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SOFTWARE

COMBINATORIAL COMPLEXITY OF THE SIGNATURE OF A NATURAL TILING

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Abstract. An additive model has been developed for calculating the combinatorial (Shannon-like) complexity of a signature of the natural tiling, which is used to describe the topological properties of micro- and mesoporous materials, in particular, zeolites. To calculate the complexity of this type, a Python program code has been compiled. The code was tested for tilings of a zeolite type. Correlations of the calculated complexity of a signature of the tiling and the combinatorial complexity of the tiling-generating structure were found.

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INTRODUCTION

Shannon's information entropy H [1], also known as Shannon's or combinatorial complexity, is usually expressed in bits and in this case has the form:

$$H = -\sum_{i=1}^{s} p_i \log_2 p_i \,, \qquad (1)$$

where $p_i = m_i/m$ is the proportion of m_i elements of the i-th type in a discrete set of m elements, each element of which is assigned to one of s types, $\sum_{i=1}^{s} p_i = 1$. Combinatorial complexity is used in chemistry as a measure of the complexity of molecular assemblies [2], including dendrimers [3], and its changes during chemical transformations [4–7], and in crystallography as a measure of the complexity of crystal structures [8–17] and chemical compositions [18], as well as systems of generating operations of crystallographic groups [19] and systems of supporting contacts in the crystal structure [20]. In mineralogy, information entropy was used by Academician N.P. Yushkin to assess the complexity of the distribution of minerals by syngonies [21]. The use of information entropy in mineralogy and crystallography is also the subject of a mini-review by Yu.L. Voitekhovsky [22].

Combinatorial complexity belongs to a broader class of species diversity indices called Hill numbers, since it is a first-order Hill number [23]. In crystallography, Hill numbers can be used to estimate the unevenness of distributions of records in structural data banks for crystallographic taxa such as syngonies, crystal classes, and space groups [24]. Species diversity of inanimate objects has some similarities with the diversity of biological species [25].

In this paper, we study the combinatorial complexity of the signature of a natural tiling [26]. A tiling is a normal (face to face) partition of space into tiles, which are generalized, not necessarily convex polyhedra, in which each vertex is incident to two or more vertices, and the faces can be curvilinear [27]. Each tiling corresponds to a certain grid formed by vertices and edges. A tiling is called natural if it meets the following conditions: the symmetry of the tiling coincides with the symmetry of the corresponding grid; the faces of the tile are strong rings (cycles that are not the sum of several smaller cycles); all strong rings of the grid, except for those belonging to the faces, have intersections, i.e. common interior points; if, in accordance with the previous points, different tilings are possible, then a single tiling is constructed by combining tiles with each other. Thus, if a natural tiling for a given grid exists at all, then there is only one. The search for natural tiling is generally non-trivial, but has been implemented in the ToposPro software package for a long time [28]. It is common to use a face symbol of the form $[A^a.B^b...]$ for tiles, which means that the tile a has A-gonal faces, b B-gonal faces, $A < B < ...$. Fig. 1 shows the natural tile $[4^{36}]$ of the pyrochlore structure. It is common to designate tiling with a signature showing the ratio of tiles, for example, the entry $2[3^4] + [3^8]$ means tetrahedral $[3^4]$ and octahedral $[3^8]$ tiles in a ratio of 2:1.

METHOD

For summing up the combinatorial complexity from several sources of information, simple additivity

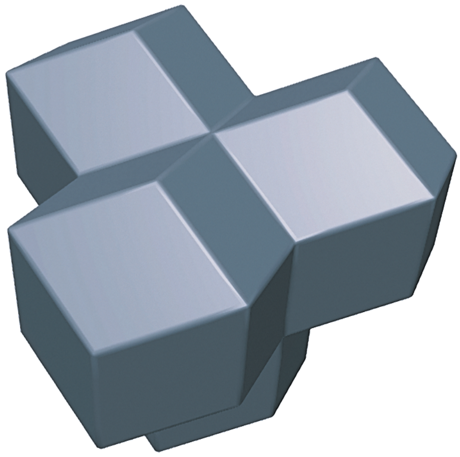

Fig. 1. Tile [4^{36}] of the eponymous natural tiling of the pyrochlore structure $Ca_2Nb_2O_6F$.

is only suitable if the sources of information are independent [29]. For example, if two subsystems of one system are isolated from each other, i.e. separated in space and do not exchange mass and energy. But if the subsystems are interdependent, as is always the case with substructures of one crystal structure [17], then strong additivity must be satisfied: in addition to the terms corresponding to individual subsystems, the sum includes an additional term that takes into account the increase in information due to mixing of subsystems. In Sabirov's works (e.g., [30]), such an additional term is called an emergent parameter. The principles of decomposition of the combinatorial complexity of discrete multisets into contributions of individual sets with an emergent parameter were considered in detail in [13].

Let us apply this approach to the signature of natural tiling, written in the standard form:

$$k_1\left[A_1^{a1}.B_1^{b1}...\right]+k_2\left[A_2^{a2}.B_2^{b2}...\right]+\ \dots$$

$$\dots+k_n\left[A_1^{an}.B_1^{bn}...\right], \qquad (2)$$

where k_1:k_2:...:k_n is the simplest integer relation between n tiles of different sorts. The set of faces of the tile of the i-th sort ($i = 1, ..., n$) is a multiset $\Phi_i = \{(A_i, a_i), (B_i, b_i), ...\}$. The combinatorial complexity of such a multiset by definition (1) is equal to:

$$H_{\text{tile},i} = -\frac{a_i}{a_i+b_i+\dots}\log_2\frac{a_i}{a_i+b_i+\dots} -$$

$$-\frac{b_i}{a_i+b_i+\dots}\log_2\frac{b_i}{a_i+b_i+\dots} - \dots. \qquad (3)$$

The set k_i of tile faces of the i-th sort is a multiset $\{(A_i, k_ia_i), (B_i, k_ib_i), ...\}$, which has the same combinatorial complexity $H_{\text{tile},i}$. Adding up the values of $H_{\text{tile},i}$ taking into account the relationship between the tiles according to the rule of strong additivity [13], we obtain:

$$H_{\text{tiling}} = \sum\nolimits_{i=1}^{n} w_i H_{\text{tile},i} + H\left(w_1, w_2, \dots, w_n\right), \qquad (4)$$

$$w_i = \frac{k_i\varphi_i}{\sum k_i\varphi_i}, \qquad (5)$$

$$H\left(w_1, w_2, \dots, w_n\right) = -\sum\nolimits_{i=1}^{n} w_i \log_2 w_i, \qquad (6)$$

where $\varphi_i = (a_i + b_i + ...)$ is the number of edges in the i-th tile, w_i are the weight factors that determine the contribution of $H_{\text{tile},i}$ to the overall complexity of the tiling H_{tiling}. The complexity calculated by formula (6) is an emergent parameter [30]. As follows from the properties of strong additivity [13], H_{tiling} is the combinatorial complexity of the multiset:

$$\Phi = \left\{\bigcup\nolimits_{i=1}^{n}\left\{\left(A_i, a_i\right), \left(B_i, b_i\right), \dots\right\}\right\}. \qquad (7)$$

Since each tiling face belongs to exactly two tiles, it is included in the multiset Φ twice, but this does not affect the size distribution of edges and does not distort the tiling stoichiometry. The unit of measurement of $H_{\text{tile},i}$ and H_{tiling} is bits/face. Multiplying H_{tiling} by the total number of edges in the tiling signature yields the total combinatorial complexity of the signature (bits/signature):

$$H_{\text{tiling,tot}} = H_{\text{tiling}} \sum k_i\varphi_i. \qquad (8)$$

Table 1 shows the Python code that allows calculating the H_{tiling} and $H_{\text{tiling,tot}}$ values for an arbitrarily large list of signatures written in a column of a file with a table in MS Excel format. The table column should not have a header, the signature format (2) is exactly the same as in the output of the ToposPro program [28] when constructing tiling, for example, "2[3^4] + [3^8]". By default, the program accesses the source data at C:\tilings\input.xlsx and creates a file in the same folder named input_with_entropies.xlsx, in which two columns (also without headers) with the calculated H_{tiling} and $H_{\text{tiling,tot}}$ values, respectively, are written next to the signature column.

The method was tested on an array of signatures of natural zeolite-type tilings presented in the accompanying materials to the pioneering work [31] and numbering 194 signatures at that time. A search was also conducted for rank (Spearman) and linear (Pearson) correlations of H_{tiling} and $H_{\text{tiling,tot}}$ with already known complexity indices of zeolite frameworks, in particular with the combinatorial complexity according to S.V. Krivovichev I_G (bit per framework vertex) and $I_{G,\text{tot}}$ (bit per reduced cell) (according to known data for 201 frameworks [32]), with the configuration complexity H_{conf} (bit

Table 1. Code for calculating the values of H_{tiling} and $H_{\text{tiling,tot}}$

```
import pandas as pd
from collections import Counter
import numpy as np

# Load the Excel file without headers
file_path = ‘C:/tilings/input.xlsx’
df = pd.read_excel(file_path, header=None)

# Function to transform the expression
def transform_expression(expression):
  def expand_term(term):
    term = term.strip()
    if term[0].isdigit():
      coefficient, inner = term.split(‘[‘, 1)
      inner = ‘[‘ + inner
      expanded_inner = expand_single_term(inner)
      expanded_inner_list = expanded_inner.strip(‘[]’).
      split(‘,’)
      return f”[{‘,’.join(expanded_inner_list *
     int(coefficient))}]”
    else:
      return expand_single_term(term)

  def expand_single_term(term):
    term = term.strip(‘[]’)
    if ‘.’ in term:
      factors = term.split(‘.’)
      expanded_factors = []
      for factor in factors:
       if ‘^’ in factor:
         base, exp = factor.split(‘^’)
         base = base.strip()
         exp = int(exp.strip())
         expanded_factors.extend([base] * exp)
    else:
      expanded_factors.append(factor.strip())
      return f”[{‘,’.join(expanded_factors)}]”
      elif ‘^’ in term:
      base, exp = term.split(‘^’)
      base = base.strip()
      exp = int(exp.strip())
      return f”[{‘,’.join([base] * exp)}]”
    else:
      return f”[{term.strip()}]”

 def modify_output(output):
   parts = output.split(‘+’)
   all_values = []
     for i, part in enumerate(parts):
       if part.startswith(‘[‘) and part.endswith(‘]’):
         inner_content = part[1:-1].split(‘,’)
```

```
        indexed_content = [f”{value.strip()}{i + 1}” for
        value in inner_content]
        all_values.extend(indexed_content)
     return f”[{‘,’.join(all_values)}]”

parts = expression.split(‘+’)
expanded_parts = [expand_term(part.strip()) for part in
parts]
final_output = ‘+’.join(expanded_parts)

return modify_output(final_output)

# Function to calculate Shannon entropy
def calculate_shannon_entropy(numbers):
   counts = Counter(numbers)
   total_count = sum(counts.values())
   probabilities = [count / total_count for count in
  counts.values()]
   entropy = -sum(p * np.log2(p) for p in probabilities
  if p > 0)
   return entropy

# Processing each expression and calculating entropy
def process_expression(expression):
   output = transform_expression(expression)
   output_numbers = list(map(int, output.strip(‘[]’).
  split(‘,’)))
   entropy = calculate_shannon_entropy(output_numbers)
   total_numbers = len(output_numbers)
   weighted_entropy = entropy * total_numbers
   return entropy, weighted_entropy

# Clean and sanitize the expression input if necessary
def clean_expression(expression):
   expression = expression.replace(‘]”’, ‘]’).replace(‘”’,
  ‘’).strip()
return expression

# Apply cleaning and process the expressions in the first
column
df[0] = df[0].apply(clean_expression)
df[[‘Entropy’, ‘Weighted Entropy’]] = df[0].apply(lambda x: pd.Series(process_expression(x)))

# Save the updated DataFrame to the same file, adding
the Entropy and Weighted Entropy columns
output_file_path = ‘C:/tilings/input_with_entropies.xlsx’
df.to_excel(output_file_path, index=False,
header=False)

print(f”Processed entropies saved to {output_file_path}”)
```

per degree of freedom) and $H_{\mathrm{conf,tot}}$ (bit per reduced cell) (according to previously obtained data for 242 frameworks [33]), as well as with the number of classes of symmetrically equivalent edges in the framework *e''*. To search for correlations, only those frameworks were used that were included in each of the three arrays of structures [31–33], with the exception of disordered frameworks (refcodes *BEA, *MRE, and *STO in the zeolite structure data bank [34]). Thus, data on 191 frameworks were analyzed.

DISCUSSION OF RESULTS

Table 2 presents the correlation coefficients of the complexity indices for zeolite-type frameworks and the corresponding natural tilings. These data show that all tested pairs of indices are positively correlated. For a random sample of $N = 191$, the *p*-significance level $p < 0.05$ corresponds to a correlation coefficient greater than 0.142; thus, all the correlations found are statistically significant. $H_{\mathrm{tiling,tot}}$ and *e''*; H_{tiling} are the weakest correlated; $H_{\mathrm{tiling,tot}}$ correlates moderately with other indices, including $I_{\mathrm{G,tot}}$ (Fig. 2a); the correlation of $H_{\mathrm{conf,tot}}$ with I_{G},tot (Fig. 2b) and $H_{\mathrm{conf,tot}}$ can be considered strong. The rank correlation coefficient in all cases, except for those in which one of the *e''* indices, turned out to be less than the linear correlation coefficient, which can easily be explained by the same logarithmic dependence of the corresponding indices on some, albeit different, structural parameters. As a comparative study of a series of structures of Hg-containing minerals and their synthetic analogues showed, all complexity indices used today in crystallography are correlated with each other to one degree or another [35].

The most complex tiling in the studied sample was the PAU framework ($H_{\mathrm{tiling,tot}} = 2928.3$ bits/signature), which also has the highest value of $I_{\mathrm{G,tot}} = 4763.5$ bits/cell. At the same time, an even more complex SFV framework ($I_{\mathrm{G,tot}} = 19557.6$ bits/cell) [32] was not included in the studied sample. The simplest tiling was the ABW framework ($H_{\mathrm{tiling,tot}} = 9.5$ bits/signature), while the simplest framework was sodalite (SOD, $I_{\mathrm{G,tot}} = 16.5$ bits/cell [32]). Tilings, like frameworks, can be classified by complexity class [36]: very simple (0–20 bits/signature), simple (20–100), medium complexity (100–500), complex (500–1000), and very complex (more than 1000). More than 80 % of tilings are simple or medium complexity (Fig. 3).

Natural tilings allow to construct migration routes of cations through framework voids and to predict ionic conductivity, therefore they are of great importance not only for zeolite-type frameworks, but also for “anti-zeolite” [37] cationic frameworks [38], as well as for mixed tetrahedral (*TT*) frameworks, for example, in the structures of boron [39] and beryllophosphates [40], and even for heteropolyhedral

Table 2. Correlations of complexity indices of zeolite frameworks and tilings

Index	Correlation type	I_{G}, bit/peak	H_{conf}, bit/d.f.	*e''*
H_{tiling}, bit/face	Linear	0.690	0.670	0.601
	Ranked	0.653	0.633	0.640
Index	**Correlation type**	**$I_{\mathrm{G,tot}}$, bit/cell**	**$H_{\mathrm{conf,tot}}$, bit/cell**	***e''***
$H_{\mathrm{tiling,tot}}$, bit/signature	Linear	0.886	0.827	0.426
	Ranked	0.779	0.774	0.615

Note: d.f. – degree of freedom.

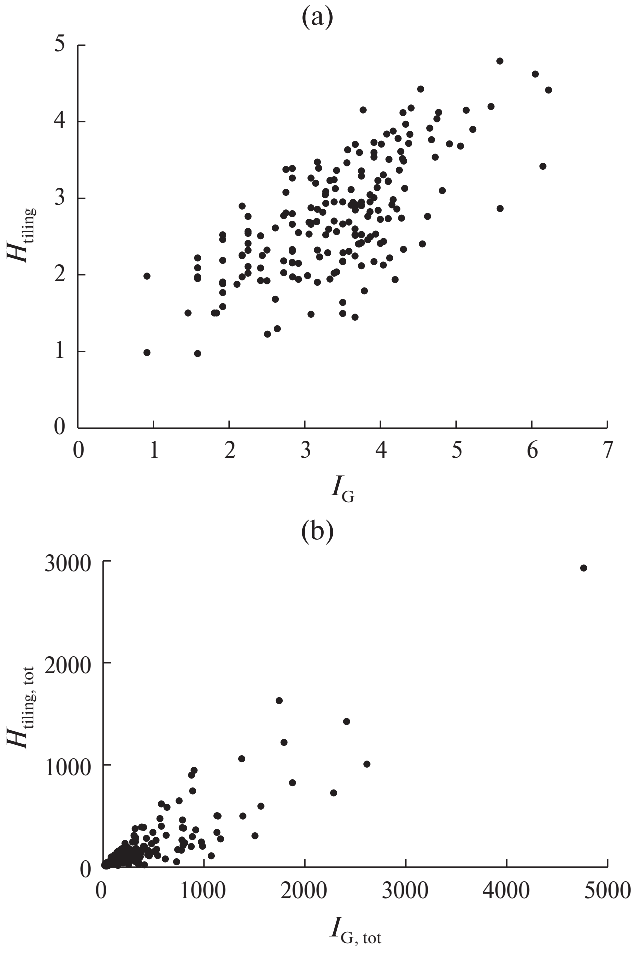

Fig. 2. Scatter diagram of H_{tiling} and I_{G} (a), $H_{\mathrm{tiling,tot}}$ and $I_{\mathrm{G,tot}}$ (b) for zeolite-type frameworks.

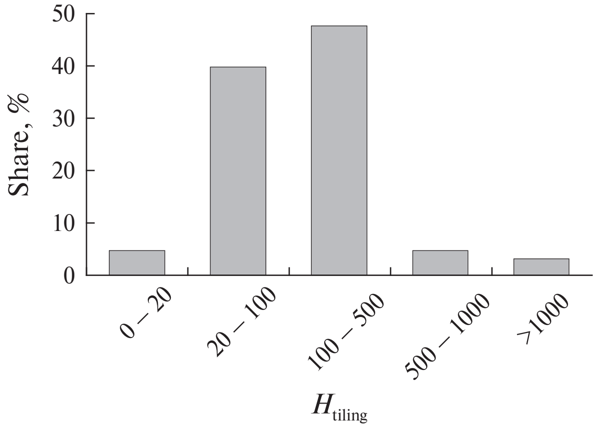

Fig. 3. The proportion of natural tilings of different complexity classes for zeolite-type frameworks.

MT frameworks, for example, in the structures of eudialyte [41], aluaudite [42], keldyshite [43], minerals of the columbite group [44], labuntsovite [45], in the structures of synthetic germanates [46], vanadates [47], molybdates [48]. Calculation of combinatorial complexity according to S.V. Krivovichev can easily calculate the complexity of any framework using the ToposPro software package [28], and additional indices, such as the Hornfeck configuration complexity, can be calculated using the Python application crystIT [15]. However, such calculations cannot be performed without a cif file with structural data. On the contrary, to calculate the combinatorial complexity of a natural tiling signature, only the tiling signature is required, not the structural data. Therefore, this complexity index can be easily used in expert systems that are not directly tied to structural data. A researcher who is not a crystallography specialist can calculate this index for a series of compounds simply from a list of signatures in a literature source.

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CONFLICT OF INTEREST

The author has no conflict of interest.

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