CONTRIBUTIONS TO THE JAMESONITE PROBLEM

By S. KOCH, GY. GRASSELLY

Institute for Mineralogy and Petrography,
University of Szeged, Hungary

and

K. PADĚRA

Institute for Mineralogy, Geochemistry and Crystallography of the Charles University of Prague, Czechoslovakia

Jamesonite is not a rare mineral. In spite of this there are questions to be elucidated concerning its composition, formula, ore microscopical properties as well as its relation to plumosite. Similarly the question of parajamesonite of Kisbánya (Herja) must also be cleared. Our task is to answer these questions.

INTRODUCTION

Among the lead-antimony sulphides jamesonite is relatively the most frequent mineral. In some localities e. g. at Zlatá-Ida (Aranyida, Czechoslovakia) it was the dominating ore of the veins. In spite of this, this interesting mineral is not well known.

Its formula was until recently assumed to be $Pb_2Sb_2S_5$ *[Eskola 1949, Köhler, 1949]*, although on the basis of analyses already in 1910 the formula $Pb_4FeSb_6S_{14}$ was proposed by *Schaller*. To day this is the generally accepted formula of jamesonite. At the same time in his work issued in 1953, *Machatschki* proposed the formula $Pb_4(SbFe)_7S_{14}$, whereas *Kostov* in his book published in 1957 uses the formula $Pb_4Sb_6S_{13}$.

The use of the name plumosite is not quite satisfactory and unequivocal. Usually the fine threaded cottonwool-feltlike jamesonite, occasionally boulangerite is denoted by this name, but according to *Ramdohr* and *Betechtin* the plumosite or »Federerz« is extremely fine threaded iron-free jamesonite. Among the occurrences of this iron-free plumosite Baia Sprie (Felsőbánya, Rumánia) is mentioned by Ramdohr.

Having personally collected material from the best jamesonite occurrences within the Carpathians I and my coworkers began to examine this ore.

The chemical examinations were carried out by *Gy. Grasselly*, the roentgenographical ones by *K. Paděra*.

RESULTS OF THE MINERALOGICAL AND CHEMICAL EXAMINATIONS

Within the Carpathians the jamesonite appears genetically in two different ways. At Zlatá-Ida epithermal silver-bearing jamesonite veins connected to Variscian granites are known. The youngest sulphide ore of the vein is the dominating jamesonite.

The jamesonite in mineralized veins connected to Tertiary subvolcanic rocks can be found in the following places:

in mesothermal veins in Nagybörzsöny (Hungary) and Herja (Rumania), among the minerals of the mesothermal metasomatic ore body of Rodna (Rumania),

in epithermal silver-bearing zinc-lead veins of Gyöngyösoroszi (Hungary), Baňská Štiavnica (Selmecbánya, Czechoslovakia), Keresztheg near Baia Mare (Nagybánya, Rumania), Baia Sprie (Felsőbánya, Rumania), Capnic (Kapnikbánya, Rumania),

in epithermal gold-silver veins of Valea Borcutului (Borpatak, Rumania), Săcărâmb (Nagyág, Rumania).

In this localities jamesonite occurs in economically insignificant quantities mainly as very fine-threaded plumosite.

Zlatá Ida (Aranyida, Czecho-Slovakia).

Jamesonite is the dominating ore of the epithermal veins genetically connected to the Variscian granite. The dark grey ore with silky metal lustre is a dense interwoven texture of thinner or thicker jamesonite threads reaching several cm in length. The ore replaces older idiomorphic pyrite occurring in far smaller amounts further sphalerite rich in chalcopyrite inclusions as well as freibergite and idiomorphic arsenopyrite. (Photos 1, 2, 3). Galena could not even be found in traces.

Among the older sulphide ores in sphalerite and freibergite gold grains with a diameter of 20—30 μ could be observed. Thus the slight gold content of the jamesonite originates from the older ores replaced by it. A part of its silver content certainly is derived from freibergite.

The quartz and siderite present in considerable quantities in the veins are mostly older than jamesonite, however, younger siderite replacing jamesonite could also be observed in polished ore sections.

In polished ore sections on the columnar-fibrous crystals of jamesonite sometimes a dense striation running parallel to the c crytallographical axis showing a fine threaded structure may be observed. Its reflection pleochroism is not striking. The characteristic greenish tint is well visible. Beside jamesonite the freibergite is darker, its colour is brownish with a mildly pinkish shade. In oil immersion the difference in colour is more intensive. Arsenopyrite is lighter and has a white colour with a pronounced yellowish tint.

Under crossed nicols the anisotropy effects are strong, the colours (yellow — greenish yellow — slate blue) depend upon the orientation, however, they are less intensive than the colour shown by antimonite and berthierite. As is mentioned by *Ramdohr* the extinction is parallel

Fig. 1. Jamesonite (white) replacing sphalerite (grey), pyrite (gray with rough surface), freibergite (light grey) and arsenopyrite (white with strong relief). Plain light, x50.

Fig. 2. Jamesonite replacing freibergite (grey). In the freibergite arsenopyrite as inclusion. Plain light, x50.

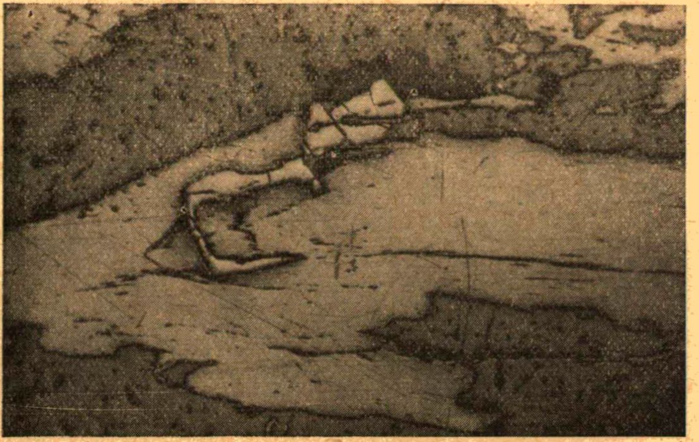

Fig. 3. Coarsely threaded jamesonite replacing arsenopyrite and sphalerite (grey). Plain light, x50.

but it will not be totally extinguished, the darkest colour is dark brownish grey. Among the wide jamesonite threads twin crystals can only rarely be observed. In contrast to *Murdoch*'s opinion it could be stated that besides ccHNO_3 also ccHCl attacks the jamesonite ore sections under development of H_2S. A dull slightly brownish grey patch forms and the crystal disintegrates into masses of fine threads running parallel to the c crystallographical axis. (Photo 4). Under crossed nicols the intensity of colours decreases considerably, the etched part shows a silky lustre.

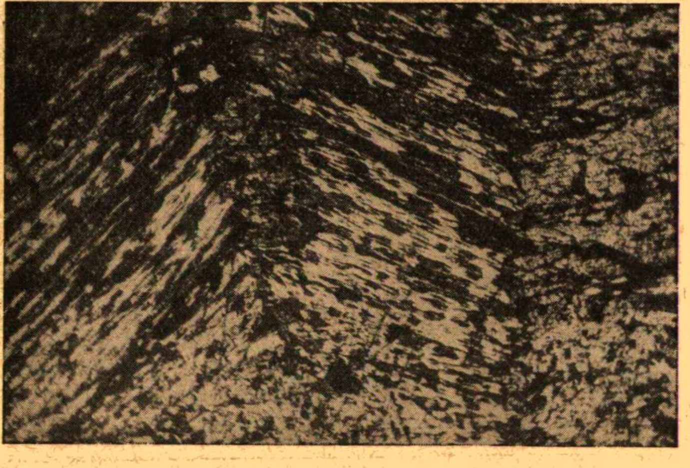

Fig. 4. Translation-twin lamellae of jamesonite developed by etching with HCl cc. Plain light, x100.

The columnar crystal aggregate built up from a mass of fine threads running parallel to the *c* crystallographical axis undergoes a translation under a tectonical load (Photo 5). Columns not etched often show a spindle-like translation phenomenon. The crystals and granules of the older, more rigid, ores are surrounded and replaced by jamesonite. The

Fig. 5. Translation phenomenon in jamesonite. Crossed nicols, x50.

jamesonite crystal aggregates intruding into small cavities, filled up subsequently by quartz or siderite, are branched. It can be well observed that the branched jamesonite fibres have mostly no terminal ends, they disintegrate into finer threads. Sometimes, however, the crystals are covered by curved platelets of a prism of fourth order. Of the jamesonite

Fig. 6. Twins of jamesonite with terminal planes embedded in quartz. x300.

crystals embedded in quartz twin crystals according to 100 can also be observed (Photo 6). The result of chemical analysis of jamesonite from Zlatá Ida is the following:

	1
Sb	27,95 %
Pb	31,69
Fe	2,13
Cu	0,08
Zn	13,30
S	23,80
SiO_2	1,10
	100,05 %

(Analyst: Mrs. E. Klivényi)

Samples for investigation were available from mesothermal pyrrhotite-sphalerite-galena veins connected to pyroxen-andesite, of Herja and mesothermal metasomatic pyrrhotite-sphalerite-galena-pyrite veins, connected to andesite, of Rodna. In its Tertiary subvolcanic occurrences the jamesonite occurs everywhere in two different ways: partly as thin covering layer consisting of dense aggregate of crystal-needles at the border of galena crystal groups replacing the galena, partly as extremely fine threaded loose plumosite forming cottonwool-like aggregates. The occurrence is economically insignificant everywhere.

Herja (Kisbánya, Rumania)

At Herja the jamesonite occurred very plentifully at the upper levels of the Salán vein. The galena-replacing jamesonite intruding frostwork-like into the galena is very frequent. (Photos 7, 8, 9). The replacement takes place starting from the boundaries of the coarsely crystallized galena aggregates of the cavity filling consisting in bulk of pyrrhotite-

Fig. 7. Jamesonite replacing frostwork-like galena. Crossed nicols, x50.

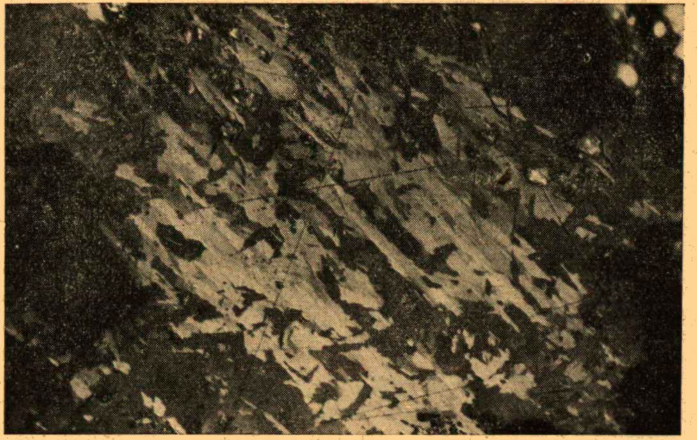

Fig. 8. Twinned jamesonite replacing frostwork-like galena. x100.

Fig. 9. Jamesonite replacing galena. Crossed nicols, x100.

sphalerite (14,69 per cent Fe). Beside the jamesonite, semseyite can be often found as sulphosalt, replacing galena. The tabular crystals or fan-like crystal aggregates of semseyite are replaced by the younger jamesonite (Photo 10). Jamesonite replacing semseyite intrudes as thin columnar crystals into it. The columnar crystals are often twins (Photo 11).

Fig. 10. Fine threaded jamesonite replacing tabular semseyite crystals. Crossed nicols, x100.

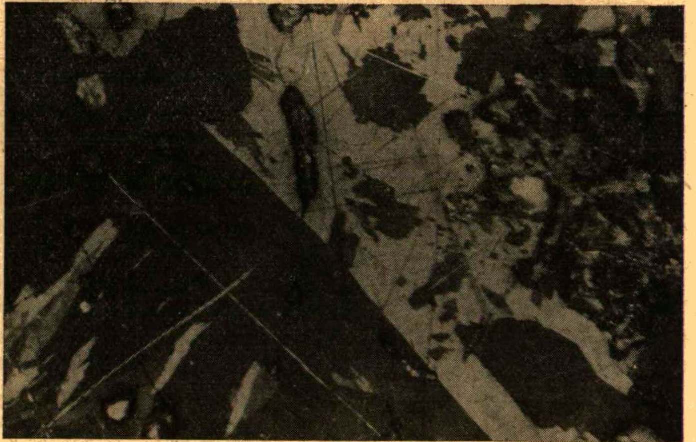

Fig. 11. Twin lamellated jamesonite replacing semseyite. Crossed nicols, x50.

The jamesonite layer consisting of dense aggregate of thin columnar crystals may be several mm in thickness. If the crystal aggregates of this layer intrude into the small cavities of the ore, they become extremely fine threaded feltlike mass. This feltlike plumosite covers the a few mm long, flat columnar jamesonite crystals of corroded surface, overgrown on the surface of the dense jamesonite coating (analysis 2).

These crystals — like the jamesonite forming their basis — are built up from frostwork-like interwoven crystal aggregates, thus they are no monocrystals. The occurrence and habit of these crystals agree with those of the parajamesonite crystals investigated by *Zsivny* and *Náray-Szabó*. The result of the chemical analysis of the dark grey plumosite coating covering these crystals and galena can be seen in analysis 3.

In the hollows of the ores of the Salán vein — particularly on level IV — the cottonwool-like soft texture consisting of interwoven aggre-

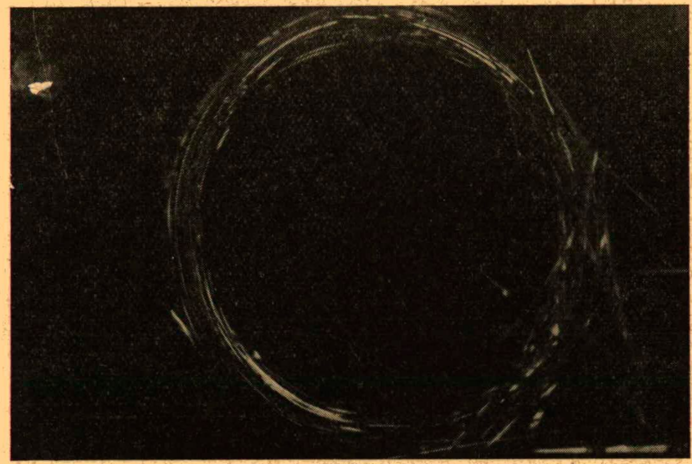

Fig. 12. Strongly curved bundles of jamesonite (plumosite) threads. x50.

gates of very fine needles of plumosite can often be found. From some hollows handfuls of fine threaded plumosite can be picked out. The threads are often curved, some threads and crystal aggregates built up from these threads even often form a circle (Photo 12). Overgrown on fine plumosite threads extremely minute ideally developed hexahedrons of pyrite, pointing to low formation temperature, can often be found. These pyrite crystals, which owing to their tinyness cannot be removed, cause the strikingly high iron content in the results of analyses of jamesonite specimens.

The semseyite, jamesonite as well as the relatively rare fizelyite formed at the cost of galena. The surface of the crystal aggregates of galena from Herja is very often corroded, the edges of the crystals are rounded and the surface of the crystallized specimens is often corroded, »parquetlike«. In the loose cottonwool-like mass of plumosite intergrown one can find the short columnar crystals of quartz, the $-\frac{1}{2}R$ rombohedrons of calcite attaining 1 cm in size as well as the globular crystal aggregates of siderite and dolomite stained grey or black by mass of plumosite inclusions (Photo 13). In the relatively rare crystals of gypsum and vivianite, plumosite also occurs plentifully as inclusions.

The composition of the loose cottonwool-like plumosite is given in analysis 4.

Fig. 13. Jamesonite (plumosite) inclusions in black calcite. Plain light, x100.

Analyses of jamesonite (plumosite) specimens from Herja:

	2	3	4
Pb	38,18 %	35,92 %	37,65 %
Sb	34,78	31,72	35,45
Fe	4,38	7,57	5,22
S	22,15	23,20	21,84
SiO_2	0,28	1,61	0,23
	99,77 %	100,02 %	100,39 %

Rodna (Óradna, Rumania)

In the lodes containing galena in Rodna the jamesonite occured everywhere. Here also the thin coating formed by frostwork-like crystal aggregates of jamesonite replacing galena as well as the plumosite masses composing loose cottonwool-like aggregates in ore hollows may be found. The plumosite occurs particularly in large amounts in the bedrock of the »Nándor« pyrite stock. In the hollows of the stock composed of pyrite—black sphalerite (14,27 per cent Fe), less galena and chalcopyrite, the cottonwool-like plumosite occurred frequently. Here also like at Herja the galena everywhere shows traces of dissolution, its crystals are rounded.

The dolomite crystals containing plumosite inclusions are stained grey and black by this mineral.

The result of the analysis of the fine threaded cottonwool-like plumosite from Rodna:

	5
Pb	37,52 %
Sb	33,76
Fe	4,94
S	21,98
SiO_2	1,71
	99,91 %

Baia Mare (Nagybánya, Rumania), Valea Borcutului (Borpatak, Rumania), Baia Sprie (Felsőbánya, Rumania)

The mode of the occurrence of the jamesonite at Baia Mare, in the mine of Keresztehegy, at Valea Borcutului and Baia Sprie is identical with the occurrence of Herja. In these localities too the jamesonite (plumosite) filling up the cavities as extremely fine threaded cottonwool-like aggregates as well as the galena replacing frostwork-like crystal aggregates can be found.

The mass of the plumosite inclusions stains black the calcite crystals at Baia Mare, whereas at Valea Borcutului and Baia Sprie beautiful baryte crystals and crystal groups can be found stained partly or completely black.

The well developed andorite crystals occurred at Baia Sprie in mine »Gyulaköze« in 1925 were embedded in plumosite.

The result of the analysis of fine threaded plumosite from Baia Sprie carried out earlier by *J. Loczka* is as follows:

	6
Pb	39,38 %
Sb	35,80
Fe	2,65
Mn	0,12
Zn	0,10
S	21,59
SiO_2	0,50
	100,14 %

Taking into account the analysis reported in the present paper as well as a few earlier results, the question was dealt with which of the formulas mentioned in the introduction can be accepted as the most probable formula for jamesonite.

In Table 1 the data of the analyses reported as well as those of the analyses according to *Loczka* and *Náray-Szabó — Zsivny — Zombory* are summarized, recalculating to 100 per cent the Pb, Sb, Fe, S content of the samples after subtracting in a suitable form the Cu, Zn, Mn, SiO_2 content present as impurities.

In the Table 1 also the formulas are denoted which are calculable from these data.

Table 1.

Specimen and No. of the analysis	Locality	Composition re-calculated to 100 per cent		Mol. quotient		Formel
Jamesonite (1)	Zlatá Ida	Pb	40,17 %	0,1939	4,15	4PbS. FeS. $3Sb_2S_3$
		Sb	35,43	0,2910	6,23	
		Fe	2,61	0,0467	1	
		S	21,79	0,6797	14,55	
»Parajamesonite« (2)	Herja	Pb	38,38	0,1852	2,35	2PbS. FeS. $2Sb_2S_3$
		Sb	34,96	0,2871	3,64	
		Fe	4,40	0,0788	1	
		S	22,26	0,6943	8,81	
Plumosite (3)	Herja	Pb	36,51	0,1762	1,27	4PbS. 3FeS. $3Sb_2S_3$
		Sb	32,23	0,2647	1,92	x3
		Fe	7,69	0,1377	1	
		S	23,57	0,7352	5,33	
Plumosite (4)	Herja	Pb	37,59	0,1814	1,94	4PbS. 2FeS. $3Sb_2S_3$
		Sb	35,40	0,2907	3,11	x2
		Fe	5,21	0,0933	1	
		S	21,80	0,6800	7,28	
Plumosite (5)	Rodna	Pb	38,21	0,1844	2,04	4PbS. 2FeS. $3Sb_2S_3$
		Sb	34,38	0,2824	3,13	x2
		Fe	5,03	0,0901	1	
		S	22,38	0,6981	7,75	
Plumosite (6)	Baia Sprie	Pb	39,67	0,1914	4,02	4PbS. FeS. $3Sb_2S_3$
		Sb	36,06	0,2962	6,23	
		Fe	2,65	0,0475	1	
		S	21,62	0,6744	14,19	
»Parajamesonite«	Herja	Pb	40,01	0,1931	3,59	7PbS. 2FeS. $5Sb_2S_3$
		Sb	34,92	0,2868	5,34	x2
		Fe	3,00	0,0537	1	
		S	22,07	0,6884	12,81	

The formulas which can be calculated from the above data are not unequivocal, especially not in the case of the two parajamesonite specimens, whereas in that of the other samples besides the PbS/Sb_2S_3 ratio of constant character, the FeS content fluctuates. It is to be noted that on the polished ore sections prepared from the samples analyzed (analysis 2) besides the jamesonite and parajamesonite also another lead-antimony sulphosalt (semseyite) can also be found, thus the fluctuation in the formula is probably due to the presence of other similar sulphosalts.

The formulas were also calculated on the basis of earlier jamesonite analyses considering the iron content as belonging to the jamesonite molecule. Hence on the basis of the data of analyses XXIV, XXXIV, XXXVII from Hintze's Handbuch der Mineralogie (Bd. I. 1. p. 1032) as well as on that of the analyses 5., and 7., from Dana's System of Mineralogy (Vol. I. p. 453), the following formulas for jamesonite can be calculated:

$$4PbS.\ 2FeS.\ 3Sb_2S_3$$
$$2PbS.\ FeS.\ Sb_2S_3$$
$$4PbS.\ FeS.\ 3Sb_2S_3$$
$$4PbS.\ FeS.\ 3Sb_2S_3$$
$$3PbS.\ FeS.\ 2Sb_2S_3$$

Thus the formula of the jamesonite varies also on the basis of the data of earlier analyses — especially changes the iron content — like the formula calculated from the recent data.

At the same time, however, if the iron content is not considered to belong to the jamesonite and only the Pb, Sb, S content is recalculated to 100 per cent after subtracting the iron content and the corresponding S content with the other impurities, in the case of all samples the formula of $4PbS.\ 3Sb_2S_3$ is obtained, corresponding to the formula $Pb_4Sb_6S_{13}$ given by *Kostov*.

As, however, we have never found iron-free jamesonite samples it may be assumed that the formula $4PbS.\ FeS.\ 3Sb_2S_3$ suggested already also by *Schaller* and confirmed by other authors can be considered as the formula of jamesonite. It should be remarked — and this explains the more or less fluctuating iron content — that a part of iron indeed often does not belong to the jamesonite molecule but it is present as impurity. This is also confirmed by the fact that on the fine threads of numerous plumosite specimens overgrown pyrite crystals in microscopical size could be observed and the sample to be analyzed could not be separated from them. In the case of such finely threaded substances as plumosite an entanglement with impurities, which can only difficultly or alltogether not be removed, must be taken into account.

THE RESULTS OF THE X-RAY EXAMINATIONS

The possible identity of parajamesonite with jamesonite

Zsivny and *I. Náray-Szabó* [1947] described from the locality of Herja (Kisbánya, Rumania) a new mineral which they called parajamesonite. Jamesonite and parajamesonite having the same chemical formula ought only to differ markedly by the powder roentgenogram. We were therefore very surprised to observe that the x-ray diagrams of the samples from the original locality of Herja (Kisbánya) were identical with those of the jamesonite.

At the x-ray investigations two samples of parajamesonite from Kisbánya (Herja) were available. The Debye-Scherrer method was applied. The conditions of the exposure were as follows: Cu anticathode, Ni filter, chamber diameter 57,4 mm.

The intensities of the interference lines were estimated in a scale of ten degrees.

Five diffraction patterns were made which always showed the identity of the dominant component of the specimens with jamesonite. To eliminate the possiblity of the modification-change due to the grinding of the samples to be investigated, three exposures were made from the needlelike crystals of parajamesonite with a normal metallic lustre, ground during various periods. The different grinding time i. e. not quite a minute, about 10 minutes and more than an hour, did not influence the d-values and the intensities of the interference lines. However, the exposure of the parajamesonite powder not ground for quite a minute furnished two different interference lines. The stronger interferences showed uninterrupted arches and belong to jamesonite. The weaker interference lines were composed of single divided dots. They often coincided with the preceeding interference lines and than their presence was revealed by the appearance of the single dots on the normal diffusion arch. Their d-values in Å units 3,10—1,91—1,64—1,245—1,112——1,046 : 0,958 and 0,918 correspond to the strong interferences of sphalerite.

The presence of some of the strongest lines of sphalerite could also be observed at the diffraction patterns of parajamesonite powder ground during a longer period. In this case, however, it was not possible to distinguish the interferences of sphalerite from the coinciding lines of jamesonite, as the latter were far more intensive against the interference lines of sphalerite. The identification of the subordinate component i. e. of sphalerite in the samples investigated rendered possible only by the diffraction pattern of the powder ground for a short time. The fourth x-ray diffraction pattern is made from the black powder with a non metallic lustre which coats the not well developed needlelike, crystals of parajamesonite. The evaluation of the diffraction pattern and the comparison with the results obtained by *L. G. Berry* [1940] for jamesonite are comprised in Table 2. They proved that the black powder is identical with jamesonite.

Table 2.

Jamesonite, Kisbánya (Herja) Cu/Ni			Jamesonite according to L. G. Berry	
No of the lines	I	d		d
1	4	4,04 Å	s	4,03 kX
2	6	3,77	s	3,76
3	1	3,69		
4	2	3,55		
5	10	3,40	vvs	3,42
6	2	3,30		
7			vvw	3,20
8	3	3,13 }	s	3,11
9	5	3,08 }		
10	3	2,94	w	2,95

Jamesonite, Kisbánya (Heria) Cu/Ni			Jamesonite according to L. G. Berry	
No. of the lines	I	d	I	d
11	9	2,81	s	2,81
12	8	2,71	vs	2,72
13	2	2,62		
14	3	2,35	vw	2,37
15	5	2,29	s	2,31
16	5	2,24	s	2,25
17	3	2,18	vvw	2,17
18	3	2,10		
19	6	2,04	vs	2,05
20	7	2,02	vs	2,04
21	2	1,95	vw	1,967
22	4	1,90	m	1,905
23	3	1,87	w	1,882
24	7	1,83	s	1,826
25	1	1,78		
26	2	1,76	vvw	1,760
27	4	1,71	m	1,717
28	1	1,65		
29	2	1,61	vvw	1,616
30	2	1,57	vvw	1,581
31	3	1,52	m	1,528
32	1	1,480		
33	4	1,449	vw	1,455
34	3	1,414	vw	1,415
35	2	1,382		
36	3	1,369	m	1,361
37			vw	1,347
38	2	1,313		
39	2	1,287	w	1,283
40	2	1,261		
41	2	1,234		
42	3	1,212	m	1,208
43	1	1,184		
44	4d	1,165	m	1,169
45	5d	1,118		
46	2	1,082		
47	1	1,064		
48	1	1,048		
49	1	0,993		
50	2	0,976		

The diffraction pattern of parajamesonite obtained by *V. Zsivny* and *I. Náray-Szabó* [1947] differs completely from our results obtained for jamesonite. The recalculated *d*-values of the results as well as the intensity of the single interferences concerning the parajamesonite obtained by the authors mentioned above are illustrated in Table 3.

Table 3.

Parajamesonite
Kisbánya (Herja)

No of the lines	I	d
1	3	4,66 Å
2	8—9	4,21
3	5	3,78
4	1	3,29
5	1	3,00
6	1	2,73
7	1	2,65
8	2	2,49
9	2	2,23
10	2	2,02
11	1	1,92
12	1	1,84

The fifth x-ray pattern was obtained from a second specimen of parajamesonite. The substance to be investigated was separated from the needle-like incompletely developed crystals with a metallic lustre.

The roentgenographical identification proved that the specimens from the Herja locality or at least most of them, considered in the collections as parajamesonites, are actually jamesonites. Whether or not parajamesonite is identical with jamesonite or if truly an independent mineral is involved, can only be decided by further investigation of parajamesonite specimens from the original locality. The rareness and inaccessibility of the further specimens of parajamesonite rendered it impossible for the authors to solve the problem decisively.

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