



Structural and chemical complexity of minerals: correlations and time evolution

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Abstract: Correlations between chemical and structural complexities of minerals were analysed using a total of 4962 datasets on the chemical compositions and 3989 datasets on the crystal structures of minerals. The amounts of structural and chemical Shannon information per atom and per unit cell or formula unit were calculated using the approach proposed by Krivovichev with no H-correction for the minerals with unknown H positions. Statistical analysis shows that there are strong and positive correlations ($R^2 > 0.95$) between the chemical and structural complexities and the number of different chemical elements in a mineral. Analysis of relations between chemical and structural complexities provides strong evidence that there is an overall trend of increasing structural complexity with the increasing chemical complexity. Following Hazen, four groups of minerals were considered that represent four eras of mineral evolution: “ur-minerals”, minerals from chondritic meteorites, Hadean minerals, and minerals of the post-Hadean era. The analysis of mean chemical and structural complexities for the four groups demonstrate that both are gradually increasing in the course of mineral evolution. The increasing complexity follows an overall passive trend: more complex minerals form with the passage of geological time, yet the simpler ones are not replaced. The observed correlations between the chemical and structural complexities understood in terms of Shannon information suggest that, at a first approximation, chemical differentiation is a major force driving the increase of complexity of minerals in the course of geological time. New levels of complexity and diversification observed in mineral evolution are achieved through the chemical differentiation, which favours local concentrations of particular rare elements and creation of new geochemical environments.

Key-words: mineral evolution; structural complexity; chemical complexity; Shannon information; chemical differentiation.

1. Introduction

The idea that complexity increases in evolution of the Universe has been widely discussed in different contexts, including biological evolution and the evolution of matter in general (e.g., Chaisson, 2001; McShea & Brandon, 2010; Lineweaver *et al.*, 2013). The concept of mineral evolution first proposed in Russian mineralogical literature (e.g., Zhabin, 1981; Yushkin, 1982) was recently developed from a general viewpoint (Hazen *et al.*, 2008, 2011; Hazen, 2013, 2014) and applied to Hg minerals (Hazen *et al.*, 2012), clay minerals (Hazen *et al.*, 2013a), carbon minerals (Hazen *et al.*, 2013b), Be minerals (Grew & Hazen, 2014) and B minerals (Grew *et al.*, 2016), leading to the formulation of mineral ecology as a field of mineralogy dealing with the study of mineral distributions in space and time (Hazen *et al.*, 2015a and b; Hystad *et al.*,

2015). The consideration of mineral evolution as a new avenue of research in mineralogy raises several important questions concerning complexity of minerals and its behaviour in the course of the evolution of the Universe.

There is no widely accepted definition of complexity of biological systems, although different measures of complexity of organisms have been proposed, including the number of cells and the number of cell-types, the gene number, and the number of different interactions of parts of an organism (McShea, 1996; Carroll, 2001). However, no universal quantitative complexity measure exists that can be applied to all biological species and their systems, even though the problem of a general trend of complexity in biological evolution has been discussed since the XIXth century (Lamarck, 1809). There is little doubt that minerals are much less complex than biological organisms, but the problem of complexity has not been

considered in mineralogy as it was in biology. As minerals are crystalline chemical compounds of natural origin, it would be appropriate to apply to them complexity measures proposed for crystals in general. However, even in crystallography, until recently there were no universal complexity measures that could be applied to all crystalline compounds, though many proposals have been put forward by various crystallographers, starting with Pauling (1929). Krivovichev (2012) proposed to estimate complexity of crystals on the basis of Shannon information content of a reduced unit cell, taking into account the number of atoms, the number of independent sites and the ratio of their multiplicities. Krivovichev (2016) demonstrated that the Shannon information content per atom correlates to a configurational entropy of crystals. Information-based complexity measures can be applied to study general trends in structural complexity *versus* chemical composition, its behaviour in the course of crystallization (Krivovichev, 2013; Cempírek *et al.*, 2016; Krivovichev *et al.*, 2016, 2017) and micro- and macroscale mineral evolution (Krivovichev, 2013; Grew *et al.*, 2016). It is important to note that Shannon information measures were first applied to the investigations of complexity of geochemical systems in general (*i.e.*, including minerals) by Petrov (1970) and Bulkin (1972a and b) (see also Yushkin, 1977).

The aim of the present study is to investigate the relations between chemical and structural complexity of minerals, both understood as amounts of Shannon information, and to apply these measures to mineral evolution, using the lists provided by Hazen *et al.* (2008) and Hazen (2013).

2. Methodology

For the investigation of chemical and structural complexity of minerals, a total of 4962 datasets on chemical compositions and 3989 datasets on crystal structures of minerals was considered. The amounts of structural Shannon information per atom ($^{str}I_G$) and per unit cell ($^{str}I_{G,total}$) were calculated using the approach developed by Krivovichev (2012, 2013, 2014, 2015, 2016) according to the following equations:

$$^{str}I_G = - \sum_{i=1}^k p_i \log_2 p_i \quad (\text{bits/atom}), \quad (1)$$

$$^{str}I_{G,total} = -vI_G = -v \sum_{i=1}^k p_i \log_2 p_i \quad (\text{bits/cell}), \quad (2)$$

where k is the number of different crystallographic orbits (independent crystallographic Wyckoff sites) in the structure and p_i is the random choice probability for an atom from the i th crystallographic orbit, that is:

$$p_i = m_i/v, \quad (3)$$

where m_i is a multiplicity of a crystallographic orbit (*i.e.*, the number of atoms of a specific Wyckoff site in the reduced unit cell), and v is the total number of atoms in the reduced unit cell.

The calculation of the information-based structural complexity parameters using equations (1)–(3) implies that positions of all atoms in the crystal structures are known. However, it is not always the case, as in crystal structures of hydrated oxysalt minerals very frequently positions of H atoms could not be determined due to the experimental problems associated with crystal quality and the low scattering power of H for X-rays, most frequently used in crystal-structure analysis. For the crystal structures of hydrated minerals with no H positions determined, Pankova *et al.* (2018) proposed to use the procedure of H-correction by introducing “dummy” H atoms into structural datasets. According to our general estimates (see also Pankova *et al.*, 2018), the H-correction for the crystal structures with unknown H positions may result in a significant increase of their structural information measures, depending upon their H content. However, no H-correction was applied to the datasets under consideration, and therefore the results we obtained can be considered only as a first approximation.

By analogy with structural complexity, chemical complexity was evaluated by the amount of chemical information per atom ($^{chem}I_G$) and per formula unit, f.u. ($^{chem}I_{G,total}$), as suggested by Siidra *et al.* (2014). Following this approach, for the idealized chemical formula of a mineral or inorganic compound, $E_{c_1}^{(1)} E_{c_2}^{(2)} \dots E_{c_k}^{(k)}$, where $E^{(i)}$ is an i th chemical element in the formula and c_i is its integer coefficient, the chemical information can be calculated as follows:

$$^{chem}I_G = - \sum_{i=1}^k p_i \log_2 p_i \quad (\text{bits/atom}), \quad (4)$$

$$^{chem}I_{G,total} = -eI_G = -v \sum_{i=1}^k p_i \log_2 p_i \quad (\text{bits/f.u.}), \quad (5)$$

where k is the number of different elements in the formula and p_i is the random choice probability for an atom of the i th element, that is:

$$p_i = c_i/e, \quad (6)$$

where e is the total number of atoms in the chemical formula:

$$e = \sum_{i=1}^k c_i. \quad (7)$$

The ideal chemical formulas of minerals used for the calculations are those approved by the International Mineralogical Association (IMA) and contained in the continuously updated lists published by Pasero (2016) at the website of Commission on New Minerals, Nomenclature and Classification IMA (CNMNC IMA). Only essential chemical elements were taken into account, without consideration of isomorphic substitutions (see also Krivovichev *et al.*, 2018).

Table 1. Information-based mean chemical and structural complexities of minerals separated into mineral-system types according to the number N of different chemical elements in the chemical formula.*

N	n	$^{chem}I_G$ (bits/atom)			$^{chem}I_{G,total}$ (bits/f.u.)			n	$^{str}I_G$ (bits/atom)			$^{str}I_{G,total}$ (bits/cell)		
		M	SD	SEM	M	SD	SEM		M	SD	SEM	M	SD	SEM
1	53	0	0	0	0	0	0	53	0.58	1.05	0.14	13	36	5
2	405	0.937	0.075	0.004	5.5	7.6	0.4	405	1.88	1.40	0.07	76	240	12
3	723	1.373	0.117	0.004	14.3	14.4	0.5	593	2.44	1.11	0.05	104	194	8
4	1347	1.612	0.135	0.004	41.4	41.9	1.1	1347	3.43	1.12	0.04	229	283	9
5	1400	1.770	0.151	0.004	71.5	73.2	2.0	951	3.66	0.98	0.03	276	423	14
6	613	1.881	0.163	0.007	102.3	97.5	3.9	386	4.02	0.96	0.05	356	405	21
7	287	1.959	0.155	0.009	130.9	120.9	7.1	180	4.25	0.79	0.06	391	361	27
8	101	2.040	0.151	0.015	159.4	105.5	10.5	59	4.74	0.88	0.12	583	479	62
9	26	2.062	0.155	0.030	218.1	119.8	23.5	11	4.84	0.38	0.12	583	213	64
10	7	2.061	0.072	0.027	408.7	272.8	106.9	4	4.87	0.39	0.20	761	100	50

* n = number of minerals taken into account; M = arithmetic mean; SD = standard deviation; SEM = standard error of mean.

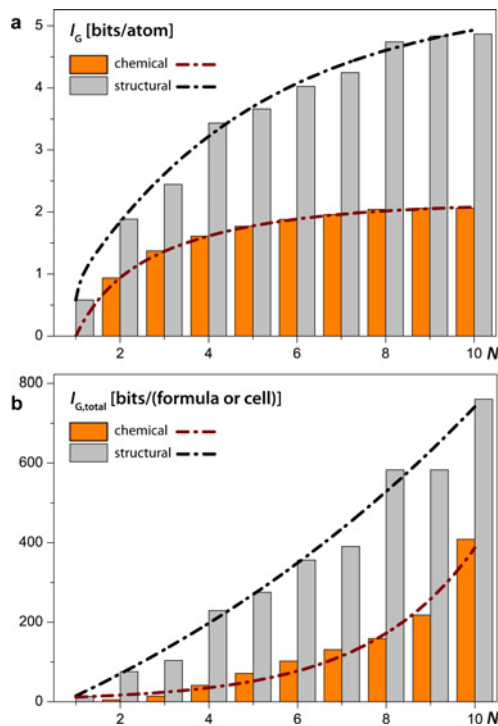


Fig. 1. Chemical and structural complexity parameters for minerals plotted against the number N of different chemical elements in a chemical formula (the mineral-system type): Shannon information per atom (a) and per unit cell or formula unit (b). The dash-and-dot lines are fitted curves corresponding to equations (8)–(11). See text for details.

3. Results

3.1. Complexity and mineral systems

Information-based chemical and structural complexity parameters for minerals calculated following equations (1)–(7) have been separated into groups according to the number N of different chemical elements present in the chemical formulae (*i.e.*, into different mineral systems)

(Krivovichev & Charykova, 2013, 2015, 2016; Krivovichev *et al.*, 2017). The mean chemical and structural complexities and associated statistical parameters are given in Table 1. Figure 1 shows distribution of Shannon information among different types of mineral systems. The dependencies of different I_G values from N were best approximated by the use of the following functions (corresponding curves are plotted as dash-and-dot lines in Fig. 1):

$$^{chem}I_G = -2.9 + 2.2 \times [1 - \exp(-N/2.8)] + 2.8 \times [1 - \exp(-N/0.7)] \quad (R^2 = 0.999), \quad (8)$$

$$^{str}I_G = -6595 + 5.85 \times [1 - \exp(-N/4.2)] + 6595 \times [1 - \exp(-N/0.1)] \quad (R^2 = 0.988), \quad (9)$$

$$^{chem}I_{G,total} = 2.2 + 10.8 \times \exp[(N - 1.2)/2.5] \quad (R^2 = 0.959), \quad (10)$$

$$^{str}I_{G,total} = -605 + 26.7 \times \exp[(N + 35)/11.6] \quad (R^2 = 0.974). \quad (11)$$

The observed relations indicate that there is a strong and positive correlation between the chemical and structural complexities and the number of essential chemical elements in a mineral.

3.2. Correlations between chemical and structural complexity

Figure 2 shows the relations between chemical and structural complexities per atom (Fig. 2a) and per unit cell or formula (Fig. 2b) calculated for different groups of minerals with the same number of chemical elements in a chemical formula. The best fitting for the $^{chem}I_G$ vs. $^{str}I_G$ and $^{chem}I_{G,total}$ vs. $^{str}I_{G,total}$ relations was obtained by

Table 2. Mean (and the 95% confidence intervals) chemical and structural complexities of minerals for the four eras of mineral evolution.

Era	n	Reference	$^{\text{chem}}I_G$ (bits/atom)	$^{\text{str}}I_G$ (bits/atom)	$^{\text{chem}}I_{G,\text{total}}$ (bits/f.u.)	$^{\text{str}}I_{G,\text{total}}$ (bits/cell)
1	12	Hazen <i>et al.</i> (2008)	0.92 ± 0.29	1.57 ± 0.81	7.3 ± 6.1	50.1 ± 56.6
2	62	Hazen <i>et al.</i> (2008)	1.11 ± 0.10	1.71 ± 0.25	12.7 ± 4.9	57.1 ± 34.1
3	422	Hazen (2013)	1.46 ± 0.04	2.73 ± 0.15	38.6 ± 5.0	157.6 ± 30.4
4	3989	Post-Hadean era	1.58 ± 0.01	3.25 ± 0.04	50.1 ± 2.3	230.0 ± 11.6

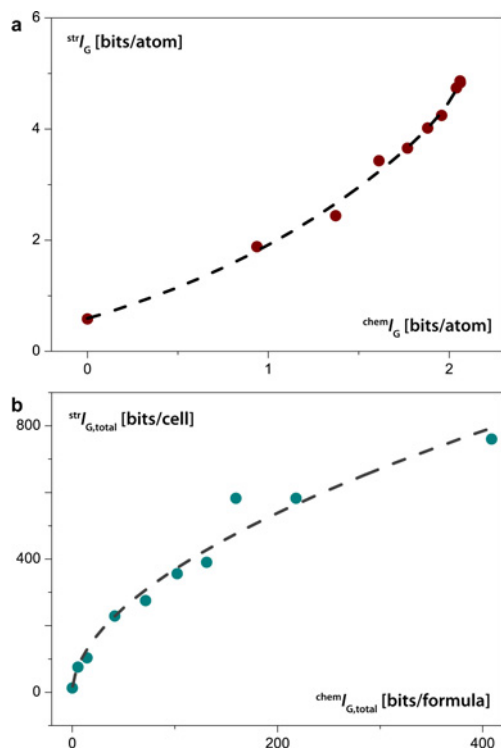


Fig. 2. Dependencies between structural and chemical complexities for different kinds of mineral systems: Shannon information per atom (a) and per unit cell or formula unit (b). Each point corresponds to a particular number N of different essential elements in a chemical formula. The dash-and-dot lines are fitted curves corresponding to equations (12) and (13). See text for details. (Online version in color.)

means of the exponential and allometric functions, respectively:

$$^{\text{str}}I_G = -0.97 + 2.3 \times [\exp(^{\text{chem}}I_G - 0.646)/1.63] \quad (R^2 = 0.988), \quad (12)$$

$$^{\text{str}}I_{G,\text{total}} = 30.2 \times [^{\text{chem}}I_{G,\text{total}}]^{0.544} \quad (R^2 = 0.957). \quad (13)$$

The observed relationships provide strong evidence that there is an overall trend of increasing structural complexity with the increasing chemical complexity of minerals.

3.3. Evolution of complexity in geological time

According to Hazen *et al.* (2008, 2013a and b), mineral evolution can be subdivided into four partially overlapping stages, each of which saw the expansion of mineralogical diversity and/or variation in relative mineral abundances.

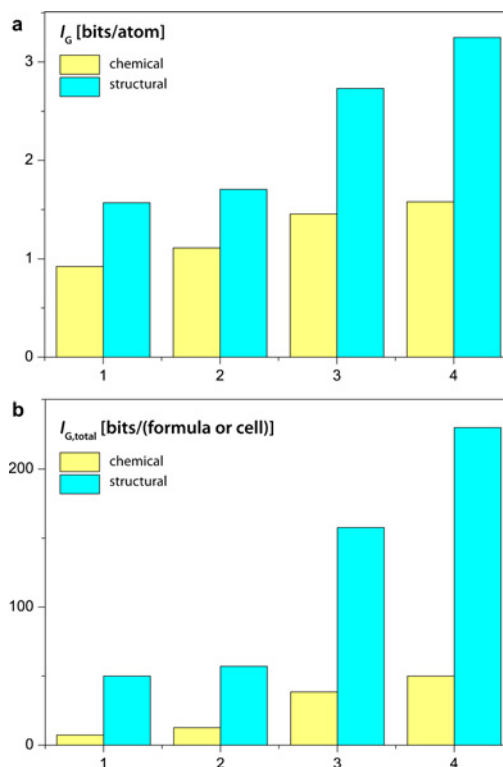


Fig. 3. Mean chemical and structural complexities for minerals occurring in different eras of mineral evolution (1 = “ur-minerals”; 2 = minerals of chondritic meteorites; 3 = minerals of the Hadean epoch; 4 = minerals of the post-Hadean era); Shannon information per atom (a) and per unit cell or formula unit (b).

The starting point of mineral evolution is that of the “ur-minerals”, the twelve earliest mineral phases to appear in the pre-solar nebulae (1). Chondritic meteorites incorporate about 60 primary mineral phases, which constitute the second phase (2). For the Hadean Eon, Hazen (2013) estimated 425 mineral species (3), whereas post-Hadean processes were responsible for the appearance of more than 5000 mineral species known today (4). The mean chemical and structural complexities for the four groups of minerals mentioned above provided in Table 2 and depicted in diagrams in Fig. 3 demonstrate that both chemical and structural complexity are gradually increasing in the course of mineral evolution.

4. Discussion

The obtained statistical correlations between information-based chemical and structural complexities of minerals and the general trend of increasing complexity in the

course of mineral evolution show that minerals become more and more complex with the flow of geological time. This agrees well with the general trend of chemical differentiation of matter as indicated by Yushkin (1982) and Hazen *et al.* (2008). In fact, the observed relations between chemical and structural complexity suggest that, at a first approximation, chemical differentiation is a major force driving the increase of chemical and structural complexity in the course of geological time. Indeed, in a companion paper, Krivovichev *et al.* (2018) showed that the average number N of chemical elements in a mineral increases for the different eras of mineral evolution, in agreement with the results of information-based considerations reported herein. It should, however, be taken into account that the list of minerals of the present era contains all minerals formed in the post-Hadean time. On the example of boron minerals, Grew *et al.* (2016) demonstrated that the variation of complexity in minerals follows a passive trend: more complex minerals arise with the passage of time, yet the simple ones are not replaced. The current work shows that this conclusion is valid for all minerals as well, with the global trend being passive due to the general increase in variance. New levels of complexity and diversification observed in mineral evolution are achieved through the chemical differentiation, which favours local concentrations of particular rare elements and creation of new geochemical environments.

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