Comparison of IR Spectra of Five Porcelain Compositions Prepared Using Algerian Raw Materials

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Abstract— **The interest in Infrared (IR) spectroscopy is an essential technique for the characterization of ceramic. Its main advantage when it comes to studying items is that it is non-destructive: the samples can be investigated without further preparation. IR spectroscopy probes molecular and crystal lattice vibrations and therefore is sensitive to the compositions, chemical environment, bonding and crystalline/amorphous structure of a sample material. Five Porcelain compositions were studied by non-destructive infra Raman spectroscopy: kaolin was derived from Debagh deposit (Guelma region Algeria), Quartz was derived from El Oued region in Algeria and PF was derived from Ain Barbar deposit (Annaba region Algeria) and both commercial Alumina and talc. Raman intensity is a very powerful tool, which allows investigating and characterizing the modifications of the structure in covalent bonded compounds. Hence the observed porcelain spectral feature at ~1050 cm^{-1} is due to SiO$_2$ for the five compositions. From our study, as well as the literature reviewed it appears that investigators uniformly confirm the benefits of IR spectroscopy concerning identification of chemical functional groups of the different chemical compositions.**

Keywords— **porcelain; IR; quartz; alumina; talc**

I. INTRODUCTION

Raman and IR spectroscopy is the study of the scattering, reflection, absorption or transmission of radiation in the spectral range of 12500 to 10 cm^{-1} (wave numbers). The mid-sub-region (4000-400 cm^{-1}) of infrared spectrum is widely used in the analysis of chemical compound by IR spectroscopy, which obtains spectra by measuring the light absorption (or transmission) [1]. Raman spectroscopy is an optical technique and, thus, can be performed through different optical devices: camera lenses, microscopes, remote fiber optic probe, etc. Raman analysis of the micro-structure (for ceramics) offers a way to identify the composition, the processing and, sometimes, the age of ancient artefacts [2].

Kaolin, feldspar and quartz are used as raw materials in conventional porcelain production. If the raw material ratios are changed or if one of these raw materials is replaced by some other materials, the characteristics of the prepared porcelains change. The physical properties vary depending on the types of the raw porcelain batch components. Porcelain bodies have now been studied and comparisons made in respect of their compositions.

In the present integrative study, five categories of porcelain were submitted to FTIR analysis in order to check the structural changes during the process. The advantage of using this technique as complementary tool is discussed.

II. EXPERIMENTAL PROCEDURES

A. Materials

The samples were prepared using raw materials typically used in the production of porcelain. The experimental compositions of the porcelain bodies were prepared from Kaolin, Quartz, Feldspar, Alumina and Talc. On the basis of chemical properties of the raw materials which showed in an earlier paper [3], the composition of the porcelain bodies was estimated without any additives. Five compositions (PDl, PD2, PD3 PD4 and PD5) were obtained. The experiments were carried out on the prepared porcelain samples obtained from an industrial firing at 1200 ∘C. The five formulations are reported in Table 1.

TABLE I

	PD1	PD2	PD3	PD4	PD5
Kaolin DD3 (wt %)	45.00	47.00	47.00	22.50	-
Feldspar Potassium (wt %)	30.00	22.00	22.00	30.00	30.00
Quartz (wt %)	25.00	31.00	-	25.00	25.00
Al$_2$O$_3$ (wt %)	-	-	31.00	-	-
Talc (wt %)	-	-	-	22.50	45.00

B. Sample Preparation and Spectra recording

The Infrared spectroscopy (FT-IR) was recorded on a Nicolet Thermo - RI 300 spectrometer using the potassium bromide (KBr) pellet technique.

III. RESULTS AND DISCUSSIONS

Raman spectra give information on the phase nature. Since the Raman cross section is directly related to the number of electron involved in the covalent bond, the technique efficiency is very variable. For the determination of the mineral constitution the Fourier transform infrared spectroscopy (FTIR) have proved to be suitable method. With the help of vibrational spectroscopy, apart from good crystalline, also poor crystalline and amorphous inorganic components and organic substances can be identified. Good quality Raman spectra were obtained from the porcelain bodies. The FTIR spectra obtained for five mixtures show that typical bands assigned to kaolinite, quartz, alumina and talc the main minerals present in the raw materials, are distinguished in the IR spectra. By comparing Raman spectra of five mixtures, it is clearly appears that the Raman intensity of Si-O bending and stretching envelopes vary with composition. In IR spectra of porcelain samples, the similarities were found at ~ 465, ~720, and 1050 cm^{-1} (Fig.1, 2, 3, 4 and 5).
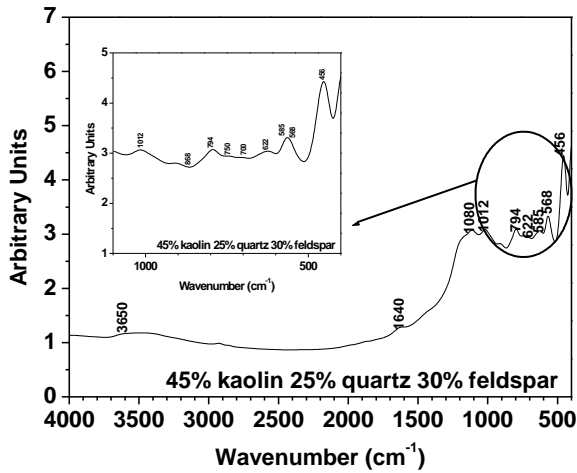
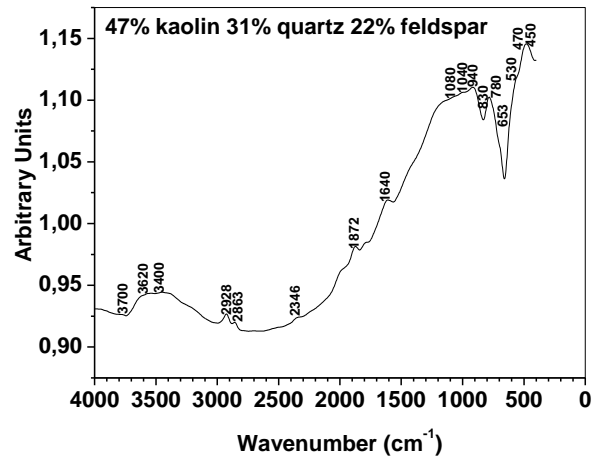

Fig. 2. Infrared spectra of PD2
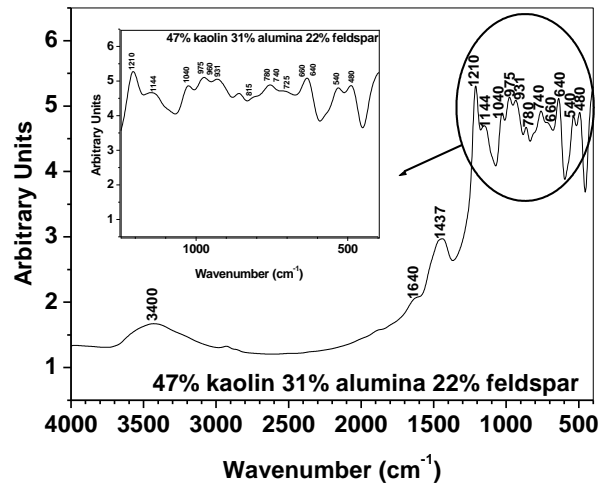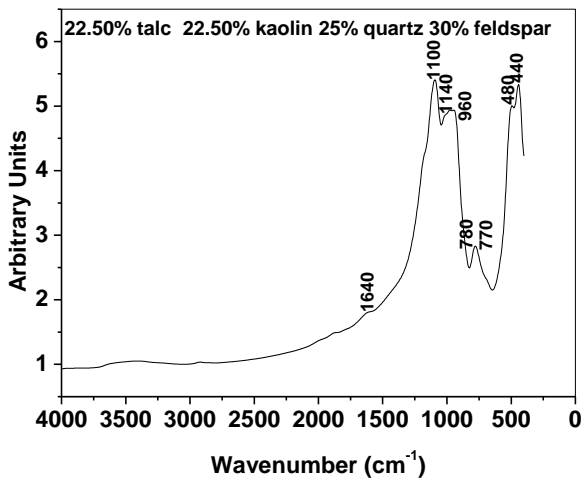

Fig. 1. Infrared spectra of PD1
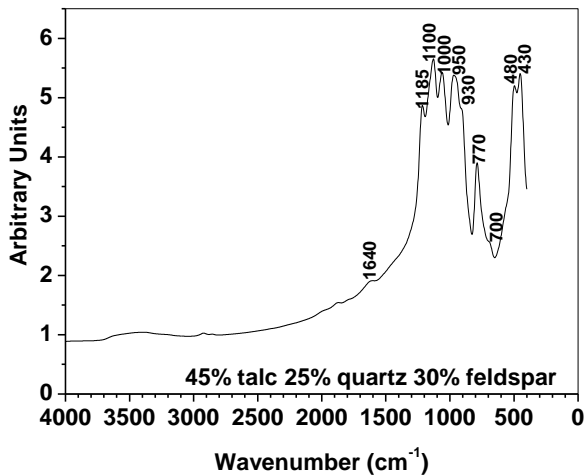

Fig. 3. Infrared spectra of PD3

Fig. 4. Infrared spectra of PD4

Fig. 5. Infrared spectra of PD5

The stronger narrow peak observed at ~458 cm⁻¹ in all spectra records on the body is present in most porcelain and arises from α-SiO$_2$ (α-quartz) . This Raman line corresponds in a displacement of the bridging oxygen along a line bisecting the Si-O-Si angle, i.e. originating from the bending vibration of isolated tetrahedral, and is the strongest mode for structures composed of connected SiO$_2$ tetrahedral units (tectosilicate structure). Thus, the mode is very sensitive to the modification of the tetrahedral network imposed by the vicinity of alkali cations [4].

Another main band in porcelain located around 1050 cm⁻¹ and originates from Si-O stretching mode. The centre of gravity of the two bands (458 cm⁻¹ and 1050 cm⁻¹) is very different depending on the kind of ceramics. In structures made of weakly connected tetrahedral units, the starching mode-related oxygen motion has large amplitude.

This induces a large polarisability change, hence a high intensity for corresponding peak at 1050 cm⁻¹. In contrast, in highly connected tetrahedral structures, the large amplitude oxygen motion takes place perpendicularly to the Si-O-Si link and the higher polarisation change, hence the larger Raman peak, is the bending mode around 458 cm⁻¹ [4]. The appearance of a new band at 1050 cm⁻¹ indicates that the quartz becomes partially amorphized. The shift of the 1080 cm⁻¹ band could be due to a change in the bond angle and therefore the vibrational frequency of the Si-O-Si group. This indicates the presence of crystalline quartz. This fact is also confirmed by the presence of the 740 and 550 cm⁻¹ bands, characteristic of only the crystalline quartz [5]. On the other hand, mullite or mullite-like glassy phase spectra (main bands at ~ 480, 960 and 1130 cm⁻¹) are observed in (prepared with kaolin and feldspars) [2].

A peak at ~517cm⁻¹ is clearly observed in Fig.1. This peak is characteristic of feldspathic porcelain bodies and is assigned to the signature of feldspar [6]. A broad band with a peak at ~3400 cm⁻¹ appeared in Fig.2 and 3 which might be interpreted as a higher level of hydroxyl water. However, further studies are necessary to confirm this trend and to understand how excess water may affect the crystalline content of porcelain. Other IR peaks and bands showed comparable results for fives samples. They were determined to correspond to H-O-H bonds (1640 cm⁻¹). By comparing Raman spectra of Fig.2 and 3, it appears clearly that the Raman cross-section of Si-O bending and stretching modes is orders of magnitude stronger than that of Al-O bond counterparts. This arises from the more covalent character of the Si-O bond. Thus in a first approximation we could consider the SiO$_4$ entities as the vibrational unit [7]. Fig. 3 show the absorption peak at around 630 cm⁻¹ which corresponds to Al–O bonds [8].

Close similarities exist in the pattern of the 1400-400 cm⁻¹ lattice vibrational region for PD4 and PD5. A strong band around 440 cm⁻¹ is essentially attributed to Mg-O-stretching vibrations [9]. Bands centered in PD4 at 1137 and 1050 cm⁻¹ are assigned to Si-O stretching vibrations. A broad band at 1137cm⁻¹ corresponds to Si–O bonds of amorphous SiO$_2$. This band is splitting at crystalline curve, which results from some ordering in Al and Si in tetrahedral site. The position of minimum value shifted toward a lower band than those of the amorphous SiO$_2$, which indicates the formation of Si–O–M (M= Al, Mg). A weak absorption at 780 cm⁻¹ is assigned to stretching modes of AlO4 tetrahedral units [10].

IV. CONCLUSIONS

The Macro- and micro-Raman spectroscopy is a good tool for the non-destructive remote identification of porcelain. There is a clear relationship between the composition of the phases of the final ceramic product and the component present in the raw material. FTIR spectra give additional support to XRD results.

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