing chemical composition content as 0.

## 4. Correlation analysis

### 4.1. Correlation between surface weathering and glass type, ornamentation, and color

When discussing whether there is a relationship between the data and the degree of correlation, it is usually analyzed for correlation. The main functions of correlation analysis are: (1) to determine the statistical association between two or more variables; (2) If there is an association, the strength and direction of the association are further analyzed. Based on this, this paper uses correlation analysis to discuss the relationship between surface weathering and glass type, ornamentation, and color.

First, determine the data type of four sets of data: weathering, glass type, ornamentation, and color. Because the weathered data is unweathered/weathered, it is a dichotomous variable, the glass type data is high potassium glass/lead barium glass, so it is a dichotomous variable, and the ornamentation and color data have three or more categories of classification variables, so the ornamentation and category are disordered categorical variables.

Second, select the appropriate test method according to the types of the two variables. Since the correlation analysis of dichotomous variables (surface weathering)-dichotomous variables (glass type) and the correlation analysis of dichotomous variables (surface weathering)-disordered categorical variables (ornamentation, color) are entitled in this title, the test methods are chi-square tests.

Finally, using SPSS, select Analysis-Crosstab: Statistics for chi-square testing. When the significance of Pearson's chi-square is less than 0.05, there is a significant correlation, indicating that the surface weathering of glass cultural relics is obviously related to it, and vice versa, there is no significant correlation, indicating that the surface weathering of glass cultural relics has no obvious correlation.

The correlation analysis by SPSS leads to the following conclusions in Table 1.

**Table 1.** Pearson's chi-square significance values

Data	Pearson's chi-square asymptotic significance (bilateral)
Surface Weathering - Glass Type	0.009
Surface Weathering - Ornamentation	0.084
Surface Weathering - Color	0.307

It can be seen that glass type and surface weathering are significantly correlated, while ornamentation and color are not significantly correlated with surface weathering [4].

#### 4.2. Chemical composition correlation analysis

The correlation analysis of 14 chemical components and surface weathering of the artifact samples was carried out separately for the two sample sets of high potassium glass and lead-barium glass. The correlation between 14 chemical components and surface weathering under various glass types was obtained by ETA detection. If the value of the symmetric measurement was greater than 0.5, it was highly correlated, and the larger the value, the greater the correlation. If the value was less than 0.5, there was no significant correlation.

The correlation degree of each component with surface weathering in the high potassium glass dataset and the lead-barium glass dataset were obtained by chemical composition correlation analysis, respectively, as shown in Table 2 below.

**Table 2.** Correlation between chemical composition and surface weathering under different glass types

Chemical composition	Progressive significance (high potassium)	Progressive Significance (Lead Barium)
SiO <sub>2</sub>	1.000	1.000
Na <sub>2</sub> O	0.357	0.575
K <sub>2</sub> O	0.919	0.687
CaO	1.000	0.947
MgO	0.897	0.734
Al <sub>2</sub> O <sub>3</sub>	1.000	1.000
Fe <sub>2</sub> O <sub>3</sub>	0.940	0.716
CuO	1.000	0.929
PbO	0.602	1.000
BaO	0.426	1.000
P <sub>2</sub> O <sub>5</sub>	0.940	0.914
SrO	0.564	0.892
SnO <sub>2</sub>	0.193	0.326
SO <sub>2</sub>	0.357	0.314

From Table 2, it can be obtained that: in the data set of high potassium glass, eight chemical components of silica, potassium oxide, calcium oxide, magnesium oxide, aluminum oxide, iron oxide, copper oxide, and phosphorus pentoxide are significantly correlated with surface weathering; in the data set of lead barium, eight chemical components of silica, calcium oxide, aluminum oxide, copper oxide, lead oxide, barium oxide, phosphorus pentoxide, and strontium oxide are significantly correlated with surface weathering.

#### 5. Cluster division

Clustering is a technique for finding the intrinsic structure between the data. Clusters organize the entire set of data instances into similar groups that are called clusters. In this paper, the chemical composition content of each artifact sampling site is used as a variable and each cluster as an instance. In clustering, the intermediate distance method and Euclidean distance square are used to operate on the data of the sampling sites in the two types and divide the categories [5, 6].

The clustering results of lead barium glass are shown in Table 3.

**Table 3.** Lead barium glass clustering results

Heritage Sampling Sites	Category	Heritage Sampling Sites	Category	Heritage Sampling Sites	Category
02	L1	33	L0	48	L0
08	L2	34	L1	49	L1
08 Severe weathering point	L2	35	L0	49 Unweathered spots	L0
11	L2	36	L1	50	L1
19	L2	37	L0	50 Unweathered spots	L0
20	L2	38	L1	51 Part 1	L1
23 Unweathered spots	L0	39	L1	51 Part 2	L1
24	L2	40	L1	52	L1
25 Unweathered spots	L0	41	L1	53 Unweathered spots	L0
26	L2	42 Unweathered point 1	L0	54	L1
26 Severe weathering points	L2	42 Unweathered spot 2	L0	54 severely weathered spots	L1
28 Unweathered spots	L0	43 parts1	L1	55	L0
29 Unweathered spots	L0	43 parts2	L1	56	L1
30 parts1	L1	44 Unweathered spots	L0	57	L1
30 parts2	L1	45	L0	58	L1
31	L0	46	L0		
32	L0	47	L0		

By analyzing the clustering results and combining them with the references, it can be found that these three subclasses satisfy.

L0.PbO – BaO – SiO<sub>2</sub>;

L1.CaO – PbO(~45%) – BaO – SiO<sub>2</sub>;

L2.PbO(~25%) – BaO – SiO<sub>2</sub>.

The clustering results of high potassium glass are shown in Table 4.

**Table 4.** High potassium glass clustering results

Heritage Sampling Sites	Category	Heritage Sampling Sites	Category	Heritage Sampling Sites	Category
01	L3	07	L4	18	L4
03 Part 1	L4	09	L4	21	L4
03 Part 2	L3	10	L4	22	L4
04	L3	12	L4	27	L4
05	L3	13	L3		
06 Part 1	L3	14	L3		
06 Part 2	L3	16	L3		

By analyzing the clustering results, combined with the references, it can be found that these two subclasses satisfy.

L3.K<sub>2</sub>O – CaO(~6%) – SiO<sub>2</sub> ;  
L4.K<sub>2</sub>O – SiO<sub>2</sub> .  
Five subtypes were finally identified [2, 5, 7].  
L0.PbO – BaO – SiO<sub>2</sub> ;  
L1.CaO – PbO(~45%) – BaO – SiO<sub>2</sub> ;  
L2.PbO(~25%) – BaO – SiO<sub>2</sub> ;  
L3.K<sub>2</sub>O – CaO(~6%) – SiO<sub>2</sub> ;  
L4.K<sub>2</sub>O – SiO<sub>2</sub>

## 6. Rationality analysis

Based on the subclasses of chemical composition content, it can be inferred from the clustering results that high potassium glass may be divided into two clusters, therefore, a rational analysis of high potassium glass with two clusters must be carried out. A and B are the clusters. Let A contain m samples of glass artifacts, and B contain n samples of glass artifacts. Set each chemical composition content index as  $g_i$  ( $i \in [1, 14]$ ), i.e.,  $g_1, g_2, \dots, g_{14}$ , so that the content of 14 chemical components of each artifact sample is the score value of the component index of that sample.

Let the m samples be in category  $Ag_i$ . The mean and standard deviation of the scores are  $\mu_A(g_i)$  and  $\sigma_A(g_i)$ , and the mean and variance of the scores of the n samples in category B are  $\mu_B(g_i)$  and  $\sigma_B(g_i)$ . Define  $g_i$ . The correlation coefficient with the categorical variable c is

$$P(g_i, c) = \frac{\mu_A(g_i) - \mu_B(g_i)}{\sigma_A(g_i) + \sigma_B(g_i)} \quad i = 1, \dots, m \quad (1)$$

When  $|P(g_i, c)|$  is larger, it means that the chemical composition content  $g_i$  level taken in class A is more different from that taken in class B.

Let the  $i$ th chemical composition content in the  $k$ th artifact sample score value is  $e_{ki}$ , for the  $k$ th sample, calculate the following.

$$V_i = P(g_i, c)(e_{ki} - \frac{\mu_A(g_i) + \mu_B(g_i)}{2}), i=1, 2, \dots, m \quad (2)$$

$$V^+ = \sum_{V_i > 0} V_i, V^- = -\sum_{V_i < 0} V_i \quad (3)$$

$V^+$  and  $V^-$  denote the proximity of the  $k$ th artifact sample to the two classes A, B, respectively, when  $V^+ > V^-$ , the  $k$ th sample is closer to class A, and vice versa to class B.

$$PS = \frac{V_{win} - V_{lose}}{V_{win} + V_{lose}} \quad (4)$$

If  $V^+ > V^-$ , then  $V^+ = V_{win}$ ; then  $V^- = V_{lose}$ .

If  $V^+ < V^-$ , then  $V^+ = V_{win}$ ; then  $V^- = V_{lose}$ .

When the PS value of an artifact sample is large, it means that the artifact sample has similar chemical composition content to the sample in the cluster it is classified.

The PS value is calculated for each artifact sample in the high-potassium glass set in turn, and a total of  $n+m$  PS values can be obtained  $M_0$ .

The following is based on the above calculated  $M_0$ , the clustering rationality is analyzed.

Step 1: The  $n+m$  artifact samples are randomly divided into two classes under the condition that the sample points are not 1 in satisfying the clustering, for a total of  $Q$  times, and the two classes in the  $q$ th time are noted as  $CL_{q1}, CL_{q2}$  ( $q=1, 2, \dots, Q$ ).

Step 2: For all the heritage samples in both categories under each classification, the PS values of each sample were calculated according to Equations (4)~(7), and the median PS values of all samples were calculated as follows in Table 5, and the median under the  $q$ th classification scheme was noted as  $M_q$  ( $q=1, 2, \dots, Q$ ).

Table 5 contains the randomly classified scheme as well as the median per scheme.

**Table 5.** Random classification schemes and their medians

Classification scheme	First category	Second category	Median
1	CL <sub>11</sub>	CL <sub>12</sub>	M <sub>1</sub>
2	CL <sub>21</sub>	CL <sub>22</sub>	M <sub>2</sub>
...	...	...	...
Q	CL <sub>Q1</sub>	CL <sub>Q2</sub>	M <sub>Q</sub>

Step 3: 95% unilateral reference range (upper limit) for M<sub>1</sub>, M<sub>2</sub>,..., M<sub>Q</sub>, noted in L:

$$L = M' + \frac{S}{\sqrt{Q}} t_{0.05} (Q-1) \quad (5)$$

of which  $M'$  is the average of  $M_1, M_2 \dots M_Q$ , and  $S$  is the standard deviation of  $M_1, M_2 \dots M_Q$ .

When  $M_0 > L$ , it means that the classification of  $n+m$  artifact samples into A, B categories is not only the result of statistical classification, but also has practical significance, so the classification is reasonable [8].

From the results of the clustering, it is obtained that the lead-barium glass can be divided into three clusters according to the chemical composition content subclasses, so it is necessary to analyze the reasonableness of the lead-barium glass with the number of clusters as 3. The clusters are C, D, and E, respectively.

Applying the above reasonableness test for the subclassification of high potassium glass, a two-by-two test was performed for the three clusters C, D, and E. Comparing in turn the C-D clusters, D-E clusters, and D-E clusters in  $M_0$  and  $L$ . If the relationship between the sizes of the three types of  $M_0$  are greater than  $L$ , the results of subclassification of lead-barium glass can be judged to be reasonable.

From the method and steps described in the model building, we can sequentially find the four tests of  $M_0$  and  $L$ -values, as shown in Table 6 below.

**Table 6.**  $M_0$  and the values obtained for  $L$

	$M_0$	$L$
High Potassium Glass	0.881701184639396	0.7118940656627504
Lead barium glass-CD clustering	0.9341733832866326	0.6142303796481939
Lead barium glass-DE clustering	0.9467898802750668	0.6071729036469156
Lead barium glass-CE clustering	0.9955830149356246	0.6834500496770781

From Table 6, it can be obtained that: under the four tests  $M_0$  are greater than  $L$ . Therefore, the clustering result is not only the result of statistical classification, and has practical significance, and the subclass division result is reasonable.

## 7. Discriminant analysis

A multivariate statistical analysis technique called discriminant analysis determines a study object's classification based on its various eigenvalues. Establishing one or more discriminant functions in accordance with predetermined discrimination criteria, calculating the discriminant index, which can be used to identify a specific category, and determining the pending coefficient in the discriminant function with a significant amount of research object data constitute the basic principles. We used SPSS software for discriminant analysis of ancient glass product data, including 14 chemical components as explanatory variables.

After obtaining the clustering results, the subclass classification function coefficients of lead barium glass were obtained by discriminant analysis as follows (Table 7):

**Table 7.** Subclass classification function coefficients of lead barium glass

	Classification function coefficients		
	Category		
	L0	L1	L2
Silicon dioxide (SiO <sub>2</sub> )	28.290	27.950	27.636
Sodium oxide (Na <sub>2</sub> O)	35.379	34.741	32.691
Potassium oxide (K <sub>2</sub> O)	-.328	5.346	12.084
Calcium oxide (CaO)	35.787	36.269	34.992
Magnesium oxide (MgO)	34.809	33.829	30.222
Aluminum oxide (Al <sub>2</sub> O <sub>3</sub> )	27.791	27.794	27.478
Iron oxide (Fe <sub>2</sub> O <sub>3</sub> )	27.715	28.677	27.566
Copper oxide (CuO)	5.873	4.442	8.131
Lead oxide (PbO)	29.186	29.517	28.566
Barium oxide (BaO)	37.327	37.879	38.200
Phosphorus pentoxide (P <sub>2</sub> O <sub>5</sub> )	36.710	36.869	37.308
Strontium oxide (SrO)	-66.346	-67.374	-59.404
Tin oxide (SnO <sub>2</sub> )	-25.423	-27.253	-28.524
Sulfur dioxide (SO <sub>2</sub> )	9.768	8.299	9.376
(Constant)	-1420.657	-1421.677	-1402.403

The resulting classification function can be obtained as:

$$YP_{L0} = 28.290SiO_2 + 35.379Na_2O - 0.328K_2O + 35.787CaO + 34.809MgO + 27.791Al_2O_3 + 27.715Fe_2O_3 + 5.873CuO + 29.186PbO + 37.327BaO + 36.710P_2O_5 - 66.346SrO - 25.423SnO_2 + 9.768SO_2 - 1420.657 \quad (6)$$

$$YP_{L1} = 27.950SiO_2 + 34.741Na_2O + 5.346K_2O + 36.269CaO + 33.829MgO + 27.794Al_2O_3 + 28.677Fe_2O_3 + 4.442CuO + 29.517PbO + 37.879BaO + 36.869P_2O_5 - 67.374SrO - 27.253SnO_2 + 8.299SO_2 - 1421.677 \quad (7)$$

$$YP_{L2} = 27.636SiO_2 + 32.691Na_2O + 12.084K_2O + 34.992CaO + 30.222MgO + 27.478Al_2O_3 + 27.566Fe_2O_3 + 8.131CuO + 28.566PbO + 38.200BaO + 37.308P_2O_5 - 59.404SrO - 28.524SnO_2 + 9.376SO_2 - 1402.403 \quad (8)$$

For any set of data, we can bring the components into the function,  $YP_{L0}$ ,  $YP_{L1}$  and  $YP_{L2}$ , which value is larger, then belongs to which category [9].

## 8. Grey association analysis

The fundamental goal of gray correlation analysis is to decide whether or not there is a connection based on the similarity of the geometry of the sequence curves in a gray system, which is a system where some information is known and some is unknown. The degree of correlation between the respective sequences increases with the proximity of the curves; conversely, the smaller the degree of correlation. In general, gray correlation analysis uses the steps listed below.

Step1: Eliminate the dimension;

Step2: Calculate the correlation coefficient

Step3: Calculate the correlation degree.

The formula for the correlation coefficient is:

$$\xi_i(k) = \frac{\min_s \min_t |x_0(t) - x_s(t)| + \rho \max_s \max_t |x_0(t) - x_s(t)|}{|x_0(k) - x_i(k)| + \rho \max_s \max_t |x_0(t) - x_s(t)|} \quad (9)$$

Define the correlation of the sequence  $x_i$  to the reference sequence  $x_0$  as

$$r_i = \frac{1}{n} \sum_{k=1}^n \xi_i(k) \quad (10)$$

The gray correlation matrix was derived in high potassium glass and lead-barium glass, respectively, and the correlation of any chemical component with other chemical components was obtained, and the three with the highest correlation were selected to obtain the following two tables [10, 11].

The top three chemical composition results for the chemical composition correlation of lead barium glass are shown in Table 8.

**Table 8.** Chemical composition of lead barium glass correlation of the top three

Chemical composition	Relevance1	Relevance 2	Relevance3
SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	BaO	PbO
Na <sub>2</sub> O	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	SO <sub>2</sub>
K <sub>2</sub> O	Al <sub>2</sub> O <sub>3</sub>	MgO	SiO <sub>2</sub>
CaO	P <sub>2</sub> O <sub>5</sub>	PbO	SrO
MgO	Al <sub>2</sub> O <sub>3</sub>	CaO	SiO <sub>2</sub>
Al <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	MgO	PbO
Fe <sub>2</sub> O <sub>3</sub>	CaO	Al <sub>2</sub> O <sub>3</sub>	MgO
CuO	BaO	SrO	PbO
PbO	SrO	CaO	BaO
BaO	SrO	PbO	CuO
P <sub>2</sub> O <sub>5</sub>	CaO	PbO	SrO
SrO	PbO	BaO	CaO
SnO <sub>2</sub>	SO <sub>2</sub>	Fe <sub>2</sub> O <sub>3</sub>	K <sub>2</sub> O
SO <sub>2</sub>	SnO <sub>2</sub>	CuO	BaO

Some of the chemical elemental correlations in lead barium glass have the following distinguishing features.

(1) SiO<sub>2</sub>-Al<sub>2</sub>O<sub>3</sub> have the highest correlation with each other, indicating that the content of alumina has a strong influence on the content of silicon oxide.

(2) CaO-P<sub>2</sub>O<sub>5</sub> has the highest correlation with each other, indicating that the production process of the two produces a strong correlation.

The top three chemical composition results for the chemical composition correlation of high potassium glass are shown in Table 9.

**Table 9.** Top three correlations of chemical composition of high potassium glass

Chemical composition	Relevance1	Relevance 2	Relevance3
SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	CuO	P <sub>2</sub> O <sub>5</sub>
Na <sub>2</sub> O	PbO	SnO <sub>2</sub>	CaO
K <sub>2</sub> O	CaO	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>
CaO	K <sub>2</sub> O	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>
MgO	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	CaO
Al <sub>2</sub> O <sub>3</sub>	CaO	MgO	K <sub>2</sub> O
Fe <sub>2</sub> O <sub>3</sub>	CuO	P <sub>2</sub> O <sub>5</sub>	CaO
CuO	Fe <sub>2</sub> O <sub>3</sub>	CaO	Al <sub>2</sub> O <sub>3</sub>
PbO	BaO	Na <sub>2</sub> O	Al <sub>2</sub> O <sub>3</sub>
BaO	PbO	SrO	P <sub>2</sub> O <sub>5</sub>
P <sub>2</sub> O <sub>5</sub>	Fe <sub>2</sub> O <sub>3</sub>	Al <sub>2</sub> O <sub>3</sub>	SrO
SrO	P <sub>2</sub> O <sub>5</sub>	MgO	BaO
SnO <sub>2</sub>	Na <sub>2</sub> O	SO <sub>2</sub>	SrO
SO <sub>2</sub>	SnO <sub>2</sub>	MgO	CaO



Some of the chemical elemental correlations in high potassium glasses have the following distinguishing features.

(1)  $K_2O$ - $CaO$  correlates most strongly with each other, and potassium oxide is used as the main co-solvent in the production of high potassium glasses, which suggests a strong relationship with the stabilizer limestone fired to calcium oxide.

(2)  $PbO$ - $BaO$  have the highest correlation with each other, indicating that in high potassium glasses, these two components vary in pairs.

## 9. Conclusion

Through the correlation analysis of artificial type, ornamentation, and color, we concluded that only glass type was significantly associated with artificial surface weathering. Next, we conducted a correlation analysis on the weathering and chemical composition of different types of glass relics, and found that eight chemical components were significantly associated with weathering. Subsequently, we used the systematic clustering method to cluster the two types of glass products, and obtained 3 subclasses of lead barium glass and 2 subclasses of high potassium glass, and obtained 5 Fisher linear discrimination functions through discriminant analysis. By substituting the chemical composition content of each glass product into the discriminant function, we can know its subclass. Finally, we used gray correlation analysis to calculate the correlation between each chemical composition.

Due to the limitation of the size of the data set, the sensitivity analysis of the model was not conducted in this paper, so the influence of uncertainty factors on the discriminant results cannot be measured. In the process of model improvement, BP neural network can be introduced to predict the chemical composition content of ancient glass products, so as to more systematically subclassify the types of glass products.

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