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QUARTERLY REPORT

NATURAL MICA STUDIES

(Covering period September 1, 1952, to November 30, 1952)

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GENERAL

During the past three months several phases of work on the natural micas have been completed and the following papers have been accepted for publication in the American Mineralogist:

1. Studies in the mica group; Mineralogy of the rose muscovites, by E. Wm. Heinrich and Alfred A. Levinson.
2. Studies in the mica group; Relationship between polymorphism and composition in the muscovite-lepidolite series, by Alfred A. Levinson.

Work has continued or is nearing completion on several projects discussed in the last quarterly report. These include structures of the high-silica muscovites (phengites) and of the biotites. Progress on the phengites was held up temporarily, as several critical specimens described in the literature were being sought from foreign investigators. Most of these have now been received, and it is planned to have the manuscript on structures of the phengites completed shortly.

Temporary difficulties also have been encountered in the study of the biotites. During the past quarter the x-ray machine did not operate for about three weeks because of difficulty in obtaining a specific micro-switch. Furthermore, it has been found impossible to distinguish the various polymorphs of biotite and phlogopite by the powder method. The photographs contain only a few of the strongest reflections (whose spacings appear to be identical for the various forms) with a very dark background. The possibility that this difficulty may be overcome by use of a monochromator is being investigated. Nevertheless, single-crystal work on the biotites has continued. It is hoped that this phase of the study will be completed by the middle of January.

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Two large projects have been started during this quarterly period. Mr. D. W. Levandowski has undertaken a study of the zinnwaldites, and Mr. C. H. Hewitt is studying zoning and overgrowths in micas. It is hoped that a complete paragenetic chemical and structural study of zinnwaldite will enable a definite decision to be made as to its relationship to other members of the mica group. With respect to the zoning and overgrowth study, since there are differences of opinion in the literature on the subject and since an excellent collection of specimens illustrating these relationships is available at the Mineralogical Laboratory, a detailed investigation seems desirable. Such a study should yield valuable information on progressive chemical and structural changes in the micas during the crystallization. lifetimes of their deposits. Both phases will probably be finished in time for the next quarterly report.

Through the courtesy of Dr. Robert Hatch, seven samples of lepidolites and muscovites are now being analyzed at the Electrotechnical Laboratory of the U. S. Bureau of Mines, Norris, Tennessee. These samples were carefully separated during the past quarter from specimens from the Brown Derby pegmatite, Gunnison County, Colorado. The paragenetic relationships and structure of these micas are well known. The problem involved is to determine the relationship between chemical variations, paragenetic position and polymorphism in micas from a single pegmatite. Similar studies are planned for other pegmatites that contain micas of several generations.

In the past two or three years several investigators have been attempting to explain polymorphism in such minerals as micas and SiC by means of the screw dislocation theory. A brief study of this approach has been inaugurated with the translation from French of an unpublished Belgian paper, obtained through the courtesy of Professor Ralph Grim of the University of Illinois.

Tentative plans have been formulated for the final report on this project. A detailed outline has been drafted, and it is expected that work on various sections of the final report may be initiated toward the end of the next quarter.

### SYSTEMATIZATION OF THE NATURAL MICAS

#### Introduction

A workable systematization of the micas has long been one of the aims of the project. Recently a preliminary classification based on structural and chemical data, along with an account of the very voluminous synonymy of the micas, has been completed. In the following pages this

classification is presented. It is however, subject to revision for the final report.

The numerous attempts at mica classification in the past have been based almost entirely on their chemistry and fall generally into two categories: (1) those employing complex quasistructural formulae (e.g., Clark, 1889; Jacob, 1925); and (2) those involving calculation of theoretical end-member molecules (e.g., Tschermak, 1878; Kunitz, 1924; Stevens, 1946). Berman (1937) approached the problem in more realistic fashion by correlating mica formulae with unit cell contents and expressed the chemical variation by means of atomic ratios of different elements occupying equivalent structural positions. However, the work of Hendricks and Jefferson (1939), Levinson (1953), and others has demonstrated the complex polymorphism in the mica group and the necessity for both chemical and x-ray studies of micas as a basis for a sound classification.

Because of the complex chemistry of the group, the numerous textural and other minor varieties, and repeated misapplication of names, the nomenclature and synonymy of the micas have been ponderous and involved. The purpose of this work is threefold: (1) to present a reasonable and usable subdivision of the micas into naturally occurring species and major varieties on the basis of both chemistry and structure; (2) to review the synonymy of the group and thus assign minor varietal names or duplications to the species or major variety to which they correspond; and (3) to list those micas that are of indeterminate status owing to incomplete studies.

In the annotated classification that follows we have used for chemical varieties the prefixes of Schaller (1930). Wherever possible for each entry we list: (1) a simplified formula based on 24 negative ions in the unit cell; (2) the major isomorphous substitutions; (3) the structure; (4) notes; and (5) synonymy.

For the structural varieties, the method of polymorphic notation suggested by Ramsdell (1947) is adopted. The symbols and their meanings used in this work are as follows:

1M	1-layer monoclinic structure
2M muscovite	2-layer monoclinic muscovite structure
2M	2-layer monoclinic octophyllite mica structure
3M	3-layer monoclinic structure
3R	3-layer rhombohedral structure
6M	6-layer monoclinic structure
6T	6-layer triclinic structure
24T	24-layer triclinic structure
48T	48-layer triclinic structure

Chemical-Structural Classification of the Micas

Species 1. Muscovite,  $K_2Al_4(Si_6Al_2)_{80}O_{20}(OH)_4$ . Minor Na, Rb, Cs, Ba, and Ca for K; minor Mg,  $Fe^{2+}$ ,  $Mn^{2+}$  and Li; Minor  $Fe^{3+}$ , Ti, and Cr for Al; minor F for OH; maximum Li O = 3.30%, occupying vacant octahedral positions. Structure: 2M muscovite.

Synonyms: adamsite, amnochrysolite, amphilogite, biaxial mica, cat gold, cat silver, common mica, damourite (in part), didymite, didymite, ferro-ferri-muscovite, frauenglas, helvetan, heptaphyllite, isinglass, kaliglimmer, marienglass, monrepite, nacrite, oblique mica, potash mica, schernikite, zweiaxger glimmer.

Hypothetical end-members: ferri-muscovite, kryptotile, leverrierite, lithium muscovite.

Varieties. a. Barian muscovite. Ba with reported maximum of 5.91% BaO (Doelter, 1914) for K. Structure: probably 2M muscovite, material labeled oellacherite from Tyrol has the 2M muscovite structure.

The barium-muscovite from Franklin Furnace, New Jersey, described by Bauer and Berman (1933), was examined by us by means of the x-ray powder method. The resultant pattern is not that of a muscovite, nor even that of a mica. The material is too fine-grained for detailed optical studies.

Synonyms: barium muscovite, oellacherite, sandbergerite.

b. Manganian-muscovite. Reported maximum MnO = 2.32% (Ellsworth, 1932); usually some  $Li_2O$  present. Structure: 2M muscovite.

The very fine-grained, deep purple mangan-muscovite of Eskola (1914) with 2.3% MnO has been checked by us by means of x-ray powder photographs, and the pattern does not correspond to any known mica structure. The photograph shows some quartz lines; thus a chemical analysis of purified material is necessary.

Synonyms: mangan-muscovite, manganese muscovite.

Ferrian muscovite.  $Fe^{3+}$  with maximum of 5.70%  $Fe_2O_3$  (Tschermak, 1878) reported, for Al. Structure: probably 2M

muscovite, for the "alurgite" from Cajon Pass (Webb, 1939) with 5.32%  $\text{Fe}_2\text{O}_3$  has been shown to have this structure.

d. Ferroan muscovite.  $\text{Fe}^{+2}$ , with maximum of 6.55%  $\text{FeO}$  (Wülfing, 1886) reported. Structure: probably 2M muscovite.

e. Chromian muscovite. Reported maximum of 4.81%  $\text{Cr}_2\text{O}_3$  (Whitmore, et al., 1946), for Al. Structure: 2M muscovite.

This follows the usage of Whitmore et al. (1946).

Synonyms: chromglimmer, chrome glimmer, fuchsite, gaebhardite, verdite.

f. Lithian muscovite.  $\text{K}_2(\text{Al}_4, \text{Li}) \text{ca } 5.0 (\text{Si}_{6-5}, \text{Al}_{2-3})_8 \text{O}_{20}(\text{OH}_3+\text{F})_4$ .  $\text{Li}_2\text{O}$ , at least 3.30% occupying vacant octahedral positions. Usually small amounts of F for OH. Structure: modified 2M muscovite (Levinson, 1953).

g. Phengite.  $\text{K}_2(\text{Mg}, \text{Fe}^{2+})\text{Al}_4(\text{Si}_7, \text{Al})_8\text{O}_{20}(\text{OH})_4$ . High-silica muscovite with considerable  $\text{MgO}$  (7.96% Pagliani, 1937) and in some cases  $\text{FeO}$ ; some  $\text{Fe}^3$  for Al. Structure: 2M muscovite.

The nomenclature of the silica-rich end of the muscovite series as proposed by Schaller (1950) is regarded by us as unsatisfactory because: (1) there is some evidence that the original leucophyllite (Starkl, 1883) is a mixture, and in any event a new analysis and an x-ray study of the type material are needed to check its validity; and (2) the term, alurgite, which Schaller (1950) suggests as a substitute for leucophyllite, in the event that the latter should prove untenable, also has been erroneously employed to indicate a normal, i.e., low-silica, ferrian muscovite (Webb, 1939; Ödman, 1950). X-ray studies, however, indicate that not all true alurgite is two-layer monoclinic in structure; some is three-layer rhombohedral. Thus the term alurgite has at various times been used for:

- (a) a red, high-silica muscovite with minor  $\text{Fe}^3$  and Mn. (Penfield, 1893);
- (b) a red ferrian low-silica muscovite (Webb, 1939 and Ödman, 1950);
- (c) a manganian high-silica muscovite (Winchell, 1951); and
- (d) a three-layer rhombohedral polymorph of (a), discovered by Hendricks and Jefferson (1939) and verified by us on material from St. Marcel, Italy.

Because of this confusion, the use of the term alurgite for the high-silica end-member of the muscovite series is also undesirable. Less confusion accompanies the term phengite, which has been generally employed to mean high-silica muscovite.

Hypothetical end-members: ferrophengite, picrophengite.

h. Rhombohedral phengite. (3R phengite). Differs from 2M phengite (lg) in having the three-layer rhombohedral structure (3R) and  $2V = 0^\circ$  - low. No well authenticated analysis of all-uniaxial material is available.

i. Chromian phengite. Cr, with maximum of 0.78%  $Cr_2O_3$  (Whitmore, et al., 1946), for Al. Structure: 2M muscovite. This follows the usage of Whitmore et al. (1946).

Synonyms: chromochre, mariposite.

j. Rhombohedral chromian phengite. (3R chromian phengite). Presumably chemically similar to Ii, but with the three-layer rhombohedral structure and  $2V = 0^\circ$  - low. No analysis of all uniaxial material available.

k. Sericite. Fine-grained muscovite. Structure: 2M muscovite.

The term sericite has been used for: :

- (a) fine-grained muscovite, either primary or secondary;
- (b) fine-grained phengite; and
- (c) hydromuscovite.

Thus it cannot be defined exactly on a chemical basis. It remains, however, highly useful as a general, nonspecific term for fine-grained muscovite whose exact chemical nature is unknown.

Synonyms: (including pinitic pseudomorphs). achlusite, agalmatolite, aspasiolite, avalite, bildstein, bonsdorffite, catalinite, cataspilite, cordierite-pinite, cymatolite, damourite (in part), dysyntribite, epileucite, epi-sericite, fahlunite, giesickite, gigantolite, gibertite, glimmer, gongylite, helvetan, hygrophyllite, iberite, ivigitite, killinite, lardite, lepidomorphite, liebenerite, lythodes, margarodite, micarel, micarelle, oncophyllite, oncosine, onkophyllit, onkosin, oosite, pagodite, parophite, pinite, pinitoid, polyargite, pyknophyllite, pyrargillite, pyrrholite, rosellan, rosellite,

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rosite, shilkinite, sterlingite, talcite, terenite, triclasite, wilsonite.

Species 2. Paragonite  $(\text{Na}, \text{K})_2\text{Al}_4(\text{Si}_6, \text{Al}_2)_8\text{O}_{20}(\text{OH})_4$ . Structure: 2M probably 2M muscovite. Schaller and Stevens (1941) have pointed out that the series muscovite-paragonite is not completely represented in nature. If intermediate types are discovered, it might be better to regard paragonite as sodian muscovite and reduce it to varietal status.

Synonyms: hallerite, natronglimmer, pregrattit, soda mica.

Species 3. Roscoelite.  $\text{K}_2(\text{V}, \text{Al})_4(\text{Si}_6\text{Al}_2)_8\text{O}_{20}(\text{OH})_4$  Maximum  $\text{V}_2\text{O}_5 = (\text{ca.}) 20\%$ . Structure: 1M.

Although roscoelite has previously been regarded as a vanidiferous muscovite, it deserves full species rank because it is structurally distinct (Quarterly Report No. 3).

Synonyms: colomite, vanadinglimmer, vanadium muscovite.

Species 4. Lepidolite.  $\text{K}_2(\text{Li}, \text{Al})_{5-6}(\text{Si}_{6-7}, \text{Al}_{2-1})_8\text{O}_{20-21}(\text{F}, \text{OH})_{3-4}$ . Rb and Cs replace K, in some types in considerable amounts ( $\text{Rb}_2\text{O} = 3.2\%$ ,  $\text{Cs}_2\text{O} = 1.90\%$ , Lundblad, 1942). Small amounts of Mn, Mg,  $\text{Fe}^{2+}$ ,  $\text{Fe}^{3+}$  are normally present. The OH:F ratio varies considerably, and OH may become negligible.

Synonyms: irvingite, lilalith, lilalite, Li-phengite, lithia mica, lithionglimmer, lithionit, lithionite, lithionitesilicat, macrolepidolite, microlepidotite, poly-irvingite, scale stone, siderischer-fels-glimmer.

Hypothetical end-members: paucilithionite, polyolithionite (in part), protolithionite.

Varieties. a. Six-layer monoclinic lepidolite. (6M lepidolite)  
 $\text{Li}_2\text{O} = (\text{ca.}) 4.0 - 5.1\%$ .

b. One-layer monoclinic lepidolite. (1M lepidolite)  
 $\text{Li}_2\text{O} = 5.1 - 7.26\%$ .

c. Three-layer rhombohedral lepidolite. (3R lepidolite)  
Composition approaches 4b. Due to twinning (?) (Levinson, 1953).  $2V = 0\text{-small}^\circ$ .



d. Three-layer monoclinic (3M lepidolite)  $\text{Li}_2\text{O} = 4.1\%$ .  
One example from Skuleboda, Sweden (Quarterly Report No. 2).

e. Manganian-lepidolite. Maximum reported  $\text{MnO} = 7.55\%$   
(Shibata, 1952). Structure: probably variable, depending on Li  
content.

f. Magnesian-lepidolite. (cited in Berman, 1937). We are  
unable to determine if any natural material of this composition  
has been discovered.

g. Polyolithionite. (in part).  $\text{K}_2\text{Li}_4\text{Al}_2\text{Si}_8\text{O}_{20}(\text{F},\text{OH})_4$ . A  
silicon-and lithium-rich, thus aluminum poor, lepidolite.  
Structure: 1M.

Species 5. Taeniolite.  $\text{K}_2\text{Mg}_4\text{Li}_2\text{Si}_8\text{O}_{20}\text{F}_4$ ; Structure: 1M.

Species 6. Zinnwaldite.  $\text{K}_2(\text{Fe}^{2+}_{1-2}\text{Li}_{2-3}\text{Al}_2)_6(\text{Si}_{6-7},\text{Al}_{2-1})_{20}$ .  
 $\text{F}_{3-2}, \text{OH}_{1-2})_4$ .

The (Al, Fe, Li) group may be considerably deficient. Zinnwaldites are chemically and structurally much more closely related to the biotites than to the lepidolites. No zinnwaldites have been found with the six-layer monoclinic structure so common in lepidolites. Also, the two-layer monoclinic octophyllite structure has been found in zinnwaldite, but not in lepidolite. In fact, there is no sharp natural compositional boundary between zinnwaldite and lithian biotite. If a demarcation is to be made, we believe the line can best be drawn on the amount of Li and suggest that lithium-iron micas with  $\text{Li} > 1$  atoms per unit cell formula be regarded as zinnwaldites, those with  $\text{Li} < 1$  be placed with the lithian biotites.

Synonyms: cryophyllite (in part), lithioneisenglimmer, polyolithionite (in part), protolithionite, rabenglimmer.

- Varieties.
- a. One-layer monoclinic zinnwaldite (1M zinnwaldite).
  - b. Two-layer monoclinic zinnwaldite (2M zinnwaldite).
  - c. Three-layer rhombohedral zinnwaldite (3R zinnwaldite).
  - d. Ferrian zinnwaldite maximum report  $\text{Fe}_2\text{O}_3 = 10.06\%$   
(Shibata, 1952).

Species 7. Phlogopite.  $K_2(Mg_6Fe^2)(Si_6Al_2)8O_{20}(OH)_4$ . Na can substitute for K up to nearly K: Na = 1:1 (Harada, 1936); minor Rb, Cs, Ba, and very minor Ca also may proxy for K.  $Fe^2$  is almost always present, but Mg predominates greatly over  $Fe^2$ . Small amounts of Mn,  $Fe^3$  and Ti may be present. The Si: tetrahedral Al ratio may be larger than 6:2.

There is no well-defined, natural, compositional boundary between ferroan phlogopite and magnesian biotite. Because  $Fe^2$  is a strong chromophore, micas of this type even with only a small per cent of Fe are dark colored and are thus commonly classed as biotites. If a division is required, we suggest that where the ratio of  $Mg:Fe^2 > 4:2$ , the mineral should be classed as phlogopite.

Synonyms: aspidolite barium-phlogopite, barytbiotite, hydrophlogopite, magnesia mica, natronophlogopite, octophyllite, pholidolite, rhombic mica.

Hypothetical end-members: fluor-phlogopite, hydroxyl-phlogopite.

Varieties a. One-layer monoclinic phlogopite (1M phlogopite). The most common type.

b. Two-layer monoclinic phlogopite (2M phlogopite).

c. Three-layer rhombohedral phlogopite (3R phlogopite).

d. Manganophyllite.  $K_2(Mg_{5-4}, Mn_{2-1}, Fe^2_{0-0.5})_6 Fe^3_{0-1} Mn^3_{0-1} (Si_6Al_2)8O_{20}$ . Structure: Generally 1M; some approach a three layer rhombohedral structure. 2M reported by Hendricks and Jefferson (1939).

Although some investigators (e.g., Hey, 1950) class manganophyllites as varieties of biotites, most manganophyllites have little or no  $Fe^2$  and only small amounts of  $Fe^3$ . An exception is a Langban, Sweden, mica analyzed by Jakob (No. 8, p. 157, 1925) which contains 16.94%  $Fe_2O_3$ . Apparently Mn is present commonly as  $Mn^3$ , rarely as  $Mn^2$ . Jakob, (1925).

Synonyms: Manganese mica, manganophyll.

e. Titanian phlogopite.  $K_2Mg_5Ti(Si_6Al_2)O_{22}(OH,F)_2$  (Prider 1939)  $Fe^2$  and  $Fe^3$  are minor. Structure: 1M

Species 8. Biotite  $K_2(Fe^{2+}, Mg)_{6-4}(Fe^{3+}, Al, Ti)_{0-2}(Si_{6-5}, Al_{2-3})_{80}O_{20-22}(OH, F)_{4-2}$ : Some Na, Ca, Ba, Rb, and Cs for K; Mn for  $Fe^{2+}$ ; F for OH; Mg may be absent. Total F + OH may be very low (Walker and Parsons, 1926).

Synonyms: annite, anomite, caesium-biotite, chromglimmer (in part), euchlorite, eukamptite, ferromuscovite, haughtonite,, heterophyllite, hexagonal mica, iron mica, lepidomelane, meroxene, natronbiotite, octophyllite, odenite, odinite, odite, oderite, pterolite, rhombenglimmer, rubellan, siderophyllite, titanglimmer, titanmica, uniaxial mica, waddoite.

Hypothetical end-members: cryophyllite (in part), eastonite, fluor-annite, fluor-biotite, fluor-lepidomelane, fluor-meroxene, fluor-siderophyllite, hydroxyl-annite, hydroxyl-biotite, hydroxyl-lepidomelane, hydroxyl-meroxene, hydroxyl-siderophyllite, manganophyllite (in part).

Varieties a. One-layer monoclinic biotite (1M biotite) The most common type.

b. Two-layer monoclinic biotite (2M biotite).

c. Three-layer rhombohedral biotite (3R biotite).

d. Six-layer triclinic biotite (6T biotite).

e. Twenty-four-layer triclinic biotite (24T biotite).

f. Forty-eight-layer triclinic biotite (48T biotite).

S. A. Forman, Bureau of Mines, Ottawa, Canada, indicates that he has found a forty-eight-layer triclinic form (personal communication).

g. Calcian biotite. A biotite from Kaiserstuhl, Germany, has 8.17% CaO (Daub, 1913). Structure unknown.

The validity of calcian biotite as a major variety is doubtful. The existence of Ca in the biotite structure has been challenged by Jacob (1929). Several specimens labelled calicobiotite, from Italian localities, have been found to have the 1M structure.

h. Ferroan biotite. Mg is very minor or absent, Fe is present mainly as  $Fe^{2+}$ .

Synonyms: siderophyllite, lepidomelane (in part).

i. Manganian-biotite. Mn as much as 1 atom per unit cell formula. Fe present as  $Fe^{2+}$  or  $Fe^{3+}$ .

j. Ferrian biotite.  $K_2(Fe^{2+},Mg)_{3-4}Fe^{3+}_{2-3}(Si_6,Al,Ti)_{8O_{20-21}}$

Synonyms: ferribiotite, lepidomelane (in part).

Lepidomelane is commonly employed for iron-rich biotite, but the term has been used to embrace biotites rich in  $Fe^{3+}$ , those rich in  $Fe^{2+}$  and those with relatively large amounts of both  $Fe^{2+}$  and  $Fe^{3+}$  (Heinrich 1946).

k. Lithian biotite.  $K_2(Fe^{2+},Mg)_5(Li,Al,Fe^{3+})_1(Si_7,Al)_8O_{20}(OH,F)_4$ .

l. Titanian biotite.  $K_2(Fe_2,Mg_3)_5Ti(Si_6,Al_2)_8O_{22}(OH,F)_2$ .

Synonyms: ferrititanbiotite, ferriwotanite, titanbiotite, titanobiotite, wodanite, wotanite.

Both titanian biotite and titanium phlogopites are relatively poor in  $Fe^{3+}$  and also are very low in OH and F.

### Hydrous Micas

Because most investigators class these minerals with the clay group, their crystal chemistry is not considered here. However, it is interesting to note that a new interpretation (Brown and Norrish, 1952) of the chemistry of one of the species in this group, hydromuscovite, postulates the replacement of K by oxonium (hydronium) ions  $H_3O^+$ . Species and varietal names included in this group are: bastonite, brammallite, bravaisite, buldymite, damourite (in part), goeschwitzite, grundite, gumbelite, hydrobiotite, hydro-mica, illite, metasericite, Mg-illidromica, rastolyte, sarospatakite, sarospatite, sericite (in part) sodium-illite, voigtite.

### Micas of Indeterminate Status

1. Euphyllite - near  $(Na,K)Al_3Si_3O_{10}(OH)_2$ ; may be a mica intermediate between muscovite and paragonite; or brittle mica or perhaps a mixture.

2. Mahadevite - near  $(K,Na)_{0.97}(Al,Fe,Mg)_{2.66}(Si,Al)_4(O,OH)_{12}$ ; supposedly between muscovite and phlogopite in composition.

3. Manandonite - a borosilicate of Li and Al closest to lepidolite in composition;  $\text{Li}_4\text{Al}_{14}\text{B}_4\text{Si}_6\text{O}_{29}(\text{OH})_{24}(\text{?})$ -possibly not a mica.

4. Leucophyllite - a high silica phengite - perhaps a mixture.

5. Anthrophyllite - a "mica (?)" (Hey 1950, p. 283).

Other Varietal Names

Anhydromuscovite

Baddeckite

Anhydrobiotite

Bauerite

Metabiotite

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