

Reply

Reply to Bosi *et al.* (2023)

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Introduction

I began thinking about the issue of defining new mineral species about 25 years ago, and soon realised that a straightforward way to approach this complicated issue is (1) to consider all possible end-member arrangements of integer charges over the sites in a crystal structure, subject to the constraint of electroneutrality, and (2) then consider all possible arrangements of ions over the sites in that crystal structure for each of those end-member charge arrangements. The result is a list of all possible end-member formulae with that general crystal-structure arrangement (Hawthorne, 2002), and I gave various examples in that paper. Gagné and Hawthorne (2016) did this for the milarite structure-type and Hawthorne *et al.* (2018) did this for the hyalotekite structure-type, and gave this method the name *Chemographic Exploration*. Hawthorne (2021) provided a mathematical proof that a dominant end-member formula can always be written for a mineral or a crystal structure. Hawthorne (2002) also defined the characteristic properties of an end-member and showed that a single site in an end-member structure may be occupied by two different ions. Later, I realised that this characteristic may be modified by short-range order as the local version of bond-valence theory can constrain the types of short-range order that are possible, introducing additional constraints on arrangements of ions (Hawthorne, 2021).

From 1992 onward, various members of the Commission on New Minerals, Nomenclature and Classification of the International Mineralogical Association (IMA–CNMNC) developed rules for defining new-mineral species (Nickel, 1992; Nickel and Grice, 1998; Hatert and Burke, 2008; Bosi *et al.*, 2019a, 2019b). Hawthorne (2023) is a critical review of this work, but it was not meant to be disparaging; the rules of the IMA–CNMNC are very important for maintaining order in the use of mineral names and mineral formulae. However, the goals of Science are (1) to produce a fundamental and consistent understanding of Nature, and (2) to develop laws and rules, based on this fundamental understanding, that describe Nature as accurately as possible. Rules that ‘work’ only part of the time are merely a stop-gap until better understanding can produce better laws and rules, and Hawthorne (2023) is an evaluation of some of the issues that need to be addressed with regard to the definition of distinct mineral species. The rule of the dominant constituent and its various ‘correction rules’ produce formulae that fit the

definition of an end-member perhaps 95% of the time and allow the CNMNC to evaluate the validity of most new-mineral proposals. However, 95% of the time is not good enough for Science. We need to work toward a better understanding of minerals and the development of rules that work 100% of the time.

I will consider many of the statements made by Bosi *et al.* (2023) and to a lesser extent by Bosi *et al.* (2019a, 2019b), labelling them below as Statements, followed by my Response.

In their Introduction and Discussion, Bosi *et al.* (2023) make the following two statements:

Statement [1]: “Thus, according to this author, the chemical formulae resulting from application of the IMA–CNMNC rules can violate the requirements of an end-member, particularly that of electroneutrality, and these cannot derive end-member formulae for some groups of minerals. *In this discussion, we will show that such a statement is incorrect.*” [my bold italics]

Statement [2]: “Hawthorne (2023) nicely discusses specific examples where the dominant-constituent rule and dominant-valency rule lead to non-electroneutral formulae, thus violating the conservation of electric charge.”

Response: In Statement [1], the authors say that they will show that the statement “...application of the IMA–CNMNC rules can violate the requirements of an end-member, particularly that of electroneutrality...” is incorrect, whereas in Statement [2], they say that I “nicely discuss specific examples where the dominant-constituent rule and dominant-valency rule lead to non-electroneutral formulae”.

Most of Hawthorne (2023) is devoted to showing examples where purported ‘end-member’ chemical formulae derived from the IMA–CNMNC rules violate electroneutrality. It is beyond my understanding how the authors can contest the truth of the statement that “the chemical formulae resulting from application of the IMA–CNMNC rules can violate the requirements of an end-member, particularly that of electroneutrality, and these cannot derive end-member formulae for some groups of minerals” when “specific examples” of this in Hawthorne (2023) are noted in their Statement [2]. Their Statement [2] directly contradicts their Statement [1].

In their Discussion, Bosi *et al.* (2023) make the following statement:

Statement [3]: “In this regard, it is instructive to show a misunderstanding by Hawthorne (2023) on the STC method.” and Bosi *et al.* (2019a) make the following statement:

Statement [4]: “As the sequential use of the dominant-valency and the dominant constituent rules are recommended by the

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IMA–CNMNC, it is worthwhile to analyse their basic aspects, here identified as follows: (1) the principle of the formula electroneutrality; and (2) the definition of an end-member formula (Hawthorne, 2002) that includes at most one site with double occupancy (i.e. heterovalent pair of ions or ion–vacancy pair). *In addition, the concepts of total charge and charge constraint at the crystallographic sites should be considered.* [my italics]

Response: It seems to have escaped the attention of some of the authors of Bosi *et al.* (2019a, 2023) that the title of Hawthorne (2002) is as follows: “The use of end-member charge-arrangements in defining new mineral species and heterovalent substitutions in complex minerals”.

In that paper, I considered (among other things) the ^[4](Li, Zn)-bearing members of the milarite group (general formula $A_2B_2CT(2)_3T(1)_{12}O_{30}(H_2O)_{0-n}$) and used the aggregate charges at the sites in these structures together with the constraint of overall electroneutrality to assign end-member charge arrangements and then end-member chemical formulae to these minerals. Hawthorne (2002) states “All of these minerals have K dominant at the C site and Si dominant at the T(1) site, and hence these sites can be considered as fixed in the following discussion. The total anion charge is 60^- , the charge at C (= K) and T(1) (= Si_{12}) is 49^+ , and hence the electroneutrality principle requires that the aggregate charge at the A, B and T(2) sites is 11^+ . The possibilities for occupancy of the T(2) site and the resulting charge-constraints on the A and B sites are as follows...”. This approach used the aggregate charge at each site and the constraint of overall electroneutrality.

Gagné and Hawthorne (2016) developed this approach further and calculated all possible arrangements of charges at the sites in the milarite structure, subject to overall electroneutrality; those with a total T(1) charge of 48^+ (= Si_{12}) are listed in Table 1, those with a total T(1) charge of $< 48^+$ can be found in table 5 of Gagné and Hawthorne (2016). They then examined ~350 chemical analyses from the literature and identified the 29 distinct end-member formulae that are the dominant constituent in one or more of these chemical analyses, 23 of which correspond to the approved minerals of the milarite group; the remaining six dominant distinct end-member formulae did not correspond to named mineral species (although they may be considered as potential species for possible approval by IMA–CNMNC). Gagné and Hawthorne (2016) labelled this method *Chemographic Exploration* as it derives all possible end-member charge arrangements and then identifies the end-member chemical formulae that correspond to these charge arrangements. End-member chemical formulae corresponding to these end-member charge arrangements allow identification of the dominant end-member formula for any mineral formulae considered. Hawthorne *et al.* (2018) applied this method to the minerals of the hyalotekite group, again showing chemical compositions in the literature that correspond to a new end-member charge arrangement and a potential new-mineral species.

Statement [4] of Bosi *et al.* (2019a) ends with the following sentence: “In addition, the concepts of total charge and charge constraint at the crystallographic sites should be considered”. The “concepts of total charge and charge constraint at the crystallographic sites” were considered in detail by Hawthorne (2002), Gagné and Hawthorne (2016) and Hawthorne *et al.* (2018). The implied contention of the authors that the STC method of Bosi *et al.* (2019a, 2019b) is a new approach is false; the name is new but the method is not new. Bosi *et al.* (2019a, 2019b) used the concepts of total charge and charge constraint at the

Table 1. Arrangements* of aggregate charge at each site of the milarite structure-type with Si = 12 apfu.

Number	Charge at (A + B + C) sites	A ₂	B ₂	C	T(1) ₁₂	T(2) ₃	Charge at [T(1) + T(2)] sites
[1]	3	0 ₂	1 ₂	1 ₁	4 ₁₂	3 ₃	57
[2]	3	1₂	0₂	1₁	4₁₂	3₃	57
[3]	3	0 ₁ 1 ₁	1 ₂	0 ₁	4 ₁₂	3 ₃	57
[4]	3	1 ₂	0 ₁ 1 ₁	0 ₁	4 ₁₂	3 ₃	57
[5]	4	1 ₂	1 ₂	0 ₁	4 ₁₂	3 ₂ 2 ₁	56
[6]	4	2 ₂	0 ₂	0 ₁	4 ₁₂	3 ₂ 2 ₁	56
[7]	4	0 ₂	2 ₂	0 ₁	4 ₁₂	3 ₂ 2 ₁	56
[8]	5	0 ₂	2 ₂	1 ₁	4 ₁₂	3 ₁ 2 ₂	55
[9]	5	1 ₂	1 ₂	1 ₁	4 ₁₂	3 ₁ 2 ₂	55
[10]	5	2₂	0₂	1₁	4₁₂	3₁2₂	55
[11]	6	0 ₂	2 ₂	2 ₁	4 ₁₂	2 ₃	54
[12]	6	2 ₂	0 ₂	2 ₁	4 ₁₂	2 ₃	54
[13]	6	1 ₂	1 ₂	2 ₁	4 ₁₂	2 ₃	54
[14]	6	1 ₂	2 ₂	0 ₁	4 ₁₂	2 ₃	54
[15]	6	2₂	1₂	0₁	4₁₂	2₃	54
[16]	6	0 ₁ 1 ₁	2 ₂	1 ₁	4 ₁₂	2 ₃	54
[17]	6	2₂	0₁1₁	1₁	4₁₂	2₃	54
[18]	6	1₁2₁	1₂	1₁	4₁₂	2₃	54
[19]	6	1 ₂	1 ₁ 2 ₁	1 ₁	4 ₁₂	2 ₃	54
[20]	6	3₂	0₂	0₁	4₁₂	2₃	54
[21]	6	2₁3₁	0₂	1₁	4₁₂	2₃	54
[22]	7	3 ₂	0 ₂	1 ₁	4 ₁₂	2 ₂ 1	53
[23]	7	1 ₂	2 ₂	1 ₁	4 ₁₂	2 ₂ 1	53
[24]	7	2₂	1₂	1₁	4₁₂	2₂1	53
[25]	8	2 ₂	2 ₂	0 ₁	4 ₁₂	2 ₁ 2	52
[26]	8	3₂	1₂	0₁	4₁₂	2₁2	52
[27]	8	4 ₂	0 ₂	0 ₁	4 ₁₂	2 ₁ 2	52
[28]	9	4 ₁ 5 ₁	0 ₂	0 ₁	4 ₁₂	1 ₃	51
[29]	9	4₂	0₂	1₁	4₁₂	1₃	51
[30]	9	4 ₂	0 ₁ 1 ₁	0 ₁	4 ₁₂	1 ₃	51
[31]	9	3 ₁ 4 ₁	1 ₂	0 ₁	4 ₁₂	1 ₃	51
[32]	9	3₂	1₂	1₁	4₁₂	1₃	51
[33]	9	2 ₁ 3 ₁	2 ₂	0 ₁	4 ₁₂	1 ₃	51
[34]	9	2 ₂	2 ₂	1 ₁	4 ₁₂	1 ₃	51

* Those charge arrangements shown in bold are the ones that correspond to observed end-member compositions of minerals, potential minerals and synthetics.

crystallographic sites without acknowledging that it was developed by Hawthorne (2002) or used extensively by Gagné and Hawthorne (2016) and Hawthorne *et al.* (2018).

Statement from Bosi *et al.* (2019a): “The application of the site-total-charge approach should be used when the simple application of the dominant-valency rule leads to an unbalanced end-member formula.”

Response: There are two problems with this statement:

(1) There is no such thing as an ‘unbalanced end-member formula’; an end-member formula is neutral by definition, and this fact was emphasised extensively by Hawthorne (2023). They should have written “...when the simple application of the dominant-valency rule leads to an unbalanced formula”. While this is a minor error in the context of this sentence, it provides an interesting insight into the attitude of some members of CNMNC concerning criticism of their rules by other scientists.

(2) The major problem involves the hybrid nature of the so-called STC approach as stated above by Bosi *et al.* (2019a). In the above statement, they link the use of the STC approach to the dominant-valency rule which, as shown by Hawthorne (2023), violates the electroneutrality principle. Why link it to a rule that is fundamentally wrong? If all possible end-member charge arrangements are calculated according to the chemographic-exploration method of Gagné and Hawthorne (2016),

and the aggregate charges at each site in the mineral of interest are matched up with the nearest end-member charge arrangement as in Gagné and Hawthorne (2016) and Hawthorne *et al.* (2018), there is no need to couple any other rule to this approach.

Statement [5]: “The end-member formula is an *overriding* condition to identify minerals, something that Bosi *et al.* (2019a, 2019b) mentioned in their papers but which seems to have been overlooked by Hawthorne (2023) who cited their results.”

Response: Two of the authors of Bosi *et al.* (2023) seem to have forgotten that they co-authored a paper with me (Hawthorne *et al.*, 2021) entitled “Ontology, archetypes and the definition of mineral species” in which the following definition is given: “a specific mineral species is defined...by the following set of universals: name, *end-member formula* [my bold italics] and Z, space group, and bond topology of the end-member structure, with the range of chemical composition limited by the compositional boundaries between end members with the same bond topology”. I am aware that I am somewhat absent-minded, but do they think that I would overlook a key result of one of my own papers? Besides, the key role of the end-member formula has been recognised for many years: a brief investigation with Google Scholar produced references going back at least 80 years that deal with this issue.

Statement [6]: “The next step is to decide which of these two approaches best reflects the mineral properties.”

Response: What properties are the approaches meant to reflect? Refractive index, hardness, lustre? And what is meant by ‘reflect’? This statement is so vague as to be meaningless.

Statement [7]: “Moreover, it is important to note that the deficiency in these IMA–CNMNC rules was already noted by Bosi (2018) and successively addressed by the CNMNC with the paper by Bosi *et al.* (2019a).”

Response: This statement is a weak attempt to discredit Hawthorne (2023). Why is it “important to note that the deficiency in these IMA–CNMNC rules was already noted by Bosi (2018)”? I corresponded extensively with Ernst Burke (then Chair of CNMNC) in 2005–2006 about this issue, and these deficiencies in the IMA–CNMNC rules were recognised in the literature (Hatert and Burke, 2008) by the new rules that were developed to ostensibly correct the initial deficiency of the rule of the dominant constituent.

Statement [8]: “Constructive proposals for alternative improved procedures are always desirable. CNMNC is an IMA Commission with elected representatives from national mineralogical societies. Proper place for constructive discussions on matters handled by the CNMNC would primarily be those societies and through their representatives in CNMNC.”

Response: This statement is merely a piece of political propaganda. Scientific advance is driven by scientists, not by organisations of elected individuals. The latter invariably have a vested interest in, and are reluctant to perturb, the *status quo*. On the one hand, I can sympathise with this view as the CNMNC does

an amazing amount of good work regulating the approval of new minerals (most of which will not be affected by the current discussion). On the other hand, Science advances primarily by the ideas and actions of individual scientists which collectively influence the community as a whole. There is no ‘proper place’ for such activity except the scientific literature (*sensu lato*), and there should be no constraints on the scientists who can participate.

Statement [9]: “Exceptions to the CNMNC rules are possible and welcome if soundly argued, as Nature does not read mineralogical papers.”

Response: Nature may not read mineralogical papers but Nature must conform to the symmetry properties of space-time and the resulting conservation laws, whereas the current IMA–CNMNC rules do not.

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