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WinHolclas, a Windows program for hollandite-supergroup minerals

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ABSTRACT

A Microsoft® Visual Basic software, WinHolclas, has been developed to calculate the chemical formulas of hollandite-supergroup minerals based on data obtained from wet-chemical and electron-microprobe analyses. WinHolclas currently evaluates 12 valid mineral species using the Commission on New Minerals, Nomenclature and Classification (CNMMC) of the International Mineralogical Association (IMA) nomenclature scheme for the hollandite-supergroup minerals, with the simplified general formula $(A^{+}, A^{2+})(M^{4+}, M^{3+})_8O_{16}$ in the coronadite and priderite groups. The program recalculates and estimates the chemical formulas of hollandite-supergroup species based on 16 oxygen atoms, with the option of octahedral site total of 8.00 atoms per formula unit normalization. Mineral compositions of the hollandite-supergroup are calculated based on the dominant tetravalent sixfold-coordinated cation, considering the dominant tunnel cation (A^{+}, A^{2+}) and the dominant charge-compensating octahedral cation (M^{3+}, M^{2+}). WinHolclas operates in four stages: (1) it estimates cation and anion contents provided by input chemical data; (2) it determines the dominant tunnel and charge-compensating cation in each group; (3) it assigns the hollandite-supergroup minerals to one of the two groups; and (4) it classifies the hollandite-supergroup species into an appropriate groups such as coronadite and priderite. WinHolclas allows users to: (1) enter up to 42 input variables for mineral-chemical analyses; (2) type and load multiple hollandite-supergroup mineral compositions in the data entry section; (3) edit and load the Microsoft® Excel files used in calculating, classifying, and naming the hollandite-supergroup minerals, and (4) store all the calculated parameters in the output of a Microsoft® Excel file for further data evaluations. The program is distributed as a self-extracting setup file, including the necessary support files used by the program, a help file, and representative sample data files.

Keywords: hollandite supergroup; coronadite group; priderite group; nomenclature; classification; software.

INTRODUCTION

The hollandite-supergroup minerals are structurally characterized by octahedral walls (2x2 octahedra wide) cross-linked to each other to build up a tunnel structure that is large enough to host monovalent alkali and divalent alkaline-earth cations, as well as, in some cases, water molecules (Pasero, 2005; Capalbo, 2013; Biagioni et al. 2013). The general chemical formula of hollandite-supergroup minerals can be expressed as either $A^{2+}[M^{4+}{}_6M^{3+}{}_2]O_{16}$, $A^{2+}[M^{4+}{}_7M^{2+}]O_{16}$, $A^{+}[M^{4+}{}_7M^{3+}]O_{16}$ or $A^{+}[M^{4+}{}_{7.5}M^{2+}{}_{0.5}]O_{16}$ based on the nature of the tunnel as well as the charge-compensating cation, where A^{2+}=Pb^{2+}, Ba^{2+}, Sr^{2+}; A^{+}=K^{+}, Na^{+}, Tl^{+}; M^{4+}=Mn^{4+}, Ti^{4+}; M^{3+}=Mn^{+3}, Fe^{+3}, Cr^{+3}, V^{3+}; M^{2+}=Fe^{2+}, Cu^{2+} (Biagioni et al., 2013). The hollandite-supergroup minerals can be classified into two groups based on their chemical composition:

the coronadite group, which has manganese ($^{VI}M^{4+}$= Mn) as the dominant tetravalent sixfold-coordinated cation (DTSCC), and the priderite group, which features titanium ($^{VI}M^{4+}$=Ti) as the dominant cation. The naming of these minerals, commonly known as tunnel oxides, is based on the primary tunnel cation (A^{+} or A^{2+}) and the main charge-compensating octahedral cation (typically M^{3+}, and occasionally M^{2+}). The Commission on New Minerals, Nomenclature and Classification (CNMMN) of the International Mineralogical Association (IMA) has approved several new minerals within the supergroup based on the principle that each unique combination of the dominant tunnel cation (referred to as DTC) and the dominant charge-compensating cation (referred to as DCCC) corresponds to a distinct mineral species, following the concept of "valence-imposed double site occupancy" (Hatert and Burke, 2008). Considering that the previous databases and lists of mineral species did not provide a clear identification of the DTC and DCCC, as well as a clear definition lacking for the endmember formula of the hollandite-supergroup minerals, Biagioni et al. (2013) provided a consistent nomenclature for potentially new species that can be identified based on the general crystal-chemical rules. In the current nomenclature scheme, each combination of dominant A^{+}, A^{2+} (i.e., DTC), M^{4+}, $M^{2+}$and M^{3+} (i.e., DCCC) cations corresponds to a distinct mineral species in the hollandite-supergroup minerals (Capalbo, 2013; Chukanov et al., 2016).

Although various computer programs applicable to the calculation and classification of rock-forming silicates, ore-related minerals, as well as accessory minerals, have been developed over the past two decades (e.g., Yavuz, 1999, 2003, 2013; Yavuz et al., 2014, 2015; Yavuz and Yıldırım, 2018, 2020; Yavuz, 2021; Yavuz and Yavuz, 2022, 2023 a,b, 2024; Yavuz, 2024 a,b; Janoušek et al., 2024; Yavuz, 2025), those useful for the hollandite-supergroup minerals, according to the current IMA report, have not yet appeared in the literature. Taking this situation into consideration, a computer program called WinHolclas has been developed using the Microsoft® Visual Basic programming language. It can be used to calculate the chemical formulas from up to 200 analyses obtained from both wet-chemical and electron-microprobe techniques. The program calculates the compositional formulas of hollandite-supergroup minerals on the basis of 16 oxygen atoms (apfu). However, WinHolclas allows users to select the M-site total of 8.00 (apfu) normalization option from the pull-down menu of Calculate Contents of Ions in the Start-up Screen or Data Entry Screen for the compositional formulas of hollandite-supergroup minerals. The calculation and classification procedures applied to minerals by the program are carried out based on the currently accepted IMA nomenclature scheme by Biagioni et al. (2013). Additionally, it takes into account new species (e.g., ferricoronadite and thalliomelane) that post-date the IMA report. WinHolclas allows the user to display members of the hollandite supergroup in several binary and ternary classification and compositional diagrams by using Golden Software's Grapher program.

HOLLANDITE-SUPERGROUP MINERALS NOMENCLATURE

Among oxide minerals, the subgroup known as "tunnel oxides" is particularly significant in the context of microporous materials (Capalbo, 2013). The hollandite supergroup comprises various manganese (Mn) and titanium (Ti) oxides in the coronadite and priderite group. Both groups exhibit a tunnel structure that is formed by a framework of cross linked octahedral walls (Tang et al., 2016). Manganese oxides within the coronadite group of the hollandite-supergroup minerals occur abundantly in a wide variety of geological settings. These minerals, along with their synthetic counterparts, have been extensively investigated for their potential uses in radioactive waste storage, specifically for the immobilization of radioactive cesium, ionic conduction, supercapacitors, battery electrodes, catalysts, and improving pollution control for heavy metals in water contaminated by mining and other industrial activities (Pasero, 2005; Post et al., 2022). On the other hand, certain synthetic analogues of hollandite-supergroup minerals are recognized for their exceptional charge-discharge capabilities, along with their antiferromagnetic and semiconductor properties (Chukanov et al., 2016). Additionally, research indicates that at high pressures, feldspar minerals can change into a hollandite-like structure and this transformation may be significant for understanding the structure of the lower crust and upper mantle conditions (Post, 1999).

Using the available data on the hollandite supergroup species, Biagioni et al. (2013) proposed a nomenclature and classification scheme for the members of the hollandite-supergroup minerals, which has been approved by the IMA-CNMNC (see Table 1). Although some samples show monoclinic distortion (pseudo-tetragonal) space groups, with a≈c≈10.0 Å, and b≈90°, such as I2/m (e.g., cryptomelane) or $P2_1/n$ (e.g., strontiomelane), the ideal topological symmetry of hollandite-supergroup minerals within 2x2 tunnel oxides is rutile-like and tetragonal, space group I4/m (a≈10.0 Å and c≈2.9 Å), which is displayed, for example, in priderite (Biagioni et al., 2013; Chukanov et al., 2016). The hollandite-supergroup minerals are divided into two groups, including the coronadite group ($M=Mn^{4+}$) and the priderite group ($M=Ti^{4+}$), based on the dominant M cation within the octahedra. In each group, mineral species are categorized according to the combination of the DTC, either A^{2+} or A^{+}, and the DCCC,

Table 1. A list of the IMA-approved species in the hollandite-supergroup minerals (revised from Biagioni et al., 2013).

Row	Species	Symbol	Formula	DTSCC	DTC	DCCC	IMA status
	Coronadite Group (M^{4+}=Mn)						
1	Coronadite	Cor	$Pb(Mn^{4+}_6Mn^{3+}_2)O_{16}$	Mn^{4+}	Pb^{2+}	Mn^{3+}	A
2	Ferricoronadite	Fcor	$Pb(Mn^{4+}_6Fe^{3+}_2)O_{16}$	Mn^{4+}	Pb^{2+}	Fe^{3+}	A
3	Ferrihollandite	Fhol	$Ba(Mn^{4+}_6Fe^{3+}_2)O_{16}$	Mn^{4+}	Ba^{2+}	Fe^{3+}	A
4	Hollandite	Hol	$Ba(Mn^{4+}_6Mn^{3+}_2)O_{16}$	Mn^{4+}	Ba^{2+}	Mn^{3+}	A
5	Cryptomelane	Cml	$K(Mn^{4+}_7Mn^{3+})O_{16}$	Mn^{4+}	K^{+}	Mn^{3+}	A
6	Manjiroite	Mji	$Na(Mn^{4+}_7Mn^{3+})O_{16}$	Mn^{4+}	Na^{+}	Mn^{3+}	A
7	Strontiomelane	Sml	$Sr(Mn^{4+}_6Mn^{3+}_2)O_{16}$	Mn^{4+}	Sr^{2+}	Mn^{3+}	A
8	Thalliomelane	Tml	$Tl(Mn^{4+}_{7.5}Cu^{2+}_{0.5})O_{16}$	Mn^{4+}	Tl^{+}	Cu^{2+}	A
	Priderite Group (M^{4+} = Ti)						
9	Priderite	Pdr	$K(Ti^{4+}_7Fe^{3+})O_{16}$	Ti^{4+}	K^{+}	Fe^{3+}	A
10	Redledgeite	Red	$Ba(Ti^{4+}_6Cr^{3+}_2)O_{16}$	Ti^{4+}	Ba^{2+}	Cr^{3+}	A
11	Mannardite	Man	$Ba(Ti^{4+}_6V^{3+}_2)O_{16}$	Ti^{4+}	Ba^{2+}	V^{3+}	A
12	Henrymeyerite	Hmy	$Ba(Ti^{4+}_7Fe^{2+})O_{16}$	Ti^{4+}	Ba^{2+}	Fe^{2+}	A

DTSCC= Dominant tetravalent sixfold-coordinated cation, DTC= Dominant tunnel cation, DCCC= Dominant charge-compensating cation; A= Approved by the IMA.

which includes M^{3+} or M^{2+} cations that partially replace M^{4+} in the octahedra to maintain charge balance (Biagioni et al., 2013).

Manganese is the 10th most abundant element in the Earth's crust and exists in three oxidation states (Mn^{2+}, Mn^{3+}, Mn^{4+}) within minerals. This leads to the formation of over 30 different oxide and hydroxide minerals (Niu et al., 2021). To recalculate the ideal chemical formulas of the hollandite-supergroup minerals from electron-microprobe analyses, and in the absence of direct evidence regarding the valence states of manganese or iron through spectroscopic methods, Biagioni et al. (2013) recommend formalizing the approach used for several minerals in the supergroup that involves estimating the analytical data based on a total of 8 octahedral cations and 16 oxygen atoms per formula unit (apfu). Typically, when all manganese is represented as MnO_2 and all iron as Fe_2O_3, eight octahedral cations should yield over 32 positive charges and the charge balance can be achieved by distributing the total manganese content between Mn^{4+} and Mn^{3+}. In contrast, the priderite group, usually Mn-free compared to the coronadite group, maintains charge balance by partitioning the total iron content between Fe^{3+} and Fe^{2+} (Biagioni et al., 2013). According to Biagioni et al. (2013), the ideal crystal-chemical formula for minerals within the hollandite supergroup is influenced by the valence states of the tunnel and charge-compensating cations. Consequently, the formula of hollandite-supergroup minerals can be represented in one of the following formats:

i) if the main tunnel cation has a charge of 2+: $A^{2+}[M^{4+}_6M^{3+}_2]O_{16}$ or rarely $A^{2+}[M^{4+}_7M^{2+}]O_{16}$;

ii) if the main tunnel cation has a charge of 1+: $A^{+}[M^{4+}_7M^{3+}]O_{16}$ or rarely $A^{+}[M^{4+}_{7.5}M^{2+}_{0.5}]O_{16}$.

The water content in the minerals of the hollandite supergroup remains a subject of discussion. For example, water molecules in tunnel oxides are not essential, as their presence or absence does not distinguish between different species. Officially, all but two minerals (i.e., manjiroite and mannardite) in this group are classified as anhydrous. Nonetheless, trace amounts of water have been detected in some analyses of other members of the hollandite supergroup (e.g., coronadite and hollandite).

PROGRAM DESCRIPTION

WinHolclas is a user-friendly, compiled program package (≈11 Mb) developed for personal computers running on the Microsoft® Windows operating system. The program first calculates the cation and anion values (in apfu) from analyses made on hollandite supergroup by means of wet-chemical or electron-microprobe techniques and then uses these to classify the mineral

into the IMA-approved species that belong to two groups, including coronadite and priderite (see Table 1). A list of the calculation steps in the Calculation Screen and in the output of a Microsoft Excel file developed by the program is given in Table 2. Upon the successful installation of WinHolclas, the start-up screen with various pull-down menus and equivalent shortcuts appears on the screen (Figure 1a). The program allows the user to input wet-chemical or electron-microprobe hollandite-supergroup analytical data by clicking the New icon on the toolbar, by selecting the New File from the pull-down menu of the File option, or pressing the Ctrl+N keys (Figure 1b). Up to 42 chemical analytes (in wt%) are used by WinHolclas in the following order:

Sample No, SiO_2, TiO_2, ZrO_2, VO_2, MnO_2, Al_2O_3, Cr_2O_3, Fe_2O_3, Mn_2O_3, Sb_2O_3, V_2O_3, As_2O_3, Y_2O_3, La_2O_3, Ce_2O_3, Pr_2O_3, Nd_2O_3, As_2O_5, P_2O_5, V_2O_5, Nb_2O_5, Ta_2O_5, MoO_3, WO_3, UO_3, SO_3, FeO, MnO, CaO, BaO, SrO, MgO, ZnO, NiO, PbO, CuO, CoO, Na_2O, K_2O, Li_2O, Tl_2O, and H_2O.

Data from the analysis of a hollandite-supergroup mineral can also be input into a blank Excel file following the above order, saving it with the extension of ".xls" or ".xlsx". After this, it can be loaded into the Data Entry Screen of the program by clicking the Open Excel File option from the pull-down menu of File. By selecting the Edit Excel File option from the pull-down menu of File, data can be inserted into a blank Excel file (i.e., MyHollandite), saved using a different file name (with the extension of ".xls" or ".xlsx"), and then loaded into the Data Entry Screen of the program by clicking the Open Excel File option from the pull-down menu of File. Additional information about the data entry or similar topics can be accessed by pressing the F1 function key to display the WinHolclas.chm file on the screen. The current version of WinHolclas includes a total of 10 binary and ternary classification and compositional plots. Data on any of these plots can be displayed using the program Grapher by selecting the diagram type from the pull-down menu of Graph in the Calculation Screen of the program (Figure 1c).

WORKED EXAMPLES

Using the selected data set from literature as well as "Species.hol" file in the installation document (i.e., WinHolclas setup file), examples are presented that show how WinHolclas can be used in the determination of chemical formulas and hollandite-supergroup minerals classification. The previously typed or loaded analyses are processed by clicking the Calculate icon (i.e., ∑) in the Data Entry Screen of the program, after which all input and estimation parameters are displayed in columns

Table 2. Description of column numbers in the *Calculation Screen* window of WinHolclas program and an output Excel file.

Row	Explanations	Column number
1	Major oxides from the hollandite-supergroup mineral (HSM) compositions (wt.%)	1-43
2	Blank	44
3	Estimation of the stoichiometric H_2O, MnO_2, Mn_2O_3, FeO, and Fe_2O_3 contents (wt.%)	45-50
4	Blank	51
5	Recalculated ions from the HSM compositions (apfu)	52-94
6	Blank	95
11	Recalculated Mn^{4+}, Mn^{3+}, Fe^{3+}, and Fe^{2+} contents (apfu) based on the stoichiometric constraints	96-99
12	Blank	100
13	Total content of the A and M (apfu) in the general formula of HSM	101-102
14	Blank	103
15	Total cation charge of the A, M and (A+M) in the general formula of HSM	104-106
16	Blank	107
17	Dominant tunnel, M and charge-compensating cations	108-110
18	Blank	111
19	Group name and species from the HSM compositions	112-113
20	Blank	114

(apfu) = Atoms per formula unit.

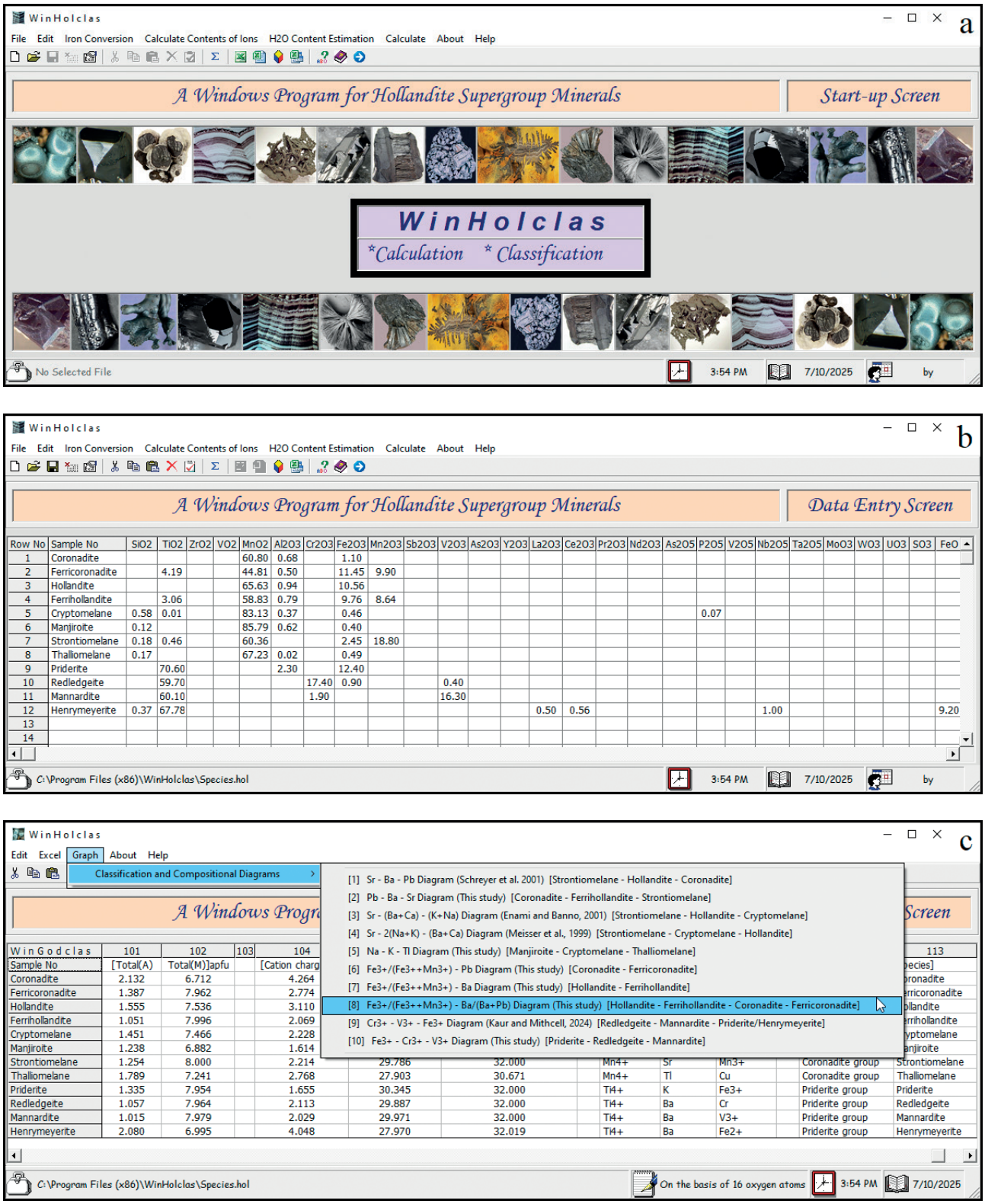


Figure 1. a) A screenshot of the WinHolclas Start-up window with various pull-down menus and equivalent shortcuts. b) A screenshot of the WinHolclas *Data Entry* window with a total of 42 analytes (wt%). c) A screenshot of the WinHolclas *Calculation Screen* with plot options from the pull-down menu of the *Graph*.

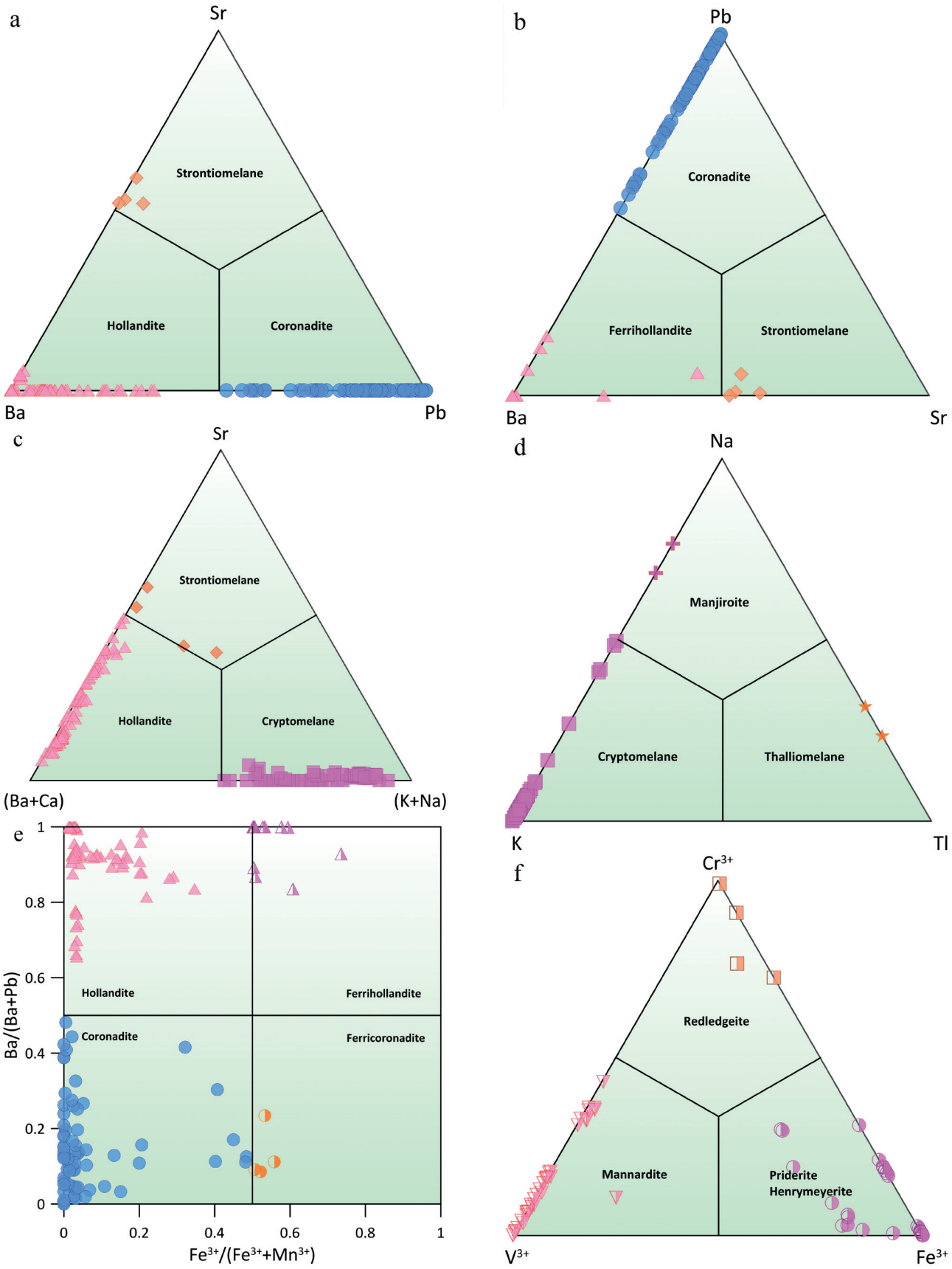
Table 3. Chemical compositions of selected hollandite-supergroup minerals with calculations and classifications by WinHolclas.

Row		S1	S2	S3	S4	S5	S6	S7	S8	S9	S10	S11	S12
1	SiO_2	0.00	0.00	0.00	0.00	0.58	0.12	0.18	0.17	0.00	0.00	0.00	0.37
2	TiO_2	0.00	4.19	0.00	3.06	0.01	0.00	0.46	0.00	70.60	59.70	60.10	67.78
3	MnO_2	60.80	44.81	65.63	58.83	83.13	85.79	60.36	67.23	0.00	0.00	0.00	0.00
4	Al_2O_3	0.68	0.50	0.94	0.79	0.37	0.62	0.00	0.02	2.30	0.00	0.00	0.00
5	Cr_2O_3	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	17.40	1.90	0.00
6	Fe_2O_3	1.10	11.45	10.56	9.76	0.46	0.40	2.45	0.49	12.40	0.90	0.00	0.00
7	Mn_2O_3	0.00	9.90	0.00	8.64	0.00	0.00	18.80	0.00	0.00	0.00	0.00	0.00
8	V_2O_3	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.40	16.30	0.00
9	La_2O_3	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.50
10	Ce_2O_3	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.56
11	P_2O_5	0.00	0.00	0.00	0.00	0.07	0.00	0.00	0.00	0.00	0.00	0.00	0.00
12	Nb_2O_5	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	1.00
13	FeO	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	9.20
14	MnO	7.12	0.00	5.12	0.00	2.08	3.18	0.00	0.00	0.00	0.00	0.00	0.00
15	CaO	0.00	0.00	0.00	0.00	0.27	0.22	0.14	0.00	0.00	0.00	0.00	0.02
16	BaO	0.00	5.16	17.59	14.63	0.13	0.16	3.95	3.53	6.70	20.20	19.40	18.25
17	SrO	0.00	0.00	0.00	2.77	0.00	0.00	9.15	0.01	0.00	0.00	0.00	0.00
18	MgO	0.00	0.00	0.00	0.00	0.05	0.18	0.00	0.05	0.00	0.00	0.00	0.00
19	ZnO	0.11	0.33	0.00	0.03	5.23	0.03	0.00	0.04	0.00	0.00	0.00	0.00
20	NiO	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.23	0.00	0.00	0.00	0.00
21	PbO	28.66	24.50	0.00	0.00	0.00	0.00	0.00	0.14	0.00	0.00	0.00	0.00
22	CuO	0.05	0.00	0.00	0.00	0.12	0.03	0.00	5.47	0.00	0.00	0.00	0.00
23	CoO	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.64	0.00	0.00	0.00	0.00
24	Na_2O	0.000	0.00	0.00	0.09	0.44	2.99	0.17	0.04	0.60	0.00	0.00	0.40
25	K_2O	0.00	0.00	0.00	0.05	3.50	1.39	1.42	0.14	5.60	0.00	0.00	0.05
26	Tl_2O	0.00	0.00	0.00	0.00	0.00	0.00	0.00	17.67	0.00	0.00	0.00	0.00
27	H_2O	1.11	0.00	0.00	0.00	0.000	3.92	0.00	0.32	0.00	0.00	0.00	0.00
28	Σ (wt%)	99.63	100.84	99.84	98.65	96.44	99.03	97.09	96.19	98.20	98.60	97.70	98.13
29	H_2O molecules $_{calculated}$ (wt%)	0.000	1.683	1.045	1.207	1.994	0.000	1.788	0.000	1.679	0.943	0.985	1.044
30	H_2O $_{calculated}$ (wt%)	0.000	0.000	0.004	0.037	0.099	0.099	0.081	0.081	0.050	0.039	0.064	0.052
31	MnO_2 $_{calculated}$ (wt%)	54.612	0.000	53.757	0.000	69.431	82.946	0.000	57.827	0.000	0.000	0.000	0.000
32	Mn_2O_3 $_{calculated}$ (wt%)	6.815	0.000	13.077	0.000	15.087	3.133	0.000	10.356	0.000	0.000	0.000	0.000
33	FeO $_{calculated}$ (wt%)	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	9.033
34	Fe_2O_3 $_{calculated}$ (wt%)	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.186
35	Si^{4+}	0.000	0.000	0.000	0.000	0.074	0.014	0.025	0.026	0.000	0.000	0.000	0.049
36	Ti^{4+}	0.000	0.493	0.000	0.318	0.001	0.000	0.047	0.000	6.484	5.995	6.034	6.815
37	Mn^{4+}	5.804	4.848	5.145	5.624	6.091	6.527	5.715	6.154	0.000	0.000	0.000	0.000
38	Al^{3+}	0.123	0.092	0.153	0.129	0.055	0.083	0.000	0.004	0.331	0.000	0.000	0.000
39	Cr^{3+}	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	1.836	0.200	0.000

Table 3. ...Continued

Row		S1	S2	S3	S4	S5	S6	S7	S8	S9	S10	S11	S12
40	Fe^{3+}	0.127	1.349	1.101	1.016	0.044	0.034	0.253	0.057	1.139	0.090	0.000	0.019
41	Mn^{3+}	0.658	1.180	1.136	0.910	1.202	0.224	1.960	1.001	0.000	0.000	0.000	0.000
42	V^{3+}	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.043	1.744	0.000
43	La^{3+}	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.025
44	Ce^{3+}	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.027
45	P^{5+}	0.000	0.000	0.000	0.000	0.008	0.000	0.000	0.000	0.000	0.000	0.000	0.000
46	Nb^{5+}	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.060
47	Fe^{2+}	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	1.010
48	Mn^{2+}	0.927	0.000	0.601	0.000	0.224	0.307	0.000	0.000	0.000	0.000	0.000	0.000
49	Ca^{2+}	0.000	0.000	0.000	0.000	0.037	0.027	0.021	0.000	0.000	0.000	0.000	0.003
50	Ba^{2+}	0.000	0.317	0.955	0.793	0.006	0.007	0.212	0.213	0.321	1.057	1.015	0.956
51	Sr^{2+}	0.000	0.000	0.000	0.222	0.000	0.000	0.727	0.001	0.000	0.000	0.000	0.000
52	Mg^{2+}	0.000	0.000	0.000	0.000	0.009	0.031	0.000	0.011	0.000	0.000	0.000	0.000
53	Zn^{2+}	0.012	0.038	0.000	0.003	0.490	0.003	0.000	0.005	0.000	0.000	0.000	0.000
54	Ni^{2+}	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.028	0.000	0.000	0.000	0.000
55	Pb^{2+}	1.186	1.032	0.000	0.000	0.000	0.000	0.000	0.006	0.000	0.000	0.000	0.000
56	Cu^{2+}	0.006	0.000	0.000	0.000	0.012	0.003	0.000	0.636	0.000	0.000	0.000	0.000
57	Co^{2+}	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.079	0.000	0.000	0.000	0.000
58	Na^{+}	0.000	0.000	0.000	0.024	0.108	0.660	0.045	0.012	0.142	0.000	0.000	0.104
59	K^{+}	0.000	0.000	0.000	0.009	0.567	0.202	0.248	0.028	0.872	0.000	0.000	0.009
60	Tl^{+}	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.770	0.000	0.000	0.000	0.000
61	OH^{-}	1.139	0.000	0.000	0.000	0.000	2.977	0.000	0.329	0.000	0.000	0.000	0.000
62	∑ (apfu)	8.844	9.349	9.091	9.047	8.928	8.120	9.253	9.030	9.288	9.021	8.993	9.075
63	$Mn^{4+}{}_{calculated}$	5.804	0.000	5.145	0.000	6.091	6.527	0.000	6.154	0.000	0.000	0.000	0.000
64	$Mn^{3+}{}_{calculated}$	0.658	0.000	1.136	0.000	1.202	0.224	0.000	1.001	0.000	0.000	0.000	0.000
65	$Fe^{3+}{}_{calculated}$	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.019
66	$Fe^{2+}{}_{calculated}$	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	1.010
67	∑ A (apfu)	2.132	1.387	1.555	1.051	1.453	1.238	1.254	1.789	1.335	1.057	1.015	2.080
68	∑ M (apfu)	6.712	7.962	7.536	7.996	7.467	6.882	8.000	7.241	7.954	7.964	7.979	6.995
69	∑ Cation charge A	4.264	2.774	3.110	2.069	2.231	1.614	2.214	2.768	1.655	2.113	2.029	4.048
70	∑ Cation charge M	25.940	29.226	27.753	29.931	28.567	27.185	29.786	27.903	30.345	29.887	29.971	27.970
71	∑ Cation charge (A+M)	30.204	32.000	30.864	32.000	30.798	28.799	32.000	30.671	32.000	32.000	32.000	32.019
72	Tunnel cation	Pb^{2+}	Pb^{2+}	Ba^{2+}	Ba^{2+}	K^{+}	Na^{+}	Sr^{2+}	Tl^{+}	K^{+}	Ba^{2+}	Ba^{2+}	Ba^{2+}
72	M cation	Mn^{4+}	Mn^{4+}	Mn^{4+}	Mn^{4+}	Mn^{4+}	Mn^{4+}	Mn^{4+}	Mn^{4+}	Ti^{4+}	Ti^{4+}	Ti^{4+}	Ti^{4+}
73	DCC cation	Mn^{3+}	Fe^{3+}	Mn^{3+}	Fe^{3+}	Mn^{3+}	Mn^{3+}	Mn^{3+}	Cu^{2+}	Fe^{3+}	Cr^{3+}	V^{3+}	Fe^{2+}
74	Group	CorG	CorG	CorG	CorG	CorG	CorG	CorG	CorG	PdrG	PdrG	PdrG	PdrG
75	Species	Cor	Fcor	Hol	Fhol	Cml	Mji	Sml	Tml	Pdr	Red	Man	Hmy

Note: (apfu) = Atoms per formula unit. Samples sources: S1, S2, S3, S5, S6, S9, S10, S11, S12 = from Handbook of Mineralogy (Anthony et al., 2001-2005); S4 = from Biagioni et al. (2014); S7 = from Meisser et al. (1999); S8 = from Gołębiowska et al. (2021). CorG = Coronadite group; PdrG = Priderite group; Cor = Coronadite, Fcor = Ferricoronadite, Hol = Hollandite, Fhol = Ferrihollandite, Cml = Cryptomelane, Mji = Manjiroite, Sml = Strontiomelane, Tml = Thalliomelane, Pdr = Priderite, Red = Redledgeite, Man = Mannardite, Hmy = Henrymeyerite. The formulas were recalculated to content of ions on the basis of 16 oxygens (apfu).


a
Sr
Strontiomelane
Hollandite
Coronadite
Ba
Pb
b
Pb
Coronadite
Ferrihollandite
Strontiomelane
Ba
Sr
c
Sr
Strontiomelane
Hollandite
Cryptomelane
(Ba+Ca)
(K+Na)
d
Na
Manjiroite
Cryptomelane
Thalliomelane
K
Tl
e
Hollandite
Ferrihollandite
Coronadite
Ferricoronadite
Ba/(Ba+Pb)
Fe3+/(Fe3+Mn3+)
0
0.2
0.4
0.6
0.8
1
f
Cr3+
Redledgeite
Mannardite
Priderite
Henrymeyerite
V3+
Fe3+

1-113 of the Calculation Screen (see Table 2). Pressing the Ctrl+F keys or clicking the Open File to Calculate option from the Calculate menu also executes the processing of a selected data file with the extension of ".hol" which refers to the hollandite supergroup. By clicking the Send results to Excel file icon in the Calculation Screen, all calculations can be stored in an Excel file (Output.xlsx) and then displayed by clicking the Open and edit Excel file icon.

The validity of program output has been tested with representative coronadite and priderite group minerals selected from the literature (see references in Table 3 and Figure 2). WinHolclas calculates the compositional formula for a given hollandite-supergroup mineral analysis on the basis of 16 oxygen atoms (apfu). Alternatively, by clicking the Octahedral Cations = 8.00 (apfu) Normalization option from the pull-down menu of Calculate Contents of Ions in the Start-up Screen or Data Entry Screen, the program calculates hollandite-supergroup minerals according to the selected criteria. All input and calculated parameters are presented in the Calculation Screen and Excel output file for the selected mineral analyses. In the general formula for minerals in the coronadite group, manganese exists in oxidation states of Mn^{3+} and Mn^{4+}, with the latter being the predominant form in tunnel oxides. If the total manganese is already given as total MnO_2 (wt%), for example, in the chemical composition of the coronadite group, the program allows users to estimate the partition of Mn^{4+} and Mn^{3+} based on neutral charge balance requirements. In some of the older chemical analyses, total manganese content may be given as MnO (wt%) for the coronadite group minerals. In this case, first, the MnO to MnO_2 conversion should be applied to those analyses, and then calculated by the program. For example, a coronadite group mineral with the following analytical data (see Table 2 in Corona-Esquivel et al., 2000; wt%); FeO 0.84, MnO 63.27, BaO 13.75, ZnO 2.22, CuO 7.04, total 87.12 is defined as coronadite. If these values are entered into the program, WinHolclas will perform the necessary calculations but will not be able to carry out any classification processes. However, by applying the conversion procedure to the same mineral analysis, with the following analytical data (wt%); FeO 0.84, MnO_2 77.54, BaO 13.75, ZnO 2.22, CuO 7.04, total 101.39, it yields the empirical formula $Ba_{0.717}(Mn^{4+}_{5.981}Mn^{3+}_{1.151}Cu^{2+}_{0.708}Zn^{2+}_{0.218}Fe^{3+}_{0.093})_{\sum 8.868}O_{16}$ on the basis of 16 oxygen atoms (apfu) and classifies the mineral as hollandite according to the current hollandite-supergroup nomenclature scheme proposed by Biagioni et al. (2013).

Classification of a given analysis as a distinct mineral species into its proper group is carried out based on the combination of DTC and DCCC by the program. The total Fe present in ferrihollandite and priderite is commonly expressed as Fe_2O_3 (wt%) due to the DCCC from mineral analysis obtained by electron-microprobe technique. Although spectroscopic methods are useful in determining the valence state of iron in the hollandite-supergroup minerals, especially for ferrihollandite, priderite, and henrymeyerite, studies on this matter are limited. According to McCammon et al. (1999), the separation of ferric and ferrous iron from microprobe-derived total FeO (wt%) analysis based on the stoichiometric approach (e.g., Droop, 1987) is not reliable, as all naturally occurring priderite group titanates and many synthetic examples are known to be nonstoichiometric. For example, in a priderite mineral analysis, the total iron

Figure 2. Selected plots of classification and compositional diagrams of the hollandite-supergroup species from the pull-down menu of *Graph* in the *Calculation Screen* of the WinHolclas program, using the selected mineral analyses from the literature. a) Compositional plot of the coronadite-group minerals in a ternary Sr-Ba-Pb diagram (from Schreyer et al., 2001) [filled diamond from Schreyer et al. (2001), Enami and Banno (2001); filled triangles from Albuquerque et al. (2017), Jirásek et al. (2015); filled circles from Albuquerque et al. (2017), Liakopoulos et al. (2001)]. b) Compositional plot of the coronadite-group minerals in a ternary Pb-Ba-Sr diagram (this study) [filled diamond from Albuquerque et al. (2017), Jirásek et al. (2015); filled circles from Albuquerque et al. (2017), Liakopoulos et al. (2001); filled triangles from Albuquerque et al. (2017), Cabella et al. (1992), Enami and Banno (2001), Miura et al. (1987), Biagioni et al. (2014)]. c) Compositional plot of the coronadite-group minerals in a ternary Sr-(Ba+Ca)-(K+Na) diagram (from Enami and Banno, 2001) [filled diamond from Schreyer et al. (2001), Enami and Banno (2001); filled triangles from Albuquerque et al. (2017), Jirásek et al. (2015); filled squares from Jirásek et al. (2022), Liakopoulos et al. (2001), Niu et al. (2021)]. d) Compositional plot of the coronadite-group minerals in a ternary Na-K-Tl diagram (this study) [filled squares from Jirásek et al. (2022), Liakopoulos et al. (2001), Niu et al. (2021); filled plus from Nambu and Tanida (1967), Gutzmer and Beuks (2000); filled stars from Gołębiowska et al. (2021)]. e) Compositional plot of the coronadite-group minerals in a $Fe^{3+}/(Fe^{3+}+Mn^{3+})$ versus Ba/(Ba+Pb) diagram (this study) [filled triangles from Albuquerque et al. (2017), Jirásek et al. (2015); filled circles from Albuquerque et al. (2017), Liakopoulos et al. (2001); half-filled circles from Albuquerque et al. (2017), Chukanov et al. (2016); half-filled triangles from Miura et al. (1987), Albuquerque et al. (2017), Biagioni et al. (2014), Cabella et al. (1992), Enami and Banno (2001)]. f) Compositional plot of the priderite-group minerals in a ternary Cr^{3+}-V^{3+}-Fe^{3+} diagram (from Kaur and Mitchell, 2024) [half-reverse filled squares from Gatehouse et al. (1986), Karpenko et al. (2011), Rezvukhin et al. (2020); half-reverse filled triangles from Fu et al. (2023), Karpenko et al. (2011), Reznitsky et al. (2018), Scott and Peatfield (1986), Yang et al. (2024); half-filled circles from Allialy et al. (2011), Chakhmouradian and Mitchell (2001), Foley et al. (1994), Gaspar et al. (1994), Kogarko et al. (2007), Maitra and Bhattacharyya (2015), Reguir et al. (2003)].

content is given as FeO (8.36 wt%) with the following analytical data (see Table 1 in Gaspar et al., 1994; wt%); TiO_2 79.36, Al_2O_3 0.06, Cr_2O_3 0.08, V_2O_3 0.52, Nb_2O_5 0.17, FeO 8.36, CaO 0.15, BaO 1.95, SrO 0.01, MgO 0.27, Na_2O 0.10, K_2O 9.26, total 100.29. Using the ferric and ferrous iron separation method by Droop (1987) for this sample, the program estimated FeO and Fe_2O_3 contents from total FeO 8.36 (wt%) as 6.702 and 1.842 (wt%), respectively. WinHolcals determines the DTSCC, DTC and DCCC as Ti^{4+}, K^+, and Fe^{2+}, respectively. As no species corresponding to this composition exists in the current classification scheme (i.e., DTSCC=Ti^{4+}, DTC=K^+, DCCC=Fe^{2+}), the program designates it as 'Not classified' rather than applying the name of one of known species listed in Table 1. However, with the conversion of total FeO to total Fe_2O_3 the program classifies the analysis as priderite.

WinHolclas calculates and classifies the hollandite-supergroup mineral analyses in the Calculation Screen, as well as in the output Excel file, based on the current nomenclature scheme (e.g., see rows 1-71 in Table 3), with the determination of dominant constituents, groups and, specific species (see rows 72-75 in Table 3). The number of ions is calculated on the basis of 16 oxygen atoms (apfu). In a case where a chemical composition corresponds to a hitherto unknown species within the hollandite supergroup (i.e., a new species), WinHolclas warns the user with a "Not classified" statement in column 113 of the Calculation Screen for species. WinHolclas provides options to display binary and ternary classification and compositional diagrams in the Calculation Screen by using the Grapher program. Some of these plots, with the selected hollandite-supergroup mineral data from the literature, are given in Figure 2. All input and calculated parameters from the Output tab of an Excel file (i.e., Output.xlsx) are automatically transposed by the Transpose tab of the program. This procedure provides the user with the ability to prepare a quick table for presentation as well as publication by using the Copy-Paste option.

SUMMARY AND AVAILABILITY OF THE PROGRAM

WinHolclas is a user-friendly program that is specially developed for personal computers running on the Windows operating system to estimate and classify the hollandite-supergroup minerals using data obtained from both electron-microprobe and wet-chemical analyses. The program processes multiple analytical data sets (up to 200) for each program execution. The current version of WinHolclas recalculates ions and classifies a total of 12 species for a given analysis into one of two groups (i.e., hollandite and priderite) using the current IMA-approved nomenclature scheme (Biagioni et al., 2013). The program generates two main windows. The first window (i.e., Start-up/Data Entry Screen), with several pull-down menus and equivalent shortcuts, enables users to edit a given hollandite-supergroup mineral analysis based on chemistry (wt%). By clicking the Calculate icon (i.e., ∑) in the Data Entry Screen, all input and estimated parameters by WinHolclas are displayed in the second window (i.e., Calculation Screen). The program reports the output in a tabulated form with a numbered column from 1 to 113 in the Calculation Screen window, as well as in an Output Excel file. These columns include hollandite supergroup compositions (wt%), recalculated ion values (apfu), dominant cations (i.e., DTC and DCCC), groups (i.e., (DTSCC), and species. The results in the Calculation Screen can be exported to a Microsoft® Excel file (i.e., Output.xlsx), by clicking the Send Results to Excel File (Output.xlsx) icon or selecting the Send Results to Excel File (Output.xlsx) option from the pull-down menu of Excel. This file is then opened by Excel by clicking the Open and Edit Excel File (Output.xlsx) icon or selecting the Open Excel File (Output.xlsx) option from the pull-down menu of Excel. WinHolclas is a compiled program that consists of a self-extracting setup file containing all the necessary support files (i.e., "dll" and "ocx") for the 32-bit system. By clicking the setup file, the program and its associated files (i.e., support files, help file, data files with the extension of ".hol", ".xls", ".xlsx" and plot files with the extension of ".grf" are installed into the personal computer (i.e., the directory of C:\Program Files\WinHolclas or C:\Program Files (x86)\WinHolclas) on Windows XP and subsequent operating systems. An installation of the program into a personal computer with a 64-bit operating system may require the "msflexgrd" adjustment (see explanations in the Supplementary Material). The self-extracting setup file (i.e., the WinHolclas setup.exe file) is approximately 11 Mb and can be obtained from the journal's server as a Supplementary Material.

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