

Research Article





Silicates in Kamargaon (L6) chondrite: A Raman spectroscopic study

Abstract

The Raman spectroscopic technique has been utilized to determine the silicate composition and structure of Kamargaon meteorite (an ordinary chondrite L6 type; Kamargaon: 26°39'01 " N and 93° 46' 02 " E, India). The micro-Raman spectra in the range of 100-2000cm-1 revealed principal characteristic bands of the major minerals: olivine, pyroxene and plagioclase. Fourier transformed infrared technique is used as complementary to the Laser Raman spectroscopic method. The presence of some mineral phases such as kamacite and taenite as well as troilite and chromite were determined by X-ray diffraction method. The present study demonstrates the usefulness of Laser-Raman spectroscopic method in identifying high pressure mineral phases which are present in the shocked meteorite.

Keywords: silicate, Raman, Kamargaon, infrared, x-ray diffraction

Introduction

Meteorites are the rocks from outer space that helps us to understand the origin and evolution of solar system, and various processes related to the early solar system. The Kamargaon chondrite (Figure 1) is a single fall (Kamargaon: 26°39′01″ N and 93° 46′ 02″ E), on 13th November, 2015; its total known weight 12.095kg.^{1–3} Earlier studies focused on the compositional analyses of the meteorite and assigned Kamargaon as an L6 ordinary chondrite.^{1–3} A cosmic ray exposure age of Kamargaon is 7 Ma.² Presence of irregular fractures of olivine and pyroxene grains observed previous authors² and entails the stage of shock metamorphism of Kamargaon is S3.



Figure I Photograph of a fraction of the Kamargaon (L6) chondrite.

The Raman, infrared and X–ray diffraction processes are the most essential tools in mineralogical research.⁴ These techniques play an imperative role in the in–situ studies of major and minor minerals in stony meteorites samples.⁵ Raman spectroscopy is a non destructive, non–contact, highly sensitive powerful rapid technique for investigating the structure and composition of materials. Raman spectroscopy has been used for the theoretical modeling of inorganic and organic materials, mathematical modeling for biomolecular quantification etc. The major advantage of this technique is its applicability of wide Volume I Issue 6 - 2018

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range of substances, and has no need for sample preparation prior the analysis. Though this technique can applied on measuring an extend spectrum of materials, metals and alloys are not Raman active. Additionally we always have to consider fluorescence interference in Raman measurements. The Raman spectra entrenched the chemical and structural information of meteoritic minerals. Although such compositions are not as accurate to those determined by electron microprobe technique, but the potential of Raman spectroscopic technique has been largely exploited for the identification of shockinduced features or polymorphs in meteorites.^{6,7} However, the typical textures produces as a result of structural deformation of the crystals by shock can be easily visible under the microscope. Raman spectroscopy can be used to study such deformations.8 However, Pittarello et al.,⁹ pointed out Raman spectroscopy as an alternative technique to characterization of meteorite.9 There were many studies on mineralogical and geochemical characterization of meteorites which fell in India, but there were no reports on spectroscopic studies on meteorites that fell in India were carried out.¹⁰⁻¹³ Raman spectroscopy has already been used for meteorite characterization and identification of shock-induced features,7-9 this paper reports the vibrational characteristics of olivine and pyroxene on the ordinary chondrite and to investigate the presence of any high pressure phase in Kamargaon.

Methods

Raman spectroscopy

The Laser–Raman spectra were collected on bulk meteorite sample with a Jobin–Yvon Horiba LabRam–HR Micro Raman spectrometer using Nd: YAG laser with a power of ~5mW as an illumination source having wavelength 532nm. The Raman instrument was equipped with an Olympus microscope with 10X, 50X and 100X objectives, using the method described elsewhere.¹⁴ A motorized x–y stage was included in this arrangement and using 1800 grooves /mm grating in the range from 100 to 3000cm⁻¹. Powdered samples (~20mg) were selected for the present investigation instead of polished thin sections, as the latter will have textural and crystallite orientation effects in spectroscopic and powder XRD studies. An edge filter was used, for measuring the exact Stokes lines. The data interpretation procedure used a Gaussian

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a needs 14 Minerals 700, 1050 cm⁻¹. The meeds held

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fit to find the exact position of the maximum of each peak.¹⁴ Minerals were identified by comparing the band positions in our spectra with the standard Raman data (RRUFF–database http://rruff.info/). Spectra were collected with counting times ranging between 10 and 60s.

Infrared spectroscopy

The infrared spectrum was acquired using Perkin–Elmer system 2000 FTIR spectrophotometer with helium–neon laser as the source reference, at a resolution of 4cm⁻¹. The powdered sample was homogenized in spectrophotometric grade KBr (1:20) in an agate mortar and was pressed 3mm pellets with a hand press. The experimental condition was identical to those used in studies^{15–17} of Dergaon and Mahadevpur meteorites.

X-ray diffraction

The composition of the powdered meteorite sample was determined by the X–ray diffraction technique (XRD) using PHILIPS PW 3710/ 31 diffractometer, scintillation counter, Cu*Ka* radiation (λ =1.5406Å) and Ni filter at 40 kV and 35 mA. This instrument is connected to a computer system using APD program and PDF–2 database for mineral identification. We used a 20 range of 10–80° with a step size of 0.02° and a 0.5s count time per step. The slits used consisted of 1° fixed divergence and anti–scatter slits and a 0.2mm receiving slit.¹⁴

Results and Discussion

Raman spectra of Kamargaon reveal the mineralogical compositions (Table 1) that typical to the stony chondrites. Figure 2 shows the Raman spectra in the range of 100–1200cm⁻¹ that associated to the principal characteristic bands of olivine, pyroxene and plagioclase. Out of 81 optic modes of olivine, only 36 are Raman active.¹⁸ Detailed olivine Raman peak assignments has been reported by Chopelas.¹⁹ The olivine Raman spectrum is generally divided into three spectral regions,²³ i.e. below 400cm⁻¹, 400–700cm⁻¹, and

700–1050cm⁻¹. The peaks below 400cm⁻¹ are commonly referred as lattice modes that arise due to rotational and translational motions of SiO, units, and translational motions of octahedral crystal lattice. The peaks in between 400–700cm⁻¹ are attributed to the internal bending vibrational modes of the SiO₄ ionic groups.¹² The internal stretching vibration modes of the SiO, ionic groups are generally observed in the region of 700–1050cm⁻¹. In the multi–phase spectra, olivine minerals are commonly identified from the characteristic doublet near 820cm⁻¹ and 850 cm^{-1} . This doublet is attributed to the couple symmetric (v1) stretching and anti-symmetric (v3) stretching modes of Si-O bonds in SiO₄ tetrahedra.²⁰⁻²² In general, five characteristic peaks have been identified for olivine ²³ based on the characteristic SiO, vibrational modes. These are: peak 1 (819-826cm⁻¹), peak 2 (849-858cm⁻¹), peak 3 (881-883cm⁻¹), peak 4 (914-920cm⁻¹), and peak 5 (951-967cm⁻¹). These characteristic peaks are found in Kamargaon meteorite at wavenumbers 821, 853, 922 and 957cm⁻¹. The peaks 821, 853 and 957cm^{-1} in Kamargaon spectra are assigned to the A symmetry and peak at 922cm⁻¹ to $B_{3\sigma}$ symmetry of forsterite mineral.^{1,14}



Figure 2 Raman spectra of some minerals recorded on the Kamargaon meteorite; a) mixture of minerals (pyroxene + olivine + plagioclase); b) pyroxene.

Table I Raman peak position (cm ⁻¹) for Kamargaon meteorite (OI: olivine, Px: pyroxene, OPx: orthopyroxene, CPx: clinopyrox
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Wavenumber	Composition	Assignments	Wavenumber	Composition	Assignments	
178	PI	T-O-T lattice	579	СРх	O-Si-O bend	
224	Px	M-O stretch	644	OI	Si-O-Si bend	
234	OPx	M-O stretch	660	OPx	Si-O-Si bend	
246	CPx	Ca-O stretch	668	CPx	Si-O-Si bend	
254	CPx	Ca-O stretch	680	OPx	Si-O-Si bend	
285	PI	T-O-T lattice	690	OPx	Si-O-Si bend	
293	Px	Mg-O stretch	746	Px	Si-O-Si bend	
301	OPx	M-O stretch	752	CPx	Si-O-Si bend	
323	CPx	M-O stretch	821	OI	v1(Si-O-Si)	
337	OPx	M-O stretch	853	OI	v3(Si-O-Si)	
387	Px	Mg-O stretch	922	OI	v3 anti sym. stretch	
405	OPx	Ca-O stretch	938	OPx	Si-O(br) stretch	
411	Px	Ca-O stretch	957	OI	v3 anti sym. stretch	
427	CPx	Mg-O stretch	982	OI	Si-O(br) stretch	
475	PI	T-O-T band/stretch	1008	OPx	Si-O(br) stretch	
510	PI	T-O-T band/stretch	1012	CPx	Si-O bend	
548	Px	O-Si-O bend	1025	OPx	Si-O bend	
570	PI	Si-O-Si	1048	CPx	Si-O bend	

The Raman spectra of olivine observed in different points of Kamargaon meteorite reveal homogeneity in chemical compositions (Figure 3). The peak positions of the doublets vary only about 820-822cm⁻¹ and 853–854cm⁻¹. The relative height of the characteristic doublet of olivine is a function of crystal orientation. The peak positions of the doublet vary with fayalite (F_{a}) /forsterite (F_{a}) composition. It is reported ²⁴ that the peak positions shift upwards with increasing of the F_0 values ($F_0=Mg/Mg+Fe$), which can be used to determine the fayalite and/or forsterite content in the solid solution of the olivine minerals. Using Raman spectral data with compositional (Mg/ Mg+Fe) ratios,²⁴ it is found that approximately about 65 to 89 mol% of forsterite is present in Kamargaon. Presence of forsterite is also observedin X-ray diffraction analysis. Full width at half maximum (FWHM) value determine for olivine Raman line at \sim 820cm⁻¹ (v1) is ~17cm⁻¹ and this value is identical to that of strongly shock stage.¹ The SiO₄ stretching vibrational modes of the peaks 821cm⁻¹ and 852cm⁻¹ shifted to the higher lower wavenumber region suggests that there is no coordination change of Si.



Figure 3 Raman spectra of the olivine recorded from different points of the Kamargaon meteorite which show homogeneity in chemical composition.

Raman spectra of pyroxene characterized ²⁵ by asymmetric peaks near 1000 cm⁻¹, asymmetric single or double peak at ~670cm⁻¹, and four peaks in the range 200-400cm⁻¹. The frequencies of the Raman peaks are gradually shift with Mg/Fe and W_o (wollastonite) content in pyroxenes. Different cations (Mg, Fe, Ca, etc.) are responsible for the translations and tilt or torsion motions of SiO₄ tetrahedra,²⁶ which exhibits the bands below 600cm⁻¹. In general the titling and torsion observed correspondingly 600-500cm⁻¹ and 500-300cm⁻¹. The Raman spectra of Kamargaon (Figure 2) were characterized by the vibrational modes as: three peaks observed at 234, 301, 337cm⁻¹ below 360cm⁻¹, one peak observed at 680cm⁻¹ in the range from 600– 700cm^{-1} , and four peaks observed at 938, 1008, 1012 and 1048cm^{-1} in the range 900-1050cm⁻¹. The spectral pattern with the peaks at 334, 680 and 1008cm⁻¹ are indicative to orthopyroxene.²⁵ However, the Raman peaks at 323, 427, 579 and 1012cm⁻¹ has indicative to a spectral pattern of clinoenstatite.²⁷ Some weak phase of pyroxenes is also observed in the Raman spectra of Kamargaon.25

In Raman spectra, the symmetric T–O stretching modes and O–T–O stretching and deformation modes of TO₄ (where T being Si or Al) appears ~510cm⁻¹, the T–O–T lattice modes arises ~285 cm⁻¹, and the lattice T–O–T and T–O lattice modes are generally observed in between 170 to 180cm⁻¹. These bands were used to identify plagioclases in Kamargaon meteorite. The observed peak positions at 510cm⁻¹ and 178cm⁻¹ suggests the presence of plagioclase in the Kamrgaon meteorite sample. The presence of plagioclase indicates to weak nature shock²⁸ in Kamargaon, which have been

already reported.² The antisymmetric Si–O stretching vibrations and antisymmetric O–Si(Al)–O deformations exhibits in the mid infrared spectra of Kamargaon correspondingly between 800 to 1150cm⁻¹ and 400cm⁻¹ to 500cm⁻¹. These band profiles are generally depends on the crystalline structure of the silicates and can therefore be used to identify the mineral phases. Mid infrared spectra of Kamargaon indicates the presence of a mixture of olivine, pyroxenes, plagioclase and chromite (Table 2).

The infrared–active Si–O modes in olivine are reported ^{29–30} at around 900–1000cm⁻¹; the strong bands at 965cm⁻¹ and 1075cm⁻¹ arises in pyroxene,³¹ similarly three bands at 995cm⁻¹, 1145cm⁻¹ and 1160cm⁻¹ are arises in plagioclase.³² Moreover, the band found at 508cm⁻¹ in Kamargaon spectra can be interpreted as Si–O and Mg–O vibration modes in enstatite. The peaks at 995–1057cm⁻¹ arise due to Si–O asymmetric stretching vibration (TO₂–T₂O₅). The peaks in the range 913–972cm⁻¹ and 874–884cm⁻¹ correspondingly arise due to Si–O asymmetric vibrations (TO₃) and (T₂O₇–TO₄). The Si–O–Si bending vibrations are found in between 458–495 cm⁻¹. The peak ~687cm⁻¹ attributed to the symmetrical bending vibration of O–Si (Al)–O. The infrared spectra of chromite in L6 meteorite has been reported by Gyollai et al.,³³ The infrared peaks at 524, 703, 935, 1424, 1458, 1543 and 1648cm⁻¹ indicates to presence of chromite in Komargaon meteorite.

The X-ray diffraction revealed pyroxene, olivine, plagioclase, and some mineral phases like: kamasite, taenite, troilite, and chromite (Table 3). The electron microprobe analysis also exhibits the evidence of these minerals. Average mineral phase compositions are determined using EPMA has been discussed by the author elsewhere.¹ The abundances include ~45% olivine, ~33% pyroxene, 8% feldspar (maskelynite), 8% metal, 4% troilite, and 2% other accessories (e.g. chromites). Fracturing phases of silicate is observed. The recrystallized feldspar with irregular shaped (>100µm in size) and emerges as maskelynite. Within the matrix, quenched metalsulfide melt texture and shock melt veins are observed. The general composition of metals in quenched textures are kamacite, while the troilite and taenite are also occurs within the matrix. These are occur both as separate grains and paired assemblages. Based on several grains analyses, the olivine compositions show a restricted range (Fa: 22.9±2.1 and Fo:77.24±1.74). The mean pyroxene composition is $Wo_{1.48 \,\pm\, 0.2} En_{77.83 \,\pm\, 0.7} Fs_{20.68 \,\pm\, 0.4}$. Kamacite shows Ni 6.8±0.4 wt% and has Co content of 7.0mg/g. In taenite, Ni ranges between 20 to 22 wt% and composition of trolite is Fe ~63wt% and S ~37wt%. Usually the Fa content of L 5/6 meteorites (e.g Araki, Kaprada, Katol) lies in the region 23.0 to 25.8mol %; $\mathrm{F_s}$ content 18.7 to 22.6mol % and Co content in kamacite is 7.0mg/g. The concentration of Co inkamacite is match with the range (0.7-1.0wt%) observed by many authors in other L chondrites. $^{\rm 34-35}$ The planner fractures within olivine and pyroxene grains indicate shock metamorphism of Kamargaon and presence of veins of feldspar and troilite suggest shock stage belong to S3-S4.36 As the Raman spectroscopy is not sensitive to most pure metals and alloys, therefore these phases did not exhibits characteristic Raman spectra. Additionally, in Raman and infrared spectroscopy, Fe-Ni metal has no active modes; andtroilite is considered as a weak Raman scatterer.37 Generally, Raman peak position of chromite exhibits a peak shift in between 680cm⁻¹ to 770cm⁻¹, which overlaps the pyroxene peak maxima. All identified mineral phases using Raman and infrared spectroscopy have been confirmed by the X-ray diffraction technique.

Wave number	Compound	Assignment	Wave number	Compound	Assignment
409	Px	Mg-Si translation	692	Px	Si-O-Si stretching
419	OI	v2(Si-O-Si)	703	Ch	M-O stretching
427	OI	Mg-Si translation	821	OI	Si-O (sym) stretching
438	Px	Si-O-Si lattice	839	OI	vI (Si-O-Si)
458	Px	Mg-Si translation	874	Px	Si-O (br) stretching
465	Px	Mg-Si translation	884	OI	v3 (Si-O-Si)
474	OI	Internal bending	904	Px	Si-O-Si (asym.) stretching
495	OI	Internal bending	912	Px	Si-O-Si (asym.) stretching
508	Px	Internal bending	924	OI	Si-O (br) stretching
524	Ch	M-O stretching	935	Ch	M-O stretching
531	PI	O-Si(Al)-O	944	Px	SiO4(anti sym.) stretching
545	OI	Si-O-Si bending	958	OI	Si-O-Si (n br.) stretching
557	OI	Si-O-Si bending	966	Px	Si-O (asym.) stretching
566	OI	Si-O-Si bending	975	Px	Si-O (asym.) stretching
588	PI	Depolarization of SiO ₄	992	Px	Internal mode
595	PI	Depolarization of SiO ₄	1002	OI	v3 (Si-O-Si)
603	PI	Internal SiO ₄ vibration	1022	Px	Si-O-Si (n br.) stretching
615	OI	v4(Si-O-Si)	1057	OI	Si-O-Si (n br.) stretching
643	Px	Si-O deformation	1424	Ch	M-O stretching
658	Px	Si-O deformation	1458	Ch	M-O stretching
660	Px	Si-O deformation	1543	Ch	M-O stretching
672	Px	Si-O deformation	1648	Ch	M-O stretching

Table 2 Infrared peak position (cm ⁻¹) for Kamargaon meteorite	(Ol: olivine, Px: pyroxene, Pl: plagioclase, Ch: chromite)
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Table 3 X-ray diffraction (Å) of Kamargaon meteorite; Kam: kamacite; Tae: taenite; Tr: troilite; Ch: chromite; Fo: forsterite; Fa: fayalite; Px: pyroxene

Kamargaon	Kam	Тае	Tr	Ch	Fo	Fa	Рх
5.097	-	-	-	-	5.113	-	-
3.891	-	-	-	-	3.89	-	-
3.729	-	-	-	-	3.728	-	-
3.654	-	-	-	-	-	3.587	-
3.347	-	3.34	-	-	-	-	-
3.24	-	-	-	-	-	-	3.241
3.037	3.032	-	-	-	-	-	-
2.999	-	-	2.98	-	-	-	-
2.955	2.953	-	-	-	-	-	-
2.879	-	2.879	-	-	-	-	-
2.823	-	-	-	-	-	2.858	-
2.709	-	-	-	-	-	2.652	-
2.655	-	-	2.66	-	-	-	-
2.602	-	-	-	-	-	2.592	-
2.525	-	2.526	-	-	-	-	-
2.498	-	-	-	2.499	-	-	-
2.426	-	-	-	-	-	2.426	-

Table	Continued
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Kamargaon	Kam	Тае	Tr	Ch	Fo	Fa	Рх
2.317	-	-	-	-	2.319	-	-
2.275	-	2.279	-	-	-	-	-
2.232	-	-	-	-	-	-	2.233
2.186	-	2.187	-	-	-	-	-
2.068	-	2.07	-	-	-	-	-
2.03	2.031	-	-	-	-	-	-
1.998	-	-	-	-	-	-	1.999
1.969	-	-	1.967	-	-	-	-
1.924	-	-	1.923	-	-	-	-
1.877	-	-	-	-	-	-	1.878
1.868	-	-	-	-	-	-	1.867
1.864	-	-	-	-	1.864	-	-
1.813	-	-	-	-	1.813	-	-
1.781	-	-	-	-	-	1.781	-
1.722	-	-	1719	-	-	-	-
1.687	-	-	-	-	-	-	1.686
1.643	-	-	-	-	-	1.644	-
1.592	-	-	-	1.592	-	-	-
1.573	-	-	-	-	-	-	1.574
1.497	-	-	-	-	1.498	-	-
1.481	1.481	-	-	-	-	-	-
1.461	-	-	-	1.461	-	-	-

Conclusion

In this study silicate mineral of Kamargaon meteorite is analyzed using Raman and infrared spectroscopic techniques. The results of Raman and infrared spectroscopic analysis are found to be consistent with the electron-probe microanalysis and x-ray diffraction results. Predominance of olivine, pyroxene and plagioclase in Kamargaon meteorite were identified from both Raman and infrared analyses. Observed FWHM value of Raman line is identical to strongly shock stage. The mineral phases (kamacite, taenite, troilite, chromite) have been identified by X-ray diffraction technique. The infrared spectrum (peaks at 524, 703, 935, 1424, 1458, 1543 and 1648cm⁻¹) also indicate the existence of chromite. The X-ray diffraction technique permits to identify some mineral phases which have not been detected using Raman and infrared spectroscopic methods. Thus, each of the methods provided useful information about the meteorite. We conclude that Raman and infrared spectroscopic technique with XRD and EPMA, is a powerful tool for investigating shock metamorphosed meteorite.

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Conflicts of interest

Authors declare that there is no conflicts of interest.

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