1	Concerning Tetrahedrites: How Much to Lump and How Far to Sp	lit?
2	Word counts Total: 3182 Abstract, text, acknowledgements and refer	ences:
3	3123	
4	Revision #1	
5		
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9		
10	Abstract	
11	Currently there are two related but distinct approaches to the classification	ı of
12	minerals. The traditional time-independent classification uses rules specified	by the
13	IMA-CNMNC that can carefully split mineral species, but may elide valuable in	nformation
14	about their formation. In contrast, an emerging time-dependent classification	appears to
15	be able to add to our knowledge about planetary evolution, yet may lump min	erals into
16	broadly defined kinds even if important distinctions should be made. An exam	ination of
17	the tetrahedrite group provides valuable insights on both approaches. As new	′ly
18	redefined by Biagioni, et al. (2020), the generalized tetrahedrite formula	
19	$(A_6(B_4C_2)D_4X_{12}Z_1)$ has six sites that can accommodate substitutions and a system	stematic
20	splitting of all possibilities could lead to more than 200 unique species. In con	trast,
21	applying guidelines for lumping largely as suggested by Hazen, et al. (2022) of	ould lead

- to a single kind. Deciding how much to lump and how far to split may ultimately depend
- 23 on the intentions of the observer.
- 24
- 25 E-mail: johnsonne@vt.edu
- 26 **Keywords:** philosophy of mineralogy, tetrahedrite, nomenclature, classification

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28

Introduction

29 The name tetrahedrite is well known in mineralogy, it being the most common 30 sulfosalt mineral. But an abundance of additional names have been used and/or 31 proposed as a result of its complicated chemistry and history as a source of copper and 32 silver. From the argentum rude album of Agricola (1546) through the grey ores (fahlerz 33 and *cuivre gris*) of the 18th century, numerous variants in the 19th century (Palache, et 34 al., 1944), to modern work, perhaps never in the field of mineralogy has a single mineral 35 been split so often by so many. But despite the complexity of its nomenclature, the 36 structure of tetrahedrite is straightforward. It can be considered as a sulfide analogue to 37 the framework aluminosilicate sodalite (Barth, 1932), with transition metal - sulfur 38 tetrahedra in place of the (AI, Si)O₄ tetrahedra. The 'cage' generated by this framework 39 is generally collapsed down onto a transition metal - sulfur octahedron (rather than 40 hosting a large anion or polyanion), and alternating rings of the framework are spanned 41 by four semimetal - sulfur trigonal pyramids (Biagioni, et al., 2020). 42 Two factors have complicated mineralogical and material science research on 43 tetrahedrite. First, the variety of possible chemical substitutions emphasized the 44 inadequacy of the historical nomenclature and made a redefinition sorely needed. 45 Formal work on this was initiated by the IMA-COM (Möelo, et al., 2008) and culminated 46 with the application of current IMA-CNMNC guidelines to the tetrahedrite group 47 (Biagioni, et al., 2020). The second factor lies in the promise tetrahedrite has shown as 48 a thermoelectric material. As a semiconductor, tetrahedrite has a relatively high 49 electrical conductivity, but vibrations of metal atoms within the cage and the lone

50 electron pairs of the semi-metals result in a poor thermal conductivity. This produces the 51 'phonon glass, electron crystal' (PGEC) phenomenon (Beekman, et al., 2015) that 52 generates electricity. Over the last two decades this research has curiously left many of 53 the known tetrahedrite compositions poorly investigated. The focus has largely been on 54 the Sb-dominant compositions (tetrahedrites sensu stricto) synthesized with 55 substitutions for the framework cations, including some unlikely to occur naturally, such 56 as AI (Tippireddy, et al., 2020), or Mg (Levinsky, et al., 2018). Part of this focus is due to 57 the lower toxicity of Sb-bearing phases compared with other thermoelectric materials 58 like Bi₂Te₃ or PbTe (Suekuni, et al., 2013; Makin, et al., 2022). But it is also likely due in 59 part to unfamiliarity with the nomenclature, owing (for non-mineralogists) to the 60 bewildering assortment of names. 61 The recent re-examination of the structure and chemistry of tetrahedrite (sensu lato)

by Biagioni, et al. (2020) resulted in a formal redefinition and a number of named series and species, and their Table 2 included all extant and a number of probable, but not yet formally recognized, species. With its complicated chemistry and tangled history of names, such a review and redefinition was long overdue and Biagioni, et al. (2020) accomplished this in a manner both thorough and thoughtful. It should come as no surprise, however, that even such a detailed review might leave some aspects of the mineral group unaddressed or incompletely considered:

•The nature of the classification requires very careful chemical analyses and, in
some cases, structural analyses to determine Z-site occupancies, which precludes any
use in the field or routine petrographic study. This contrasts with the approach taken for

the eudialyte group wherein the contents of the X-sites are ignored in assigning an

- 73 appropriate mineral name owing to difficulties in determining CI, F, OH, O, CO₃, H₂O
- and vacancies during routine analyses. (Johnsen, et al., 2003).

•It defines a freibergite series while eliminating the eponymous mineral itself and

76 combines within this series minerals with both occupied and vacant Z-sites, inconsistent

77 with previous IMA recommendations for the classification of alkali-group, calcic-group

and X-vacant group tourmalines (Henry, et al., 2011).

•It discusses Z-site vacancies solely in the context of kenoargentotetrahedrite-(Fe),

80 despite long standing questions about the presence or absence of a 13th sulfur atom in

81 tetrahedrite (effectively an occupied or unoccupied Z-site, Johnson, et al. 1986, Sack, et

82 al. 2022). The subsequent discoveries of kenorozhdestvenskayaite-(Zn) (Qu, K., et al.,

83 in press a), and kenorozhdestvenskayaite-(Fe) (Qu, K., et al., in press b) suggests that

84 this may be a topic requiring further consideration.

•It does not address similarities between the tetrahedrite group and the galkhaite

group (Chen and Szymanski, 1981; Kasatkin, et al., 2018), which are also made up of a

87 sodalite-like framework of transition metal - sulfur tetrahedra (in the case of galkhaite,

HgS₄), but with each cage occupied by an atom of Cs or TI instead of a transition metal

- sulfur octahedron. The subsequent discovery of posepnyite (Skacha, et al., 2020),

90 where the dominant cation in the B site is Hg instead of Cu suggests a connection

91 between the two groups which may also require additional consideration.

•It speculates on the possibility of new tellurian species arsenogoldfieldite and

93 stibiogoldfieldite, both of which have now been reported (Sejkora, et al., 2022; Biagioni,

94 et al., 2022), but only briefly mentioned the possibility of cations other than copper in the

95 framework. This likely due to Makovicky and Karup-Moller (2017) who found

96 substitution, omission and exchange calculations on natural tellurian tetrahedrites

97 (sensu lato) to be hindered by poor or incomplete data.

98 The redefinition has also predictably rekindled a debate, since it can be argued that

99 it is a template for excessive splitting (Nikischer, 2020) or that it presents opportunities

100 to more precisely characterize new species (such as the 15 new Sb-dominant minerals

101 since the publication of Biagioni, et al. 2020). But regardless of any weaknesses or

102 perspective, their Table 2 only lists minerals by formula rather than indicating which

103 sites accommodate which substitutions in which series or species. A more

104 comprehensive table could both clarify areas for future research and uniquely illuminate

the lumping vs. splitting question, thereby impacting a current philosophical discussion

106 in mineralogy.

107 Hazen (2019) proposed a new type of mineralogical classification, one very different

108 than that which currently exists and driven by the need to interpret data collected during

109 planetary exploration missions. This was termed an evolutionary (i.e. time-dependent)

110 system to contrast it with the current time-independent IMA guidelines. He

111 demonstrated that a time-independent classification may elide important contextual

112 information on mineral formation, illustrating this with a discussion of different diamond

113 morphologies and properties that arise from different parageneses. Hazen & Morrison

- 114 (2022) surveyed mineral formation environments and identified a series of 57 unique
- 115 paragenetic modes which could be used to classify minerals in this evolutionary

116 scheme. Following this, Hazen, et al. (2022) explored the lumping and splitting of 117 minerals in this context, providing criteria for determining when to lump or split. They 118 suggest lumping minerals only if part of a continuous solid solution, if they are 119 isostructural or part of a homologous series, and form in the same paragenetic mode. 120 Splitting should occur only if they form via two or more paragenetic modes, and/or 121 cluster analyses reveal multiple and distinct sets of attributes. To stress test these 122 criteria, they examined eight mineral groups in detail (cancrinite, eudialyte, hornblende, 123 jahnsite, labuntsovite, sartorite, tetradymite, tourmaline) in which 20 or more current 124 species could be lumped together into natural kinds. Obviously, the present work also 125 addresses this debate, albeit from a different perspective, and raises the question if the 126 newly defined tetrahedrite group could provide additional insight.

127

128

Lumping vs Splitting: Speculation ad Absurdum?

129 As shown above, a time-dependent classification might be considered more closely 130 tied to mineral lumping whereas a time-independent one could be thought skewed 131 towards mineral splitting. To apply this to the tetrahedrite group, let us first speculate as 132 to how much lumping could occur. All of the tetrahedrite minerals are isostructural and 133 to date, there is no evidence of anything but complete solid solution across all 134 compositions. The lumping question then comes down to the number of observed 135 paragenetic modes. An examination of the RRUFF database (https://rruff.info/ima; 136 accessed 3 July 2023, Downs, 2006) finds (as of this writing) 28 extant tetrahedrites 137 (sensu lato) with listed paragenetic modes . Of these, all have PM33 (hydrothermal

metal deposits) as a paragenetic mode and only three list any others (two with PM32
(Ba-Mn-Pb-Zn metamorphics) and one with PM37 (layered igneous deposits)). Clearly,
most tetrahedrites would meet all the criteria for lumping, and relaxing the paragenetic
mode criterion only slightly could allow for complete lumping. This extreme case would
then turn the tetrahedrite group into a single natural mineral kind - tetrahedrite.

143 But what of maximal splitting? Table 1 is a version of Table 2 of Biagioni, et al.

144 (2020), focusing only on Sb-dominant compositions, but describing other substitutions

145 more exhaustively. It includes the extant series and species (as of this writing), adds a

146 number of speculative series and species (in italics), and specifies which of the six

147 structural sites accommodates which substitutions following the conventions used by

148 Biagioni, et al. (2020). It must be emphasized that these speculative series and species

149 are just that, speculations for the purpose of exploring this debate and not a 'back-door'

150 attempt to further redefine the tetrahedrite group without IMA-CNMNC sanction. For

151 compactness sake, the tetrahedrite series is the only one completely filled out with eight

152 unique species; the rest can be completed in an analogous fashion. The table also:

•Speculates about a Cu-rich series with the Z-site unoccupied. Following the

154 convention established by Biagioni, et al. (2020) this series is referred to as

155 "kenotetrahedrite".

Abandons the freibergite series of Biagioni, et al. (2020), dividing it into separate
speculative series "argentotetrahedrite" and "kenoargentotetrahedrite".

158	 Speculates about a "kenohakite" series, an "argentohakite" series, and a 				
159	"kenoargentohakite" series in direct analogy with the matching series - extant and				
160	speculative - for tetrahedrite.				
161	 Speculates about a "kenorozhdestvenskayaite" series to accommodate the newly 				
162	discovered kenorozhdestvenskayaite-(Fe) (Qu, K., et al., in press b).				
163	•Speculates about an "unnamed series 1" and "kenounnamed series 1" as the				
164	Se-dominant analogs of the rozhdestvenskayaite and "kenorozhdestvenskayaite"				
165	series.				
166	 Leaves out any discussion of posepnyite due to the ambiguity discussed above. 				
167	The result of this splitting speculation is twelve separate series, each with eight				
168	unique species, for a total of 96 possible tetrahedrites. Accommodating these				
169	characteristics as well as speculative Se - S ordering and the As-dominant and				
170	Te-significant (2 or more atoms per formula unit) minerals, could result in more than 200				
171	unique mineral species.				
172					
173	Implications				
174	With a potential range of one kind to more than 200 species, at what point are we				
175	lumping that which should be split or splitting that which should be lumped? As noted by				
176	Hazen, et al. (2022), Darwin originated the terminology but could offer no definitive				
177	guidance and Santana (2019) suggests the development of a hard-and-fast rule may be				
178	neither useful nor necessary. The contrast between splitting minerals into narrowly				
179	defined species or lumping them together into broader kinds resembles past				

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180 developments in both taxonomy and igneous petrology. Advances in DNA sequencing 181 now allow for species splitting when morphological characteristics might suggest 182 lumping (Bickford, et al., 2007) and the IUGS classifications of igneous rocks 183 (Streckesien, 1976) allow for lumped field classifications and split laboratory 184 classifications based on modal analyses (QAPF). 185 Consider whether the Fe, Mn, Ni, and Zn tetrahedrite variants should be lumped into 186 a single kind or split into separate species as they currently are. From the perspective of 187 a mining company, these are truly distinctions without a difference, since their interests 188 are solely in the amounts of available Cu and Ag in the mineral. Contrast this with the 189 utility of tetrahedrite as a thermoelectric material. Figure 1 is a plot of the dimensionless 190 thermoelectric figure of merit (ZT) as a function of temperature for synthetic 191 tetrahedrite-(Fe), tetrahedrite-(Mn), tetrahedrite-(Ni), and tetrahedrite-(Zn) (Heo, et al., 192 2014; Wang, 2016; Weller, 2018). The significant differences between these ZT values 193 make the trivial distinction of the mining industry into a serious research question for 194 condensed matter physics. It would seem that sometimes lumping is the better choice, 195 whereas in others splitting is preferable, and which path should be chosen is entirely 196 dependent on the aim of the research. 197

198

Acknowledgements

This work began as an exploration of the tetrahedrite name space, inspired by Tony Nikischer and furthered by a discussion with Shauna Morrison. It relies heavily on the extraordinary work on proposal IMA 18-K by Cristian Biagioni and coauthors, and the

- 202 equally extraordinary work by Bob Hazen and collaborators on the ideas of mineral
- 203 evolution. Exceptionally helpful comments and discussion were provided by Mickey
- 204 Gunter, Igor Pekov and an anonymous reviewer.
- 205
- 206

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321	Figure captions
322	
323	Figure 1. Plot of the dimensionless thermoelectric figure of merit (ZT) versus
324	temperature for synthetic C-site substituted tetrahedrites.
325	

	Dominant site occupant						
Series	A site (6	6) B site ((4) C site (2)	D site (4	4) Y site (1	2)Z site	
<u>Tetrahedrite</u>	Cu	Cu	Variable	Sb	S	S	
Tetrahedrite-(Cd)*	Cu	Cu	Cd	Sb	S	S	
Tetrahedrite-(Cu)*	Cu	Cu	Cu	Sb	S	S	
Tetrahedrite-(Fe)	Cu	Cu	Fe	Sb	S	S	
Tetrahedrite-(Hg)*	Cu	Cu	Hg	Sb	S	S	
Tetrahedrite-(In)*	Cu	Cu	In	Sb	S	S	
Tetrahedrite-(Mn)*	Cu	Cu	Mn	Sb	S	S	
Tetrahedrite-(Ni)*	Cu	Cu	Ni	Sb	S	S	
Tetrahedrite-(Zn)	Cu	Cu	Zn	Sb	S	S	
Kenotetrahedrite	Cu	Cu	Variable	Sb	S	Vaca	
Argentotetrahedrite	Ag	Cu	Variable	Sb	S	S	
Argentoetrahedrite-(Cd)*	Ag	Cu	Cd	Sb	S	S	
Argentoetrahedrite-(Fe)	Ag	Cu	Fe	Sb	S	S	
Argentoetrahedrite-(Hg)*	Ag	Cu	Hg	Sb	S	S	
Argentotetrahedrite-(Zn)*	Ag	Cu	Zn	Sb	S	S	
Kenoargentotetrahedrite	Ag	Cu	Variable	Sb	S	Vaca	
Kenoargentoetrahedrite-(Fe)*	Ag	Cu	Fe	Sb	S	Vacar	
Kenoargentoetrahedrite-(Zn)	' Ag	Cu	Zn	Sb	S	Vacar	
_	2						
Hakite	Cu	Cu	Variable	Sb	Se	Se	
Hakite-(Cd)*	Cu	Cu	Cd	Sb	Se	Se	
Hakite-(Fe)*	Cu	Cu	Fe	Sb	Se	Se	
Hakite-(Hg)	Cu	Cu	Hg	Sb	Se	Se	
Hakite-(Zn)*	Cu	Cu	Zn	Sb	Se	Se	

358								
359	Kenohakite	Cu	Cu	Variable	Sb	Se	Vacant	
360								
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362								
363	Argentohakite	Ag	Cu	Variable	Sb	Se	Se	
364								
365	Kenoargentohakite	Ag	Cu	Variable	Sb	Se	Vacant	
366								
367	<u>Rozhdestvenskayaite</u>	Ag	Ag	Variable	Sb	S	<u>S</u>	
368	Rozhdestvenskayite-(Zn)	Ag	Ag	Zn	Sb	S	S	
369								
370	Kenorozhdestvenskayaite	Ag	Ag	Variable	Sb	S	Vacant	
371	Kenorozhdestvenskayite-(Fe)*	Ag	Ag	Fe	Sb	S	Vacant	
372								
373	Unnamed Series 1	Ag	Ag	Variable	Sb	Se	Se	
374								
375	Kenounnamed Series 1	Ag	Ag	Variable	Sb	Se	Vacant	
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377	Extant series and species ar	e in pla	ain text. Sp	peculative se	eries and	d species a	re in italics	
378	Species noted with an "*" are	e newly	y described	d - either in p	print or i	n press - sir	nce the	
379	redefinition by Biagioni, et al. (2020).							
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Figure 1

