Characterising Chinese Ru ware in the Sir Percival David collection at the British Museum using handheld XRF analysis

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ABSTRACT

Ru ware is a very rare and highly prized stoneware from the end of the Northern Song period of China (960–1127 CE). Stylistic and art historical work by Regina Krahl (2021) suggests that a brush washer in the Sir Percival David Collection, housed in the British Museum, might be Ru rather than Korean Goryeo ware as previously thought. This paper reports the analysis of the glaze of this piece by handheld XRF in comparison with 10 pieces of Ru and 10 pieces of Goryeo ware. Despite the compositional similarity of the glazes, the analysis was able to show conclusively that the piece is Ru ware. The work has implications for the analysis of Chinese stoneware and beyond, showing that it may be possible (under the right conditions) to distinguish different productions relatively quickly and easily.

Introduction

Ru (Ruyao) ware is the most prized and rarest of all Chinese ceramics (Geng, 2015; Kerr, 2004; Kobayashi, 2009; Krahl and Harrison-Hall 2009:11; Li 2007:134–36; Sun and Guo, 2016). It was produced for only 20 years at the end of the Northern Song period, during the reign of Emperor Huizong (1101–1125 CE). The end of the Song Dynasty, in CE 1127, saw the court flee south and abandon the production sites and facilities that made the ware extremely rare and very desirable even then (Krahl 2021). Contemporary potters produced a very similar ware in Goryeo, Korea, but (while it is obviously related through the colour of the glaze and forms) it is unclear whether Goryeo wares were modelled on Ru wares (Krahl 2021). Much later in Chinese history, the Yongzheng emperor (1723–1735 CE) arranged for Ru copies to be made in the imperial kilns at Jingdezhen, using original pieces as models. The rarity of Ru ware meant that very few people ever saw or handled a piece, which lent it an "almost mythical aura" (Krahl 2021). Until the twentieth century, near-mythical Ru ware was unknown in the West. In China, knowledge of it was retained through the imperial collection. However, which pieces were Ru and which were other wares was and still is debated. To date, only 88 pieces of Ru ware have been published, the last being a brush washer in the Staatliche Kunstsammlungen in Dresden, identified by Regina Krahl (Krahl 2021).

The Sir Percival David Foundation (PDF) collection, housed in the British Museum, includes a world-class collection of Northern Song Ru wares. With 13 pieces, this is the largest collection outside China. The British Museum has another 4 Northern Song Ru wares. In her work on the Dresden brush washer, Krahl studied the three Ru brush washers in the PDF collection as *comparanda*. Her attention was drawn to another brush washer (PDF.59) that had been classified as a Korean piece since the 1970s (Fig. 1).

PDF.59 originally entered the PDF collection as a Korean piece, but Sir Percival David himself was convinced it was Ru and catalogued it as such. After that it fell from favour, and for the last 50 years it was catalogued as Korean again. Krahl proposed that PDF.59 was indeed Ru and mentioned this in a brief note in her paper on the Dresden piece. This led to a query from the trustees of the PDF asking if there was an analytical approach to verify or otherwise determine whether PDF.59 could be the eighty-ninth Ru piece or the much more common Goryeo celadon ware. Stylistically, it could be narrowed down to one of the two, but the glazes are close in composition (Li et al., 2018; Wood 1999:127; Wood and Li 2014). The object was complete, so it had to be approached with a non-destructive test focussing on the glaze. As such vessels are almost totally glazed, there is very little available body clay for analysis beyond the spur marks. During the last two decades, research on Ru wares has typically been focused on the colouring mechanisms of the opaque sky blue or sky green glaze. These investigations, usually based on results from vibrational spectroscopy, X-ray fluorescence spectrometry (XRF), and scanning electron microscopy (SEM), uncovered the prominent roles of intricate chemical and physical mechanisms in the colouration and vi-

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Fig. 1. Pictures of the celadon PDF.59. © The Trustees of the British Museum.

Table 1
Samples analysed in this study.

Museum Number	Ware	Type	Museum Number	Ware	Type
1911,0607.4	Goryeo ware	*kundika*	PDF.1	Ru ware	brush washer
1936,1012.201	Goryeo ware	bowl	PDF.3	Ru ware	bowl
1973,0726.392	Goryeo ware	cup holder	PDF.60	Ru ware	dish
1973,0726.390	Goryeo ware	bottle	PDF.61	Ru ware	bottle
1983,1007.2 (cup)	Goryeo ware	cup	PDF.76	Ru ware	brush washer
1983,1007.2 (stand)	Goryeo ware	cup stand	PDF.81	Ru ware	stand for bowl
1992,0615.34	Goryeo ware	dish	PDF,A.26	Ru ware	brush washer
1992,0615.32	Goryeo ware	ewer	PDF,A.44	Ru ware	incense burner
1997,0609.1	Goryeo ware	cup stand	1936,1019.1	Ru ware	dish
2001,0808.1	Goryeo ware	vase	1971,0921.1	Ru ware	stand for bowl
			PDF.59	unknown	brush washer

Note: Full details on the pieces can be found on the British Museum collections website: https://www.britishmuseum.org/collection.

sual aspects of these celadons (Bin et al., 2004; Li et al., 2005; Shi et al., 2020; Wu et al., 2021; Yang et al., 2005). Few studies have highlighted the possibility of using energy dispersive XRF (ED-XRF) to discriminate Ru wares produced in Qingliangsi, Henan Province, from visually similar celadons produced elsewhere in China (Li et al., 2018; Wood and Li 2014).

The aim of the present study is to determine the origin of the celadon PDF.59 from the PDF collection at the British Museum using handheld XRF analysis. While the accuracy of XRF analyses on glass and glazes has been questioned, the precision and repeatability of the analysis is often excellent. The underlying assumption was that this instrument would be able to discriminate the glaze composition of Ru wares from Goryeo wares qualitatively by comparing spectra, without the need for quantitative datasets.

Materials and methods

To assess the origin of the celadon PDF.59, 10 Ru wares from the Northern Song Dynasty (1086–1125) and 10 Goryeo wares (twelfth century) were used as reference materials to identify their chemical fingerprints (Table 1).

A Bruker TRACER 5i handheld XRF spectrometer equipped with a rhodium target, a 3 mm collimator, and a silicon drift detector was used for the measurements. The experimental parameters were a 40 kV tube acceleration voltage, 30 μA current, and 15-second measurement time without filters. A short measurement time was used because of the difficulties of holding the instrument gently in contact with the objects. All the data were taken in helium atmosphere with a flow between 0.5 and 1 L/min, and each object was measured five times on different areas. Since the analyses were performed in a non-destructive way on complete vessels, the measurements were taken only on the glazed surfaces of the artefacts. It should be noted that the interpretation of surface analysis can be influenced by the penetration depth of the X-ray beam, which is a function of the density and grain size of the matrix. Depending on the composition and thickness of the glaze layer, the data recorded by the

spectrometer might be influenced by the composition of the body of the ceramic.

Bruker spectra software was used to process the raw data, and the spectra were averaged using Spectragryph software (Menges 2020). To reduce the signal variation caused by minor changes in analytical conditions, three of the five spectra with the closest count rates were selected. The values, given in peak area integrals for each element, were normalised on total counts per second (kcps) to overcome various matrix effects and differences in measurement conditions (Bouchard et al., 2011; Martin et al., 2014). The data were exported to Excel to calculate averages, standard deviations, and relative standard deviations. For this dataset, classic analysis of variance (ANOVA) and the Mann-Whitney U test are not best suited to evaluate the discriminating power of each element because the Bartlett's test of homogeneity of variance indicated that several distributions would violate the homoscedasticity assumption required to apply these methods. Thus the Welch's ANOVA was used to assess the compositional differences between the Ru wares and the Goryeo wares analysed. This method tests the null hypothesis, which is that the mean of the two groups are equal; a low Welch *p*-value means that the null hypothesis should be rejected. A Varimax-rotated principal component analysis (PCA) was then computed to achieve a better understanding of the distinction based on the multivariate data matrix. The Welch's ANOVA and the rotated PCA were computed using Xlstat (version 4.1).

Results

The averaged spectra of the 21 ceramic samples show a similar elemental composition with the detection of Na, Mg, Al, Si, P, K, Ca, Ti, V, Mn, Fe, Ni, Cu, Zn, Rb, Sr, and Ba. Some compositional differences between the Goryeo wares and the Chinese Ru wares can be seen simply by comparing the raw spectra normalised to the Rh Compton peak (Fig. 2). The counts on the Sr Kα lines (14.16 keV) and the Mn Kα lines (5.90 keV) are higher for all the Goryeo wares, with a clear gap between the two groups of celadon. The counts for the Al Kα lines (1.48 keV) and
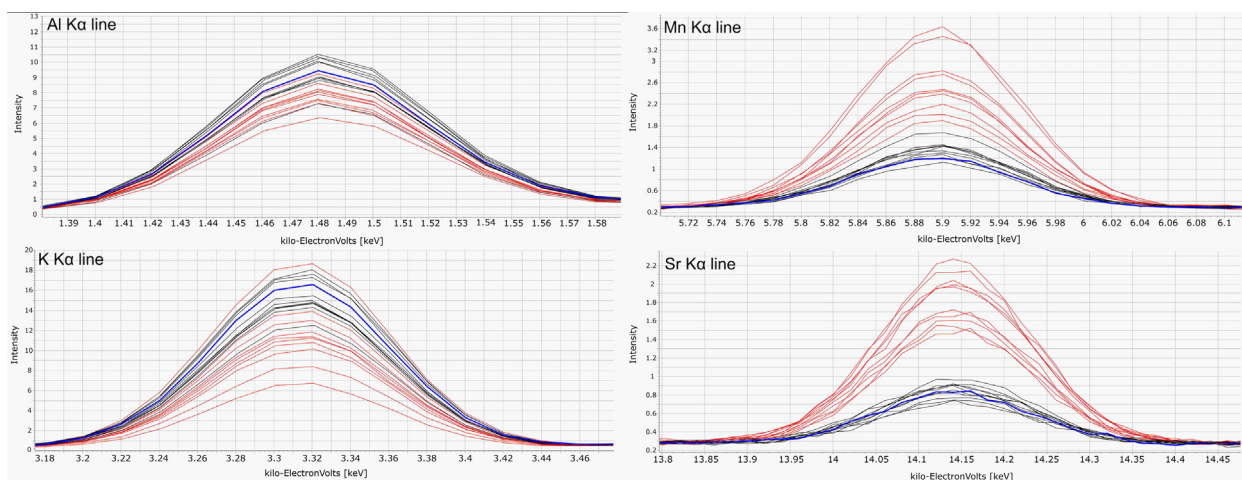
Fig. 2. Spectra of the 21 samples focused on spectral regions corresponding to the Al Kα lines, K Kα lines, Mn Kα lines, and Sr Kα lines. The red lines are the Goryeo wares spectra, the black lines are Ru wares spectra, and the blue line is the sample PDF59 spectrum. All the spectra are normalised on the Rh Compton peak.

Table 2

Means, relative standard deviations (%), Results of the Shapiro-Wilk test, and results of the Welch's ANOVA on the Ru wares and goryeo wares.

	Group	Mean	%RSD	S-W *p*-value	Welch *p*-value
Al	Goryeo wares (*n* = 10)	322.9	8.5	0.349	**0.000**
	Ru wares (*n* = 10)	377.2	5.6	0.905	
Ba	Goryeo wares (*n* = 10)	23.8	11.4	0.143	0.289
	Ru wares (*n* = 10)	22.7	9.0	0.529	
Ca	Goryeo wares (*n* = 10)	2340.2	9.9	0.160	0.058
	Ru wares (*n* = 10)	2138.7	9.9	0.119	
Cu	Goryeo wares (*n* = 10)	11.1	15.6	0.339	**0.001**
	Ru wares (*n* = 10)	14.8	14.5	0.053	
Fe	Goryeo wares (*n* = 10)	957.3	19.6	0.862	**0.002**
	Ru wares (*n* = 10)	1216.6	9.7	0.721	
K	Goryeo wares (*n* = 10)	586.0	26.1	0.697	**0.005**
	Ru wares (*n* = 10)	771.0	11.4	0.810	
Mg	Goryeo wares (*n* = 10)	16.7	30.0	0.066	0.603
	Ru wares (*n* = 10)	15.8	14.7	0.468	
Mn	Goryeo wares (*n* = 10)	148.3	26.2	0.827	**< 0.0001**
	Ru wares (*n* = 10)	66.1	11.3	0.608	
Ni	Goryeo wares (*n* = 10)	63.4	5.3	0.656	NA
	Ru wares (*n* = 10)	62.7	7.1	**0.009**	
P	Goryeo wares (*n* = 10)	21.0	36.9	0.060	0.632
	Ru wares (*n* = 10)	19.6	24.8	0.464	
Rb	Goryeo wares (*n* = 10)	35.4	10.2	**0.017**	NA
	Ru wares (*n* = 10)	30.7	11.1	0.123	
Si	Goryeo wares (*n* = 10)	2758.2	3.7	0.492	**0.009**
	Ru wares (*n* = 10)	2610.3	4.7	0.877	
Sr	Goryeo wares (*n* = 10)	150.8	14.1	0.724	**< 0.0001**
	Ru wares (*n* = 10)	54.3	15.0	0.909	
Ti	Goryeo wares (*n* = 10)	21.5	10.2	0.664	**0.001**
	Ru wares (*n* = 10)	29.1	17.9	0.413	
V	Goryeo wares (*n* = 10)	7.7	13.9	0.945	NA
	Ru wares (*n* = 10)	8.7	9.5	**0.005**	
Zn	Goryeo wares (*n* = 10)	6.5	58.0	0.085	0.311
	Ru wares (*n* = 10)	7.9	18.8	0.162	

Note: The *p*-values in bold indicate the rejection of the null hypothesis.

the K Kα lines (3.31 keV) are generally higher for the Chinese Ru wares. However, some samples from the two groups are clearly overlapping, and no clear limit can be drawn between the Ru and the Goryeo wares since the distribution of the values is more continuous. On all four plots, the spectrum of PDF.59 fits in the range of the Ru wares spectra.

The mean, relative standard deviation results of the Shapiro-Wilk normality test and the results of the Welch's ANOVA performed on the kcps-normalised data of the 20 reference samples are summed up in Table 2. The sodium values were discarded because of low counts and high variability. The Welch's ANOVA is an appropriate method for comparing sample groups with normal distributions but unequal variances.

The normality of the distributions was confirmed by the results of the Shapiro-Wilk test, except for Ni, Rb, and V. Most of the relative standard deviation (%RSD) values ranged between 5% and 15% for both the Goryeo wares and the Ru wares. The variation for Al, Si, Ca, and Ni was low for the two groups, which reflects a strong intragroup homogeneity for these elements. In contrast, the P %RSD values are high (> 20%) for both groups. For the other elements, the variation is typically higher for the Goryeo wares.

The results of the Welch's ANOVA indicate significant differences at the 0.05 level between the means of the Goryeo wares and the Ru wares for Al, Si, K, Fe, Mn, Ti, Cu, and Sr. The very low *p*-values (< 0.0001) for Mn and Sr confirmed the discriminative power of these two elements, corroborating observations made on the XRF spectra. The low *p*-values for Al, Fe, Cu, and Ti indicate that these elements can also be used to differentiate the two groups of celadon after data normalisation. On average, the Ru wares have higher values for Al, K, Fe, Ti, and Cu and lower contents of Mn, Sr, and Si. The Mn-Sr scatter plot (Fig. 3) displays a clear distinction between the Ru wares and the Goryeo wares, the latter having higher values for both Mn and Sr. The Ru wares show less scatter than the Goryeo wares, reflecting a higher homogeneity between the glazes of the different vessels. The CI ellipses are not overlapping, and the position of PDF.59 is clearly amongst the Ru wares on this plot.

On the Al-Ti and Fe-Cu scatter plots (Fig. 4), the distinction between the two groups is still visible, but the ellipses are overlapping. On the Al-Ti plot, three Goryeo celadons are included in both the Ru wares ellipse and the Goryeo wares ellipse, but PDF.59 is located only inside the Ru wares ellipse. The overlapping between the two groups is more obvious on the Fe-Cu scatter plot, where the two ellipses encompass several samples from the two groups as well as PDF.59.

To achieve a distinction between the Ru and the Goryeo wares based on a multivariate comparison, the values of the 16 elements identified in the spectra were processed using a varimax-rotated PCA. The Zn values were discarded because of the very high %RSD in the Goryeo group. Because of the low number of samples and the higher number of variables (*p* > n*), a varimax rotation was applied on the PCA to maximise the sum of the variance of the squared loadings. The aim was to make stronger associations between the factors and the most meaningful variables to achieve a better interpretation of the results (Reimann et al., 2002).

After rotation of the first four principal components, the four rotated components (RCs) accounted for 79.9% of the total variance. The first rotated component, RC1, explained 33.6% of the variance and showed high positive loadings for Al, Ti, Fe, and Cu and high negative loadings for Mn and Sr. The RC2 explained 15.6% of the variance and is characterised by high positive loadings for Si and V and high negative loadings for Ca. The RC1-RC2 plot (Fig. 5a) shows a clear pattern discriminating
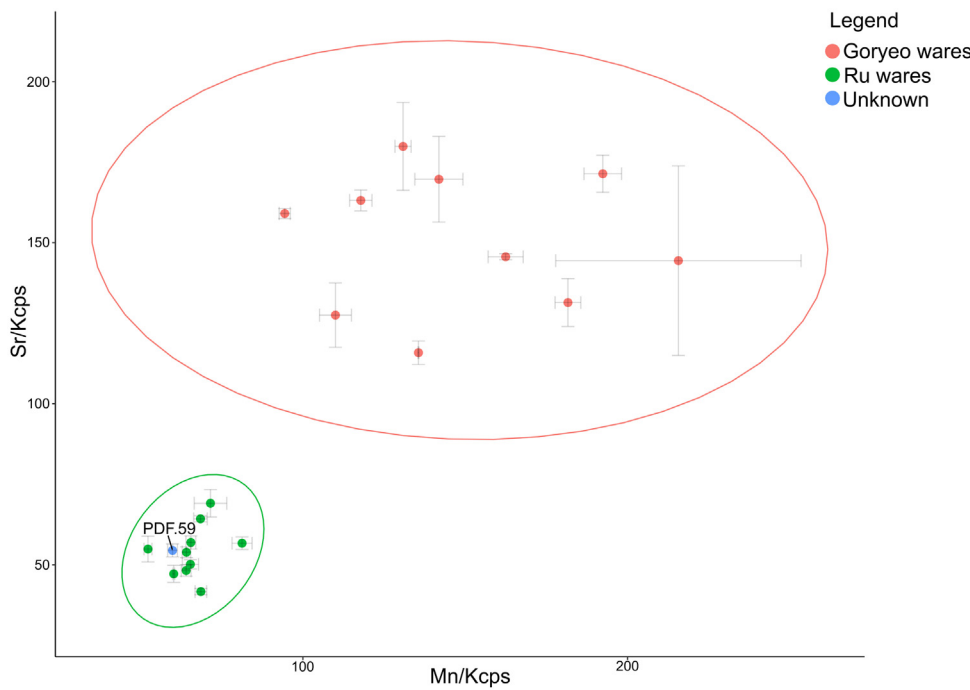
Fig. 3. Mn-Sr scatter plot with error bars showing the standard deviations (1 sigma) calculated between the three spectra averaged and the 95% CI ellipses for the two groups of celadon.
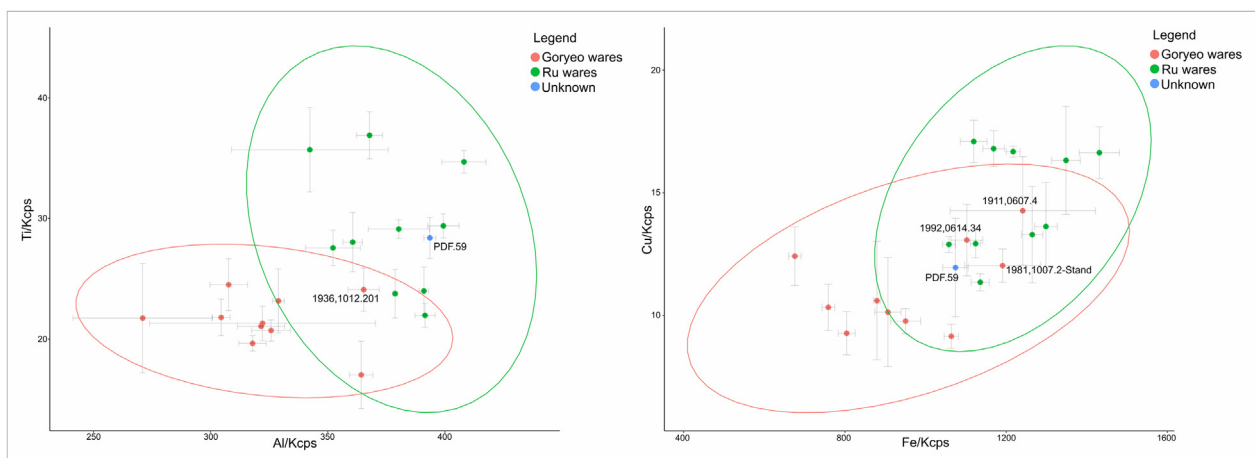

Fig. 4. Al-Ti and Fe-Cu scatter plots with error bars showing the standard deviations (1 sigma) calculated between the three spectra averaged and the 95% CI ellipses for the two groups of celadon.

the Ru wares on the right side of the plot, with positive coordinates on RC1, and the Goryeo wares on the left, with negative coordinates. On this plot, PDF.59 is close to the Ru ware sample points. The position of this sample on RC2 reflects higher values for Si and V but is still in the range of the Ru ware group. The third rotated component, RC3, explained 15.9% of the variance, with high positive loadings for Mg and P, and RC4 explained 14.6% of the variance, with high positive loadings for Ba, Ni, and Rb. The RC3-RC4 plot (Fig. 5b) does not show any pattern related to the origin of the ceramics. The position of the samples on this plot is likely related to chemical variations inside the two groups investigated.

Comparison between the results of the rotated PCA and the Welch's ANOVA shows that RC1 includes all elements with a strong discrimination power between the Goryeo wares and the Ru wares except Si. Because of the good agreement between these two results and the relatively high eigenvalue of RC1, the position of the sample points on this component should be interpreted as more meaningful when considering the origin of the ceramics. The position of PDF.59 on this component is wholly consistent with the Ru wares.

Discussion

Handheld XRF has proven to be an efficient instrument to discriminate between Ru and Goryeo wares. Even with an analysis time as short as 15 s, the five measurements taken on each sample provided useful data to identify differences in the elemental composition of the glazes. The Ru wares are mainly discriminated from the Goryeo wares because of their higher counts per second for Al, K, Cu, Ti, and Fe and lower content of Sr and Mn. This pattern is likely to reflect the use of different materials for the preparation of glazes and/or different production processes. The absence of quantitative calibration in our results prevents the comparison of this data to others published elsewhere. However, the presence of some correlation patterns can be used to suggest a hypothesis regarding the origin of the chemical differences observed. In the Goryeo wares, Mn is correlated with P and Mg, while these positive correlations are absent from the Ru wares (Fig. 6).

The use of Mg-, Mn-, and P-rich plant or wood ashes as a flux for celadon glaze has already been recorded for Chinese and Goryeo celadons, and the positive correlation between P/Mg and P/Mn is often
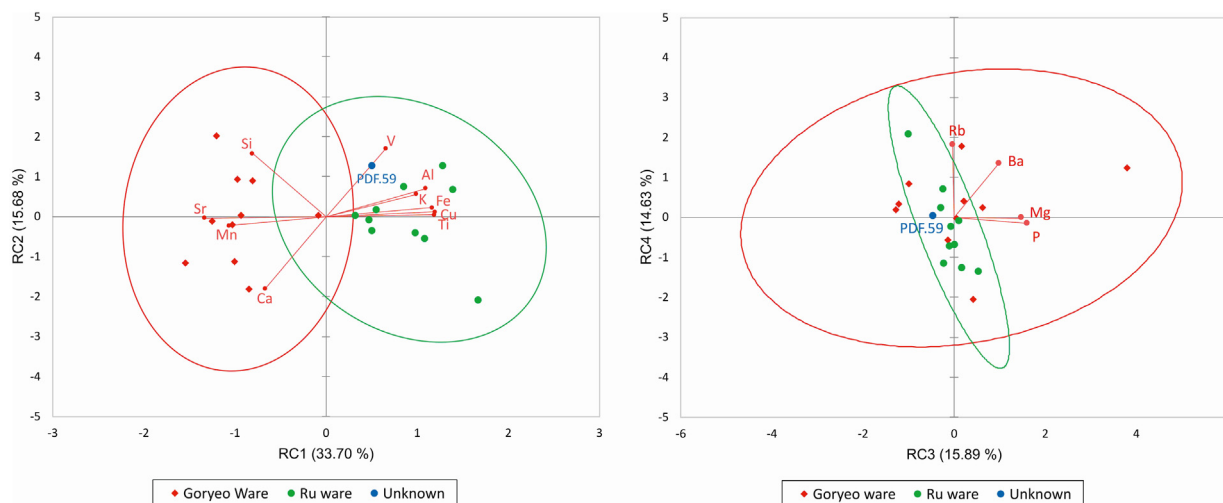
Fig. 5. Biplots of the varimax-rotated PCA with 95% CI ellipses for the two groups of celadon. (5a) Left biplot RC1-RC2. (5b) Right biplot RC3-RC4. The variables displayed on the plots are filtered with a threshold on Cos^2 on the two components > 0.5 after rotation.
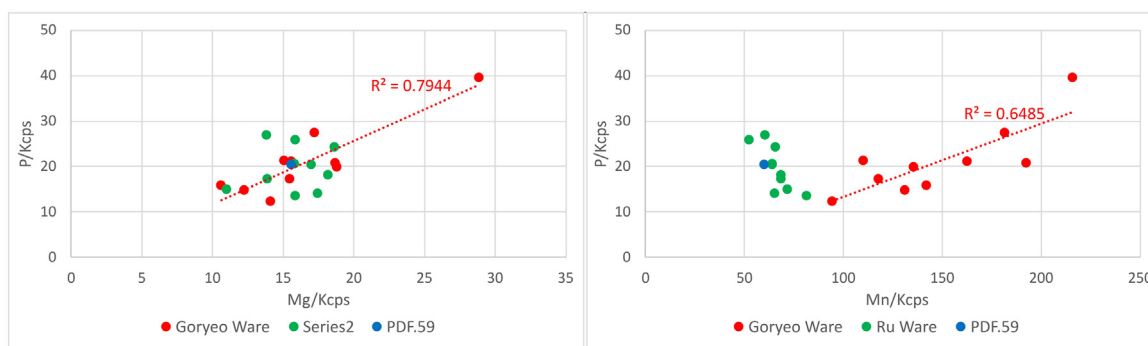

Fig. 6. Scatter plots of P/Mg and P/Mn. The good determination coefficient for the Goryeo wares might indicate the use of wood ashes or plant ashes rich in Mg and Mn.

regarded as a fingerprint for this type of material (Ham et al., 2002; Shi et al., 2020; Zhou et al., 2020). It must be noted that because of the high variability of the ash composition, depending on the type of plant or wood used (Huang et al., 2020), the absence of elemental positive correlation with P in the Ru wares does not indicate that no ashes were used in the recipe of the glaze. Moreover, mineral materials such as grounded limestone, feldspars, or muscovite could also have been used as flux for the glaze, but our data cannot clearly provide more details about this. There is a greater similarity of the compositions of the 10 Ru ware objects (along with PDF.59) than with the Goryeo group, as seen from the %RSD in Table 2. This tentatively suggests that the compositional and methodological control of production in the Ru kilns was better or tighter. It could also reflect that the kilns were in operation for a relatively short period of time.

Conclusion

The ability of XRF to separate Goryeo wares and Ru wares allowed us to attribute the celadon PDF.59 to the Ru ware group with a high degree of certainty—the position of this sample in the Mn-Sr scatter plot provides the best evidence of its origin. The high discrimination power of these two elements and the very good homogeneity of the Ru wares make this a useful screening tool for assessing provenance. Other information about the raw materials and high degree of control of the Ru kilns is evident from elemental correlations and the tight cluster of the compositional data. The method presented here shows a very simple, non-destructive, and easy process for distinguishing Ru from the

compositionally and stylistically very similar Goryeo ware. It shows the potential for further work in distinguishing other often-confused wares.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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