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Morpho-chemistry of Pyrophyllite in the Schistose Rock in a part of the Singhbhum Craton, Odisha, India

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Abstract: *This paper discusses the morphological vs compositional variation of pyrophyllite from the schistose rocks exposed near Keonjhar. Morphologically, pyrophyllite in the study area can be grouped into six categories such as: Flaky, Laminated, Bladed, Fibrous, Platy and Granular. The first three types are enriched in alumina (Al_2O_3-31 to 33%) and poor in silica (SiO_2-49 to 56%) content while the other three varieties are poor in alumina (Al_2O_3-20 to 27%) but contain higher amount of silica (SiO_2-58 to 73%). Pyrophyllite shows different habits under scanning electron microscope. Compositional difference between these micro-morpho types are established through in-situ analysis by the SEM-EDAX system. A pristine pyrophyllite exhibits acicular radiating, anhedral crystalline, globular, massive, monoclinic crystalline and banded forms. Pyrophyllite containing Fe-impurities (6.9 to 18% Fe) appear in needle, rod and prismatic form. Similarly, pyrophyllite having Mg-impurities (platy & flaky) and Mg+Fe impurities (sub-hedral crystalline) show different habits. The K-containing pyrophyllite (K_2O: 4.6 to 8%) reveals different forms like: inter-grown, porous, spotted, bedded and ooloidal. A spongy / mesh structure is shown by pyrophyllite having ~ 11% Na as contaminant.*

The study demonstrates the inter-dependance nature between different habits and composition of pyrophyllite mineral in the schistose rock. Growth of pristine type or the one having impurities like Fe, Mg, K or Na depends upon their protolithic composition. The clean variety is most likely formed through solution and precipitation without any contamination. The Fe-containing pyrophyllite appears to have iron minerals in their inter-granular space, which might have been derived from the breaking down of biotite. Similarly, both Mg and Fe + Mg-containing pyrophyllite are derived from phologopite or biotite. Both K and Na-containing pyrophyllite have probably formed at the expense of orthoclase / mica and plagioclase respectively.

Keywords: Pyrophyllite; Singhbhum craton; Micro-morphological analysis; SEM

Pyrophyllite deposit in the Keonjhar district, Odisha extends over a strike length of about 90 km from Rebna-Palaspal in the south to Dhobakuchuda-Balabhadrapur in the north. It is a soft and pale-colored mineral, a common constituent of some schistose rocks. Pyrophyllite and its occurrence in India was first reported by Mishra (1953). It was followed by Saha, and Basak, (1965) who reported the occurrence of pyrophyllite associated with the Singhbhum Granite around Keonjhargarh, Orissa. Several others the world over; Evans and Guggenheim, 1988; Zaykov *et al.,* 1988; Zelazny and White, 1989, Rieger, 1997; Zaykov *et al.,* 1988; Harben, 2002; Sahoo, 2006) described pyrophyllite as an industrial mineral and worked on pyrophyllite

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deposits. In the study area it mostly occurs as small to medium discontinuous lensoidal pockets spatially associated with the Singhbhum granite phase-II (Yadav and Das, 2017). Characterisation of pyrophyllite in respect of mineralogy, petrology and geochemistry were discussed by many researchers (Roy and Osborn, 1954; Zen, 1961; Keats, 1970; Zelazny *et al.,* 2002; Das *et al.,* 2012; Oner and Tas, 2013; Pradhan *et al.* (2014); Yadav and Das (2017). SEM study of pyrophyllite for high temperature transformation was carried out by Sanchez-Soto and Perez- Rodriguez (1989). Sykes and Moody (1978) studied the pyrophyllite and metamorphism in the Carolina schist belt. The economic potential of Pyrophyllite deposits of Keonjhar as industrial mineral and its different industrial application has been reported by Pradhan *et al.* (2015) and Ali *et al.* (2021). Limited studies on genesis / origin of pyrophyllite have been undertaken by some researchers (Bryndzia, 1988; Henderson, 1970; Sharma, 1979; Marumo, 1989; Takagi, 2000; Uygun and Solakoglu, 2002; Das *et al.,* 2012) though a common consensus on this has not yet been arrived at. Limited SEM studies on pyrophyllite are reported in the literature (Sanchez-Soto and Pedro, 1989; Mukhopadhyay *et al.,* 2010). In this context the present study is pertinent, since it revealed the detailed insight of pyrophyllite phase from the study area.

In this contribution, we undertake a detailed study on morphological attributes of pyrophyllite and its corresponding compositional variation. Our objectives were (1) to obtain different morphological variation of pyrophyllite mineral through the field and laboratory investigation, (2) to find out the compositional difference between such morphologies, if any and (3) to acquire a comprehensive understanding of the mechanisms responsible for the formation of different morphologies and establish whether these are compositional dependant.

Materials and Methods

Balabhadrapur, located in the north of Keonjar town (Fig. 1) is the study area and where from the pyrophyllite samples were collected,

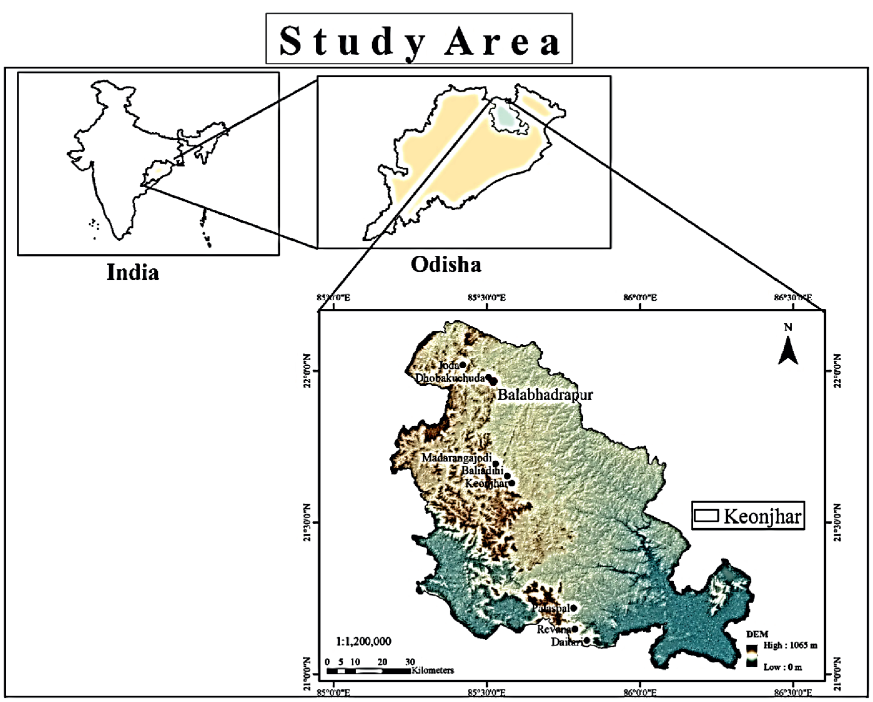

Fig. 1. Location map of pyrophyllite deposit in the Singhbhum Craton

studied and results are reported in this article. Six representative bulk samples of pyrophyllite showing different morphological habits were selected. For detailed characterization with respect to mineralogy, bulk chemistry and micro-chemistry, for this three different techniques such as optical and electron microscopy, XRD and XRF were employed. Thin polished section of six samples showing typical habits was prepared following conventional sample preparation technique and examined under Leica make high-resolution microscope(DM 2500 P) to find out the mineralogical and textural details. The mineral assemblage in these six types was also found out using a Rigaku ULTIMA IV(operating at 40kv acceleration voltage and 40mt current, having Cu target) XRD. The major and minor elements in these six morphologies were determined using PW100 Pan analytical XRF. Micro-morphological variations of pyrophyllite mineral and their in-situ compositional difference were documented using a FESEM-EDAX (ZEISS Gemini SEM 300) and AMETEK EDS system.

Results and Discussions

Morphological attributes

Based on the morphology, the pyrophyllite in the study area are grouped into six morph-types such as: Flaky, Laminated; Bladed; Fibrous; Platy; and Granular / Gritty (Fig. 2 A to F). Das

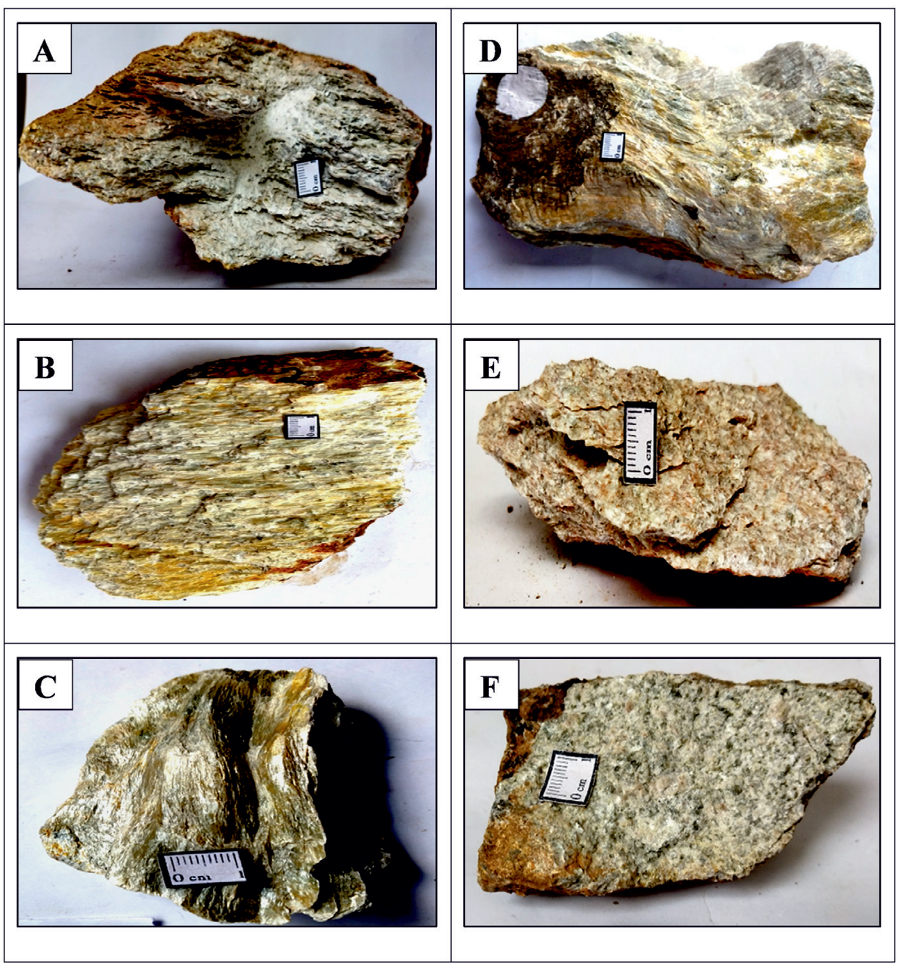

Fig. 2. Morphological variation of the Pyrophyllite schist in the study area
A-Flaky; B- Laminated; C- Bladed; D- Fibrous; E-Platy; F. Granular / Gritty

et al. (2012) described pyrophyllite from Madrangjodi, into three categories, viz, flaky, schistose and granulose, based on structure and texture. Later Yadav and Das (2017) identified three similar varieties in the Rathamara pyrophyllite deposit. Das and Mohanty (2009) described a slaty variety in addition to three varieties described by others, where they identified crude slaty cleavage in pyrophyllite.

Flaky type is coarse grained, and can split into individual flakes (Fig. 2A). Laminated types exhibit finely layered structure, akin to the pages of a book (Fig.2B). Bladed types are remarkable for their striking blade-like crystals, which often shows elongated, curvilinear shape reminiscent blades simulating kyanite (Fig.2C).

Fibrous variety consists of elongated, acicular-needle like crystals (Fig.2D). Platy type is marked by their flattened, sheet-like crystals (Fig.2E). Granular or gritty variety is recognized by its coarse-grained texture, consisting of individual grains or aggregates that impart a gritty feel upon touch (Fig.2F).

Mineral assemblage vs Morpho-types

Pyrophyllite is a soft and pale-colored lustrous schistose rock. Different minerals as interpreted from their XRD patterns, are pyrophyllite, quartz, mica, and feldspar. Other minor minerals such as hematite, rutile, and zircon are also recognized under microscope. A significant difference in the order of mineral assemblages is observed in these six morpho types (Table 1).

Besides the minerals described before, some additional minerals like chloritoid and tourmaline are reported from other pyrophyllite deposits (Das *et al.,* 2012; Pradhan *et al.,* 2014). Oner and Tas (2013) reported the presence of kaolinite (dickite), kyanite, chlorite, and alunite in addition to the major mineral phases in the Porturge region, Malatya, Eastern Turkey.

The flaky, laminated, and bladed types contain major pyrophyllite minerals while in other types, pyrophyllite appears as a subordinate phase.The fibrous, platy and granular varieties are dominated by muscovite, orthoclase, and quartz respectively (Table 1).

Morpho-types vs Composition

Six morpho-types of pyrophyllite were analysed for major and minor elements by XRF. The analytical results of all the six types and the host granite are presented in Table 2.

The chemical analysis data reveals that the pyrophyllite, in general, is constituted of silica and alumina as the major elements with sub-ordinate amount of potash. Other minor elements present are iron, titanium, and sodium. Vanadium and manganese are present in traces. The broad chemistry of pyrophyllite shows intermediate to high content of silica (49-69%); moderate alumina (21-34%) and subordinate potash content

Table 1. Mineralogical assemblages in different morph-types

Sample no.	Morpho-types	Minerals identified [In order of abundance]
A	Flaky	Pyro > Ortho > Mus
B	Laminated	Pyro > Mus > Qtz
D	Bladed	Pyro > Mus > Plag (albite)
C	Fibrous	Mus > Qtz > Pyro > Plag (albite)
E	Platy	Ortho > Qtz > Pyro
F	Granular / gritty	Qtz > Mus > Pyro

{Pyro- Pyrophyllite; Ortho- Orthoclase; Mus – Muscovite; Qtz- Quartz; Plag- Plagioclase}

Table 2. Major and selective minor element patterns (in wt%) in different morpho-types

Morpho types	Al_2O_3	SiO_2	K_2O	Fe_2O_3	TiO_2	Na_2O	V_2O_5	Mn	LOI
Flaky	31.73	56.35	6.79	0.24	0.42	0.12	0.004	0.005	4.31
Laminated	33.63	49.04	10.22	0.70	1.61	0.15	0.079	0.015	4.08
Bladed	33.92	53.02	6.89	0.38	0.81	0.14	0.006	0.009	4.69
Fibrous	27.97	58.41	4.74	0.52	0.34	0.12	0.005	0.007	6.72
Platy	21.11	69.26	6.04	0.17	0.47	0.10	0.009	0.007	2.79
Granular /Gritty	14.36	71.79	3.70	2.33	0.35	0.18	0.002	0.019	1.04
Granite	19.27	75.81	1.28	0.13	0.30	4.18	0.004	0.006	1.01

(4.7-10%). A pure pyrophyllite stoichiometrically contains 28.3% Al_2O_3, 66.7% SiO_2 and 5% H_2O.

As can be seen from the table (Table 2), and figure (Fig.3) the alumina content in flaky, laminated, bladed and fibrous types is higher than the stoichiometric data. Additional alumina content may be attributed to feldspar and mica found associated with these morpho types.

Micro-morphological attributes

The pyrophyllite mineral examined under scanning electron microscope. Showed varied types of habit/ form. These forms or micro morphologies are grouped into three categories depending upon their composition and further classified into six sub-types based on their shape.

Group I [Fig. 5.1]: 1. Acicular, radiating, 2. Anhedral; 3. Sub-globular; 4. Massive, 5. Monoclinic crystal; 6. Laminated, zig-saw.

Group-II [Fig. 5.2]: 1. Needle shaped, 2. Rod shaped, 3. Prismatic, 4. Platy, 5. Flaky, 6. Subhedral crystal

Group III [Fig. 5.3]: 1. Intergrown, 2. Porous, 3. Spotted, 4. Bedded 5. Ooloidal, 6. Spongy

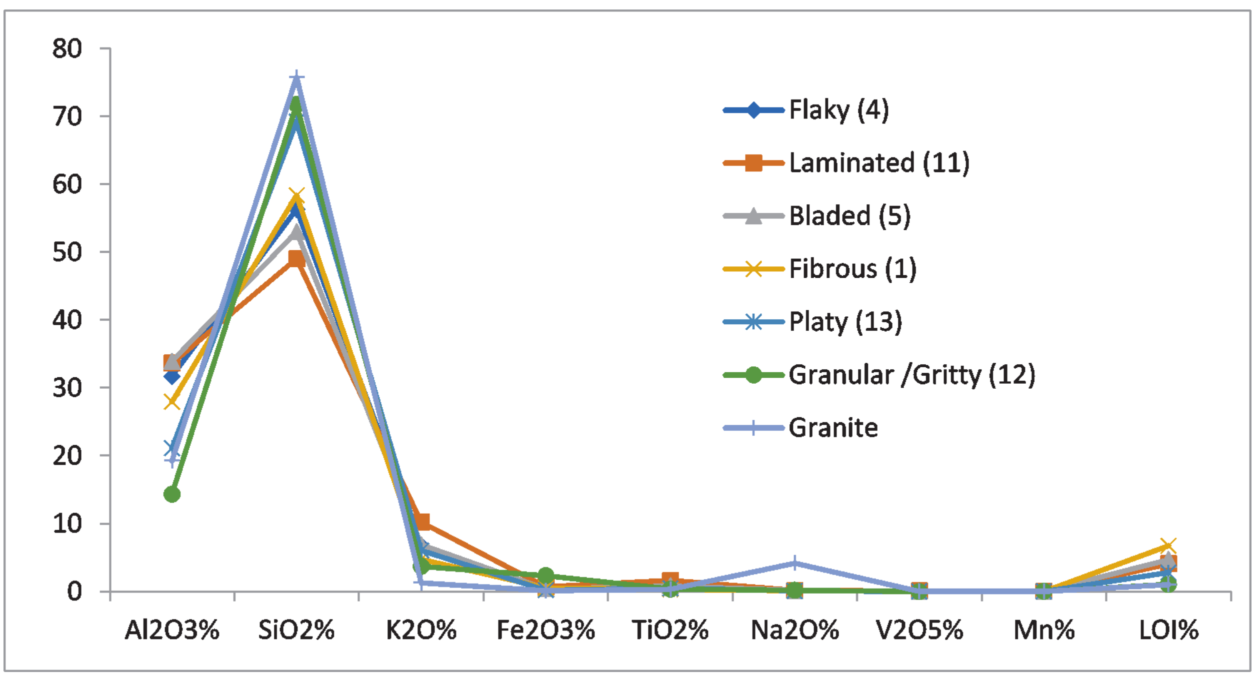

Fig. 3. Plot showing compositional difference between different morpho types and underlying host granite

Micro morpho-types vs Composition

The pyrophyllite reveals different types of habit (Fig. 4, 5, 6), which are found compositionally different when their chemistry is determined through in-situ EDS analysis (Fig. 4.1, 5.1, 6.1).

Based on the composition, they are broadly grouped into three categories as described above. The habits clubbed under group-I [Acicular radiating, Anhedral crystalline; Sub-globular; Massive, Monoclinic crystalline and Banded] are of pure pyrophyllite type (Fig. 4). Their EDS

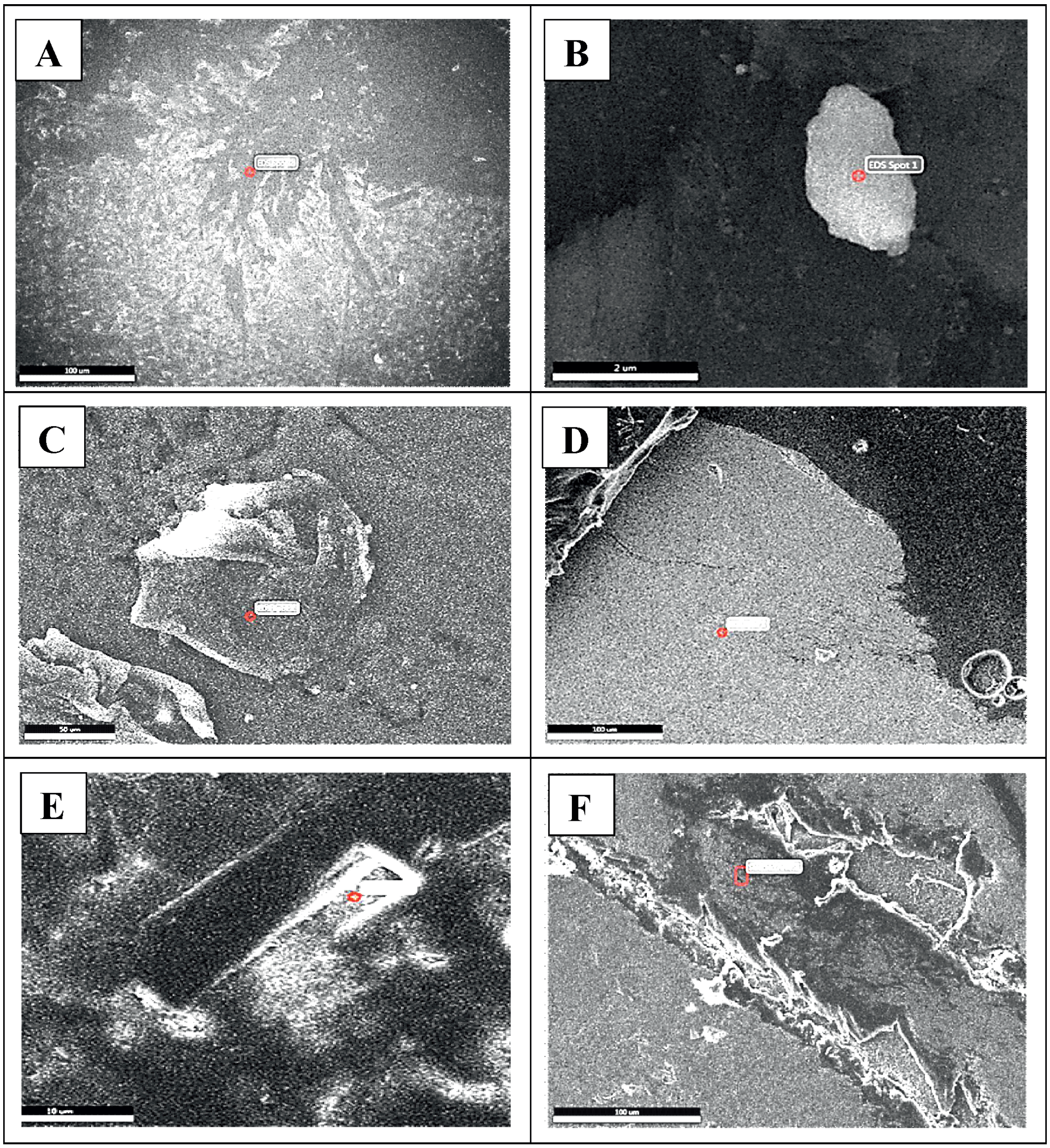

Fig. 4. Electron micrographs of pyrophyllite showing different habits
A. Acicular radiating; B. Anhedral crystalline; C. Sub-globular; D. Massive; E. Monoclinic crystalline; F. Banded
Note: Red markings are analyzed points

pattern and analytical results are presented in Fig. 4.1 and Table 3 respectively. In the group-II, the pyrophyllite containing 6.9 to 18% Fe as impurity exhibits- Needle shapes, Rods and Prismatic habit (Fig. 5.1 and Table 4). Similarly, the pyrophyllite containing 2 to 31% Mg, clubbed under this group, show Platy and Flaky shape. The platy type may be termed as low-magnesia

Table 3. In-situ compositional analysis results (in wt%) of pyrophyllite in different habits

Habit	O	Al	Si	Fe	Mg	K
A. Acicular radiating	63.1	16.9	20.0	-	-	-
B. Anhedral grain	58.5	20.1	21,4	-	-	-
C. Sub-globular	49.4	17.1	33.5	-	-	-
D. Massive	57.0	15.1	27.9	-	-	-
E. Monoclinic crystalline	55.2	13.1	31.8	-	-	-
F. Banded	60.9	13.5	25.6	-	-	-

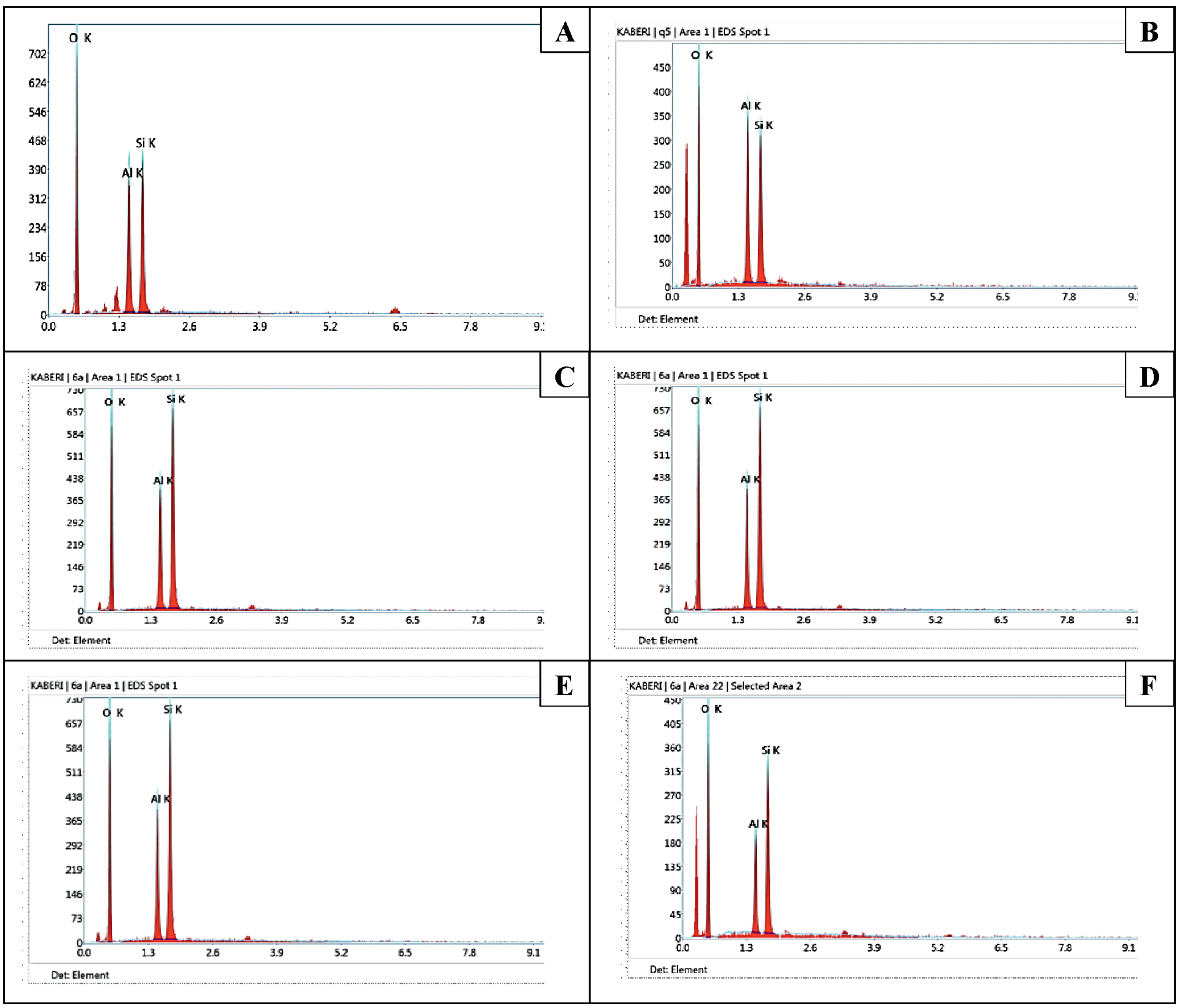

Fig. 4.1. Energy Dispersive Spectrum of Pyrophyllite grains / crystals shown in fig. 4

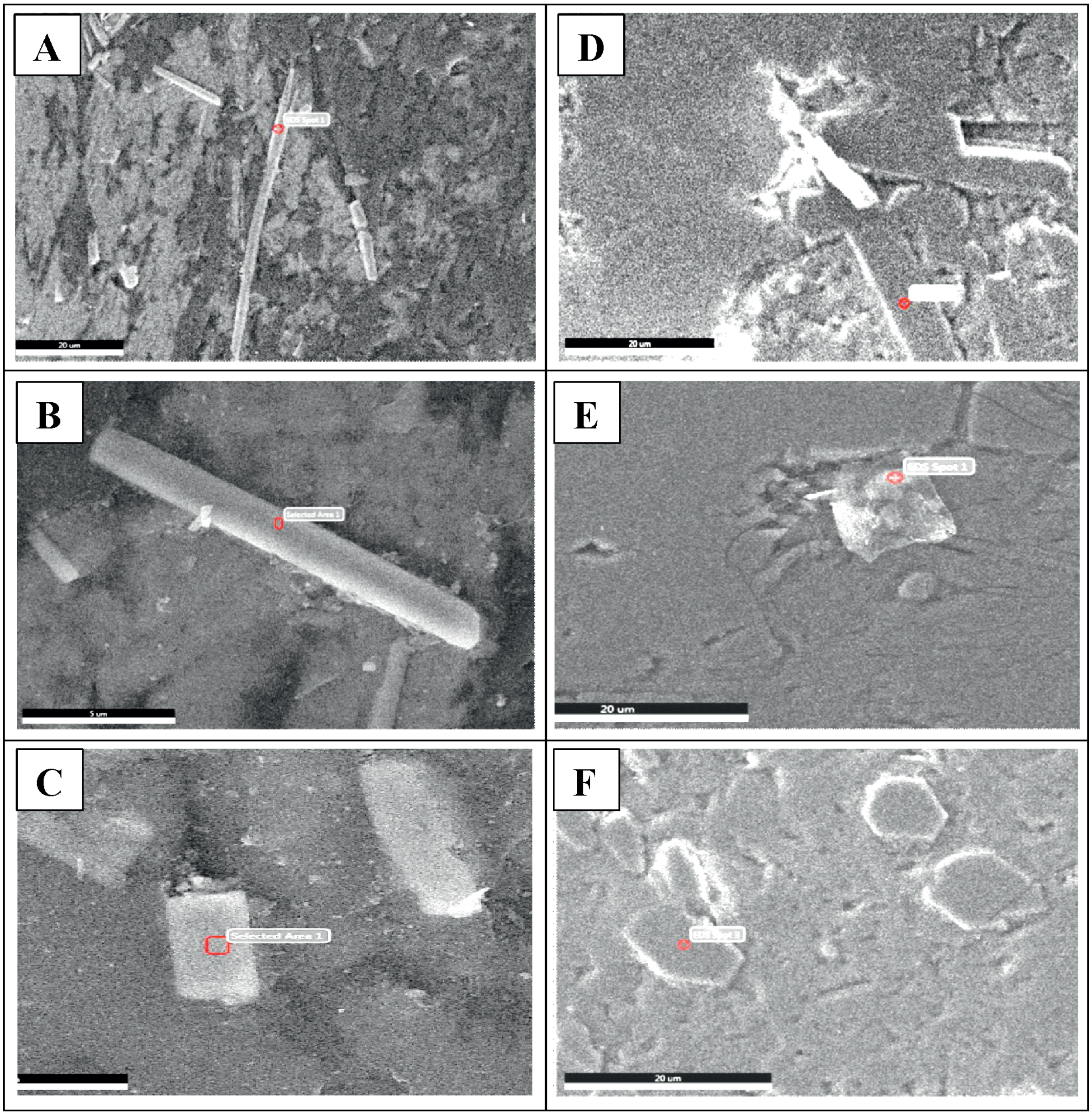

Fig. 5. Electron micrographs of pyrophyllite showing different habits
A. Needle shaped; B. Rod shaped; C. Prismatic, D. Platy; E. Flaky; F. Subhedral crystalline
Note: Red markings are analyzed points

Table 4. In-situ compositional analysis results (in wt%) of pyrophyllite in different habits

Habit	O	Al	Si	Fe	Mg	K
A. Needle shaped	57.0	18.3	16.2	18.0	-	-
B. Rod shaped	57.2	18.6	17.4	6.9	-	-
C. Prismatic	58.7	17.5	15.7	8.2	-	-
D. Platy	61.0	18.0	18.1	-	2.9	-
E. Flaky	51.2	9.3	8.6	-	31.0	-
F. Sub-hedral crystalline	58.6	18.7	17.2	3.5	2.0	-

pyrophyllite and Flaky type as high-magnesia pyrophyllite. Sometimes, the pyrophyllite appears as subhedral crystal, which have both Fe (3.5%) and Mg (2%) impurities. (Fig. 5.1, Table 4).

The pyrophyllite that constitutes of potash and soda as minor elements, besides alumina and silica shows a different form [4.6 to 8.1% K- Inter-grown, Porous, Spotted, Bedded & Ooloidal; 11.2 % Na Spongy], and included under group-III (Fig. 6.1 and Table 5).

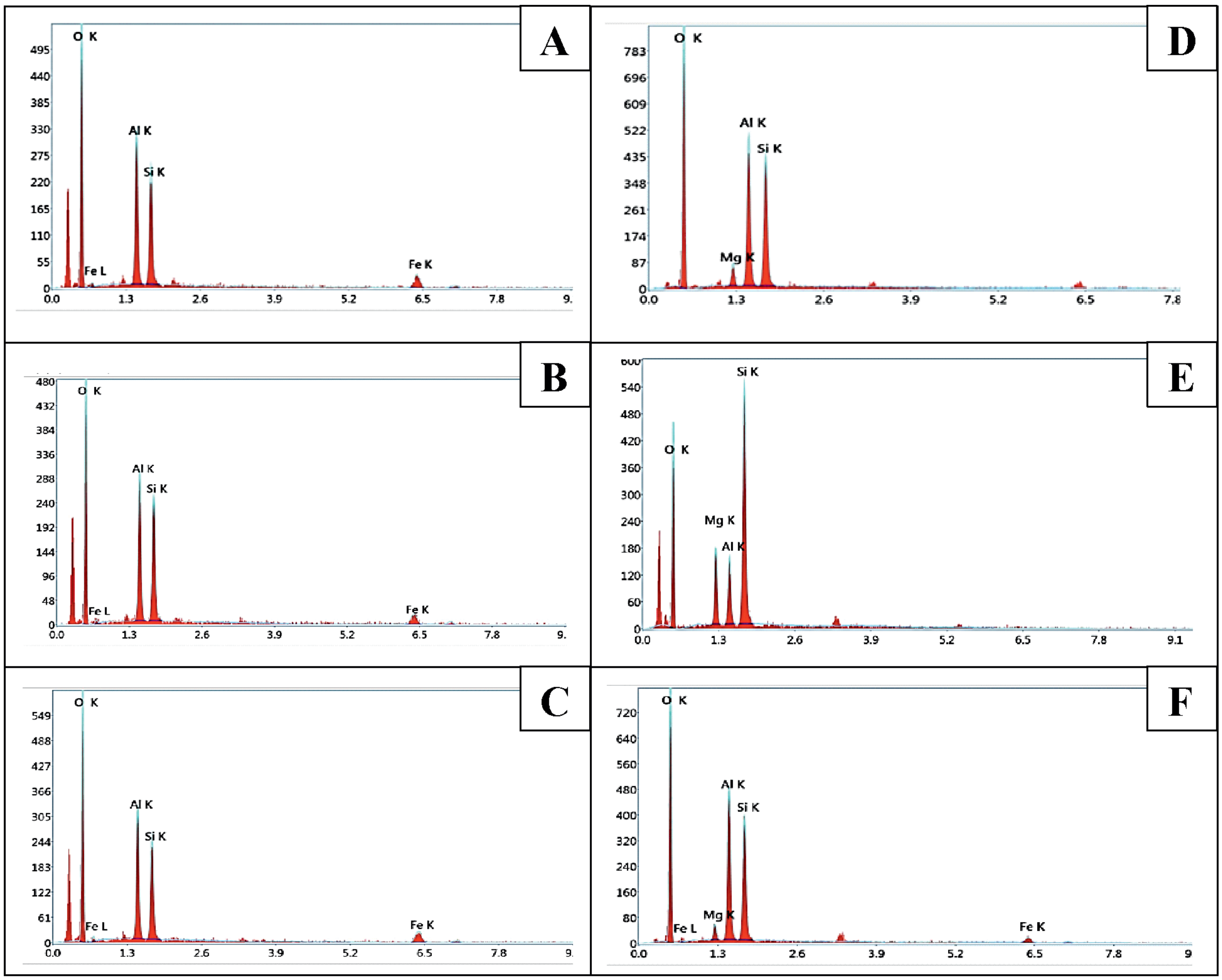

Fig. 5.1. Energy Dispersive Spectrum of Pyrophyllite grains / crystals shown in fig. 6

Table 5. In-situ compositional analysis results (in wt%) of pyrophyllite in different habits

Habit	O	Al	Si	Fe	Mg	K	Na
A. Intergrown	53.8	19.1	19.0	-	-	8.1	-
B. Porous	55.4	18.4	18.7	-	-	7.5	-
C. Spotted	55.2	17.4	20.3	-	-	7.1	-
D. Bedded	56.9	16.7	19.4	-	-	7.0	-
E. Ooloidal	56.0	10.6	28.8	-	-	4.6	-
F. Spongy	59.9	3.6	25.2	-	-	-	11.2

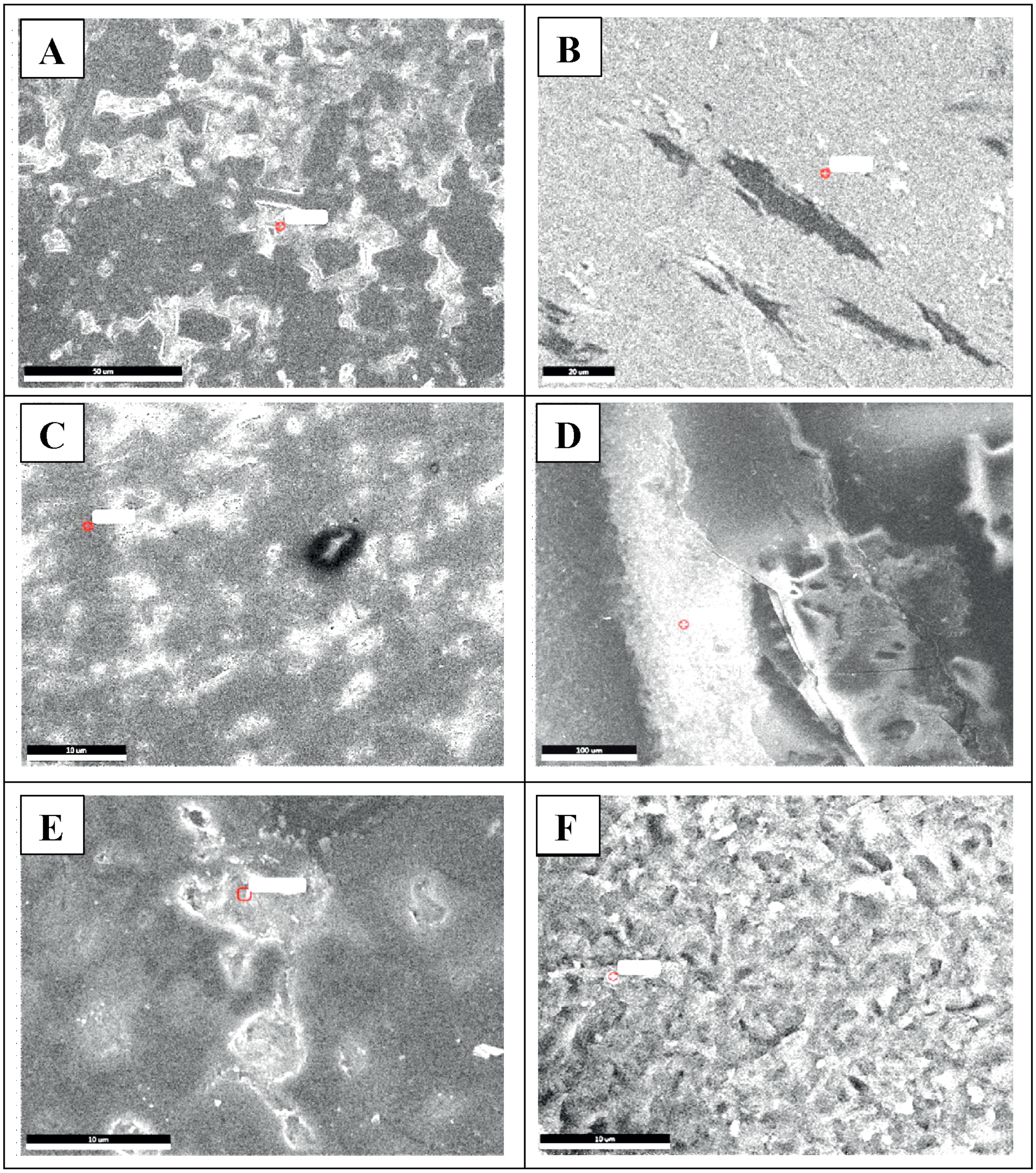

Fig. 6. Electron micrographs of pyrophyllite showing different habits
A. Inter-grown. B. Porous, C. Spotted, D. Bedded, E. Ooloidal, F. Spongy
Note: Red markings are analyzed points

Genetic Evolution of different Morpho types

The genetic evolution of pyrophyllite exhibiting different mega and micro-morphologies in the schistose rock from Keonjhar, Odisha, are interpreted from their in-situ compositional analyses. Presence of contaminants in the protolith probably played a major role in framing different habits of pyrophyllite. The chaste pyrophyllite might have formed through solution and precipitation without any contamination. The Fe-contaminated pyrophyllite is likely to have formed from a protolith having micaceous iron oxide. Both high

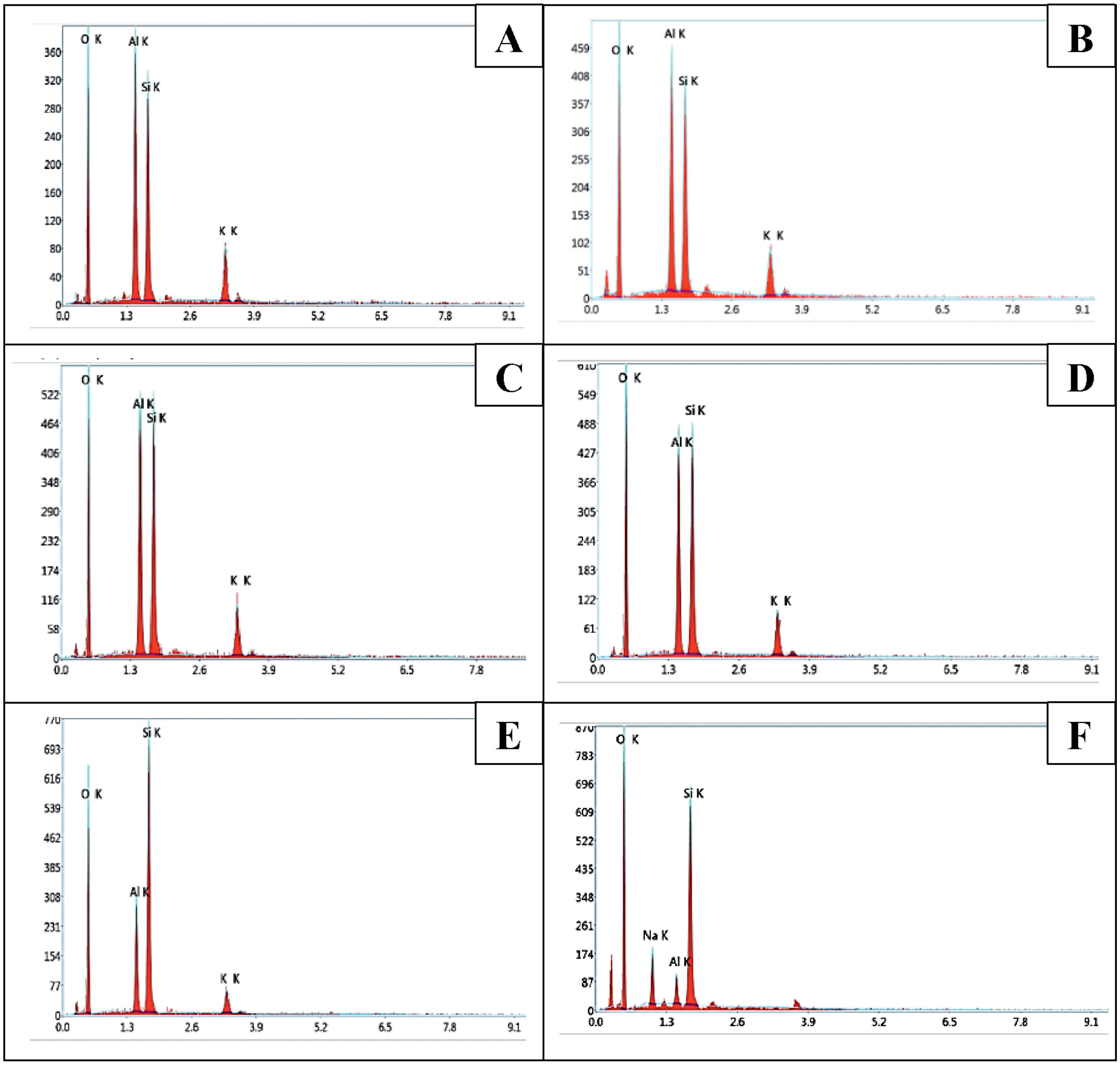

Fig. 6.1. Energy Dispersive Spectrum of Pyrophyllite grains / crystals shown in fig. 6

magnesia and low magnesia pyrophyllite are probably grown at variable expense of phlogopite mica. The pyrophyllite having both Fe and Mg impurities may be a derivative of biotite.

The potash containing pyrophyllite (4.6 to 8.1% K) is seemingly a substitute of orthoclase (14% potassium) / muscovite (9.77% potassium). Similarly, the pyrophyllite that contains Na (~11%) are possibly formed, replacing albite (Na-rich plagioclase). It may be mentioned here that the presence of both orthoclase and albite are recorded in the pyrophyllite schist from the study area (Table 1).

Conclusions

The pyrophyllite schist rock occurring over Singbhum granite Keonjhar district, Odisha shows six morpho types, viz. Flaky, Laminated; Bladed; Fibrous; Platy; and Granular / Gritty in its field disposition. The mineral assemblages in these morphologies are pyrophyllite, quartz, mica and feldspar (both orthoclase and plagioclase) in variable order of abundance. Various types of texture and micro-morphology of pyrophyllite mineral are observed under scanning electron microscope; The clean pyrophyllite, free from any contaminant, exhibits different habits like: Acicular radiating; Anhedral; Sub-globular; Massive, Monoclinic crystalline and Banded. Pyrophyllite having Fe-impurities shows needle shaped, rod shaped and prismatic form. The low magnesia and high magnesia pyrophyllite reveal platy and flaky habit respectively. The subhedral crystalline variety constitutes both Fe and Mg impurities. The potash containing pyrophyllite may appear in different shapes like inter-grown, porous, spotted, bedded, ooloidal, A spongy texture is exhibited by soda bearing pyrophyllite.

It may thus be surmised that the macro and micro-morphological variations revealed by pyrophyllite mineral are composition dependant. Based on the protolithic composition, the habit of the mineral can change.

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Conflict of Interest

The authors have no conflicts of interest with any individual or any organization.

Authors Contribution

Kaberi Sahoo: Sample preparation, analysis and manuscript writing.

Somnath Khaoash: Manuscript writing and technical editing.

Patitapaban Mishra: Analysis of samples and interpretation of analytical data.

Sujit Das: Field work and sample collection.

Birendra Kumar Mohapatra: Development of concept and technical editing of manuscript.

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