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UNIFORMITY DEGREE - INFORMATION ENTROPY - GRANULOMETRIC CHARACTERISTICS CORRELATIONS FOR POLYDISPERSE POWDERY MATERIALS

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În articol se prezintă utilizarea unui nou parametru statistic, gradul de uniformitate granulometrică pentru amestecurile poligranulare, calculat pe baza entropiei informaționale. Se evidențiază o serie de corelații între acest parametru, calculat pe baza entropiei informaționale şi diverse caracteristici ale unor distribuții granulometrice de tip Rosin-Rammler-Sperling. Dependențele deduse teoretic sunt verificate cu o serie de date experimentale referitoare la cimenturi industriale.

The paper presents the use of a new statistical parameter, the uniformity degree for powdery mixtures, calculated on the basis of information enthropy. Correlations between this parameter, calculated on the basis of information entropy, and different characteristics of Rosin-Rammler-Sperling type granulometric distributions are given. Theoretical dependencies are checked with experimental data on various types of industrial cements.

Keywords: powdery material, particle size distribution, characteristic diameter, size uniformity, information entropy

1. Introduction

Except the monograins materials, which are seldom used most of the materials in the silicates technology represents polygranular mixtures. They usually a certain diameter dispersion around a medium value and different particle size distributions (unimodal or multimodal). Depending on the specific nature of the operation from the technological flow in which the materials appear. Size uniformity of the powder material is lower when the frequencies of occurrence of different particle size classes have closer values. In some cases the uniformity degree of powder mixtures can be different even if their specific surface or mean diameter the same [1 - 4].

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It is obvious that processing two mixtures on the same technological flow will present certain differences (like in the heat transfer from the flue gases to the material during the reaction flow or in the milling bearing).

For illustration, in Fig. 1 is presented the case of two granular Rosin-Rammler-Sperling (RRS) type mixtures characterized by the same value of the characteristic diameter (x'), but with different degrees of granulometric uniformity ($n = tg\alpha$).

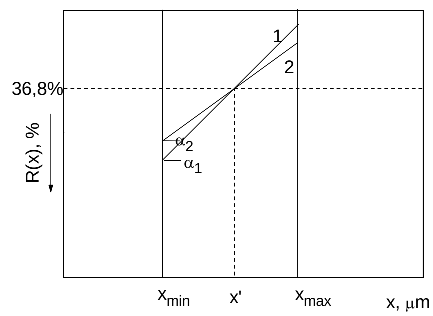


Fig. 1. Residue R(x) for two granular mixes of RRS type characterised by the same value of the characteristic diameter and the different values of the size uniformity

Fig. 1 shows that a powdery mixture can be characterized not only by means of diameters, but also making reference to the uniformity degree.

It must be also noticed the modality to estimate the granulometric uniformity degree becomes more difficult the case of polydisperse mixtures for which a distribution law applies differently over different intervals. Fig. 2 shows the case of a powder material for wich RRS law is valid on the intervals.

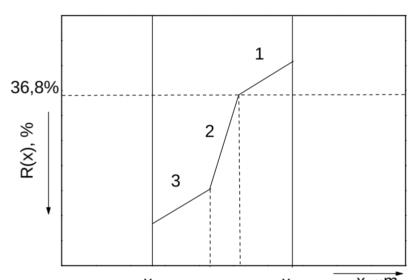


Fig. 2. Residue R(x) for a distribution of a polygranular mixture where RRS law applies on regions

It results that all factors that measures the dispersion proposed by the most common distribution laws can define the uniformity degree, and characterize a polygranular mixture only in certain situations, which are more or less in accord with the practical technological flow. Thus it is necessary to establish a general indicator for the quantitative evaluation of the polygranular mixtures uniformity, which can be used for any diameter and distribution model.

Statistics coefficients have been proposed for assessing the uniformity degree of a population [1 - 8]. Some of them are included in the defining law of the distribution type; like the uniformity index (n) of Rosin-Rammler-Sperling law. Others may be calculated from the definite expression of the distribution law.

For a polygranular mixture these coefficients have practical utility in defining the uniformity degree. Their effectiveness is real if size distribution is described by only one distribution function, and if the distribution law describes exactly the grain distribution on the full range of the characteristic granular spectrum.

However, in many practical cases during the operations of processing polygranulare mixtures these conditions are not met. Hence, the need to calculate the uniformity degree in a widespread manner, taking into account the natural approach based on the concept of order/disorder of the systems. To achieve this aim, in this paper correlations between the uniformity degree and different granulometric characteristics are established.

2. Evaluation of the polygranulate materials uniformity degree

In a general approach, a system is composed of many structural units. To these are associated existential probabilities (frequencies). The organizational/disorganizational degree of the system can be determined on the basis of some indicators provided by informational statistics [9 - 11]: Shannon informational entropy (H) and Onicescu informational energy (E_i). The most elaborated from the theoretical point of view is the *informational entropy* [9].

A granular material can be considered as a system in a systemic approach. Constructive elements are represented by the granulometric classes characterizing this material.

For the same diameter (x_{min} - minimum diameter, respectively x_{max} - maximum diameter) and the same class interval (Δx), the occurrence frequencies (probability of existence) of granulometric classes can be:

- relatively close in value, showing an advanced disorder from point entropic of view; this material is strongly nonuniform;

- strong by disproportionated, many particle size classes having lower values of the frequencies; in this case the system is strongly ordered, having a high of uniformity degree.

For a polygranular material, the distribution of particle size classes is experimentally establisheds, by classifications or evaluated on the basis of laws of distribution. In both cases uniformity degree can be assessed by calculating the informational entropy.

Shannon informational entropy (H) for discrete random variables is given by (1), where n is the number of states (events, structural units, granulometric classes of the system); f_i - existential frequency for this state i, consequently $0 \le f_i \le 1$, $i = \overline{1,n}$, $\sum_{i=1}^{n} f_i = 1$:

$$H(x) = -\sum_{i=1}^{n} f_i \cdot \log f_i \tag{1}$$

For a defined system (organized, ideally uniform) the enthalpy is zero (H = 0) because $f_1 = f_2 = \ldots = f_{n-1} = 0$, and $f_n = 1$. For a system where all states are echiprobable (disorganized, ideally nonuniform) H = log n, because $f_1 = f_2 = \ldots = f_n = 0$.

The lowest limit of the information entropy corresponds to a uniform system (monogranular material), and the upper limit depends on the number of possible states (n), in particular on the number of particle size fractions. The *standard entropy (H_n)* is defined (2) to eliminate this deficiency; H_n take values between [0,1]:

$$H_n = \frac{H}{\log n} \tag{2}$$

Polygranular dispersed mixture is ideal (nonuniform) when $H_n(x) = 1$. Therefore, the occurence of each particle size fraction is the same. If the material is monogranular $H_n(x) = 0$ (ideally uniform).

Shannon information entropy of a continuous variable is given by (3).

The amount of information provided by certain event (experiment), for a continuous random variable x, with distribution density $p(x) \ge 0$ has $\int_R p(x)dx = 1$:

$$H(x) = -\int_R p(x) \cdot \log(x)dx \tag{3}$$

For a powder material the distribution density has a specific law related to the residue according to grain diameter, x.

Herein, the informational energy (E_i) proposed by Onicescu [11, 12] which was to define the uniformity of dispersed granular mixtures [3, 13, 14]. However, our approach is a determination of the possibility of using the simpler Shannon distribution for calculation of uniformity degree.

For each density distribution (f(x)) the normalization condition checked (4):

$$\int_{x_{min}}^{x_{max}} f(x)d(x) = 1 \tag{4}$$

When x is the particle diameter and the disperse granular mixture obey the law Rosin - Rammler - Sperling, can be written as equation (5), where: n is uniformity index, and x' - the specific diameter (µm):

$$R(x) = e^{-\left(\frac{x}{x'}\right)^n} \tag{5}$$

The repartition density RRS was determined using with equations 6 and 7 taking into account the residue R(x), and the passing T(x) (% mass).

$$T(x) = 1 - R(x) = e^{-\left(\frac{x}{x'}\right)^n} \tag{6}$$

$$f(x) = \frac{dT(x)}{dx} = \frac{n}{x'}\left(\frac{x}{x'}\right)^{n-1} e^{-\left(\frac{x}{x'}\right)^n} \tag{7}$$

Based on (5) the mean diameter and diameter for the 0.5 passage-residue can be define (8, 9).

$$\bar{x} = \int_{x_{min}}^{x_{max}} x \cdot f(x)dx \tag{8}$$

$$0,5 = e^{-\left(\frac{x_{50}}{x'}\right)^n} \qquad x_{50} = x' \cdot (\ln 2)^{1/n} \tag{9}$$

The integral equation 4 is preferred for calculating the entropy when the particle size distributions for powder mixtures are described quantitatively by distribution RRS (10).

$$H = -\int_{x_{min}}^{x_{max}} f(x) \cdot log[f(x)]dx \tag{10}$$

Based on this equation we tried to find possible dependencies between H and various characteristics of granular mixtures when x_{min} = 1 mm and where x_{max} takes different values in the range 80 - 200 μm.

For the calculation of the specific surface of the powder mixtures the equation (11) was used, where ρ is the cement density:

$$S = \frac{6}{\rho \cdot x'} \int_{x_{min}}^{x_{max}} \frac{1}{x} f(x)dx \tag{11}$$

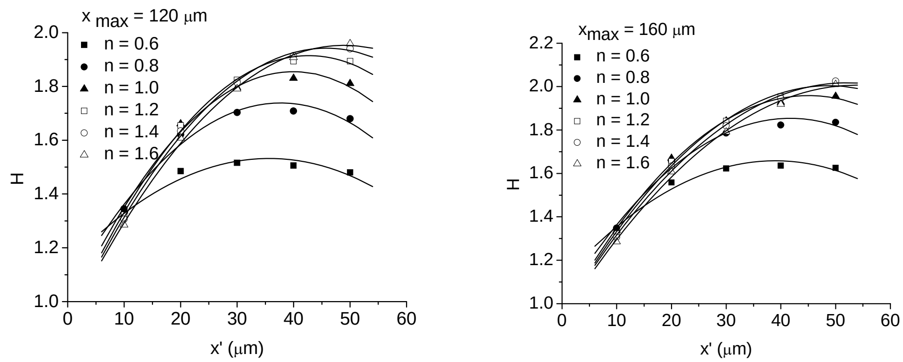
3. Ressults and disscution

In this paper are presented the theoretical results, derived from the numerical simulations which show the existence of dependencies between information entropy and various granular characteristics of powder mixtures with RRS distribution. A series of 18 granular cements wase considered for the known density (ρ) and RRS distribution curves parameters x' şi n (Table 1). In Figs. 3, 4, 5, 7 and 9 are presented some results calculated for several industrial cements [1, 2, 4, 11]. The statistical dependencies between the same statistical quantities obtained for these industriale cements are presented in Figs. 6, 8 and 10.

Table 1

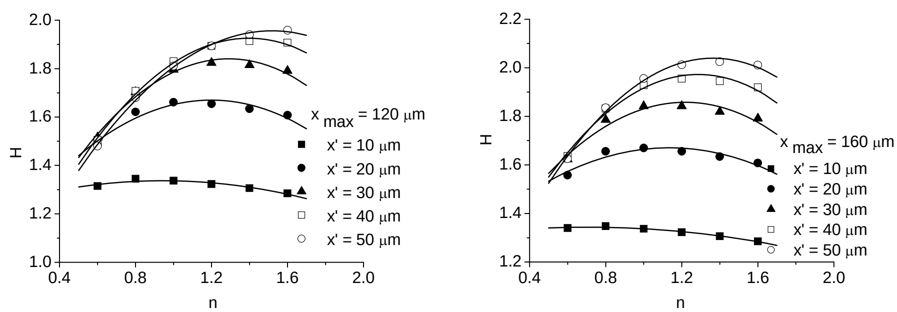
Granulometric characteristics şi densities of several industrial cements [1, 2, 5, 12]

Cement	C_1	C_2	C_3	C_4	C_5	C_6	C_7	C_8	C_9
Density, g/cm^3	3.15	3.10	3.03	3.18	3.10	3.03	3.15	3.10	3.05
x', μm	31.3	24.8	19.6	21.0	16.0	11.3	13.0	10.8	8.6
n	0.99	0.90	0.80	1.11	0.99	0.84	1.14	1.02	0.92
Cement	C_{10}	C_{11}	C_{12}	C_{13}	C_{14}	C_{15}	C_{16}	C_{17}	C_{18}
Density, g/cm^3	3.12	3.05	3.00	3.04	2.98	2.92	3.04	3.01	2.96
x', μm	27.6	22.7	15.3	26.5	18,8	14.5	19.7	16.5	15.0
n	1.09	0.94	0.83	1.12	0.98	0.85	1.12	0.99	0.82



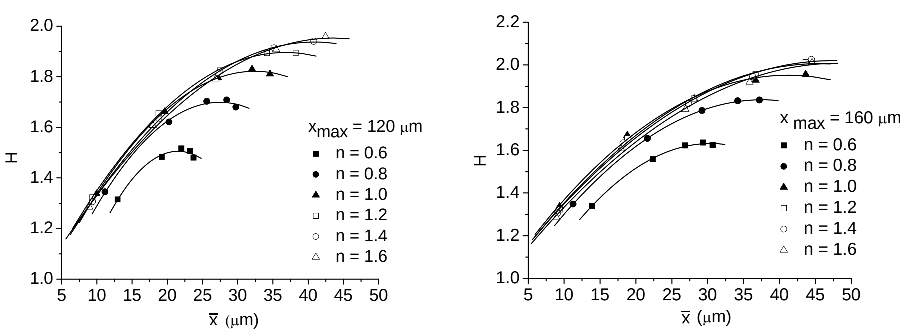
(a) (b)

Fig. 3. Variation of the information entropy with the characteristic diameter, x'
($H = a + b\,x' + c\,(x')^2$)



(a) (b)

Fig. 4. Variation of the information entropy with the uniformity index, n
($H = a + b\,n + c\,n^2$)



(a) (b)

Fig. 5. Variation of the information entropy with the mean diameter, $\bar{x}$
($H = a + b\,\bar{x} + c\,\bar{x}^2$)

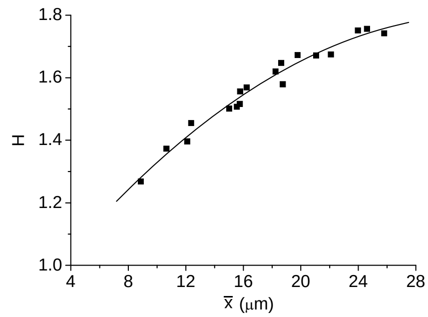
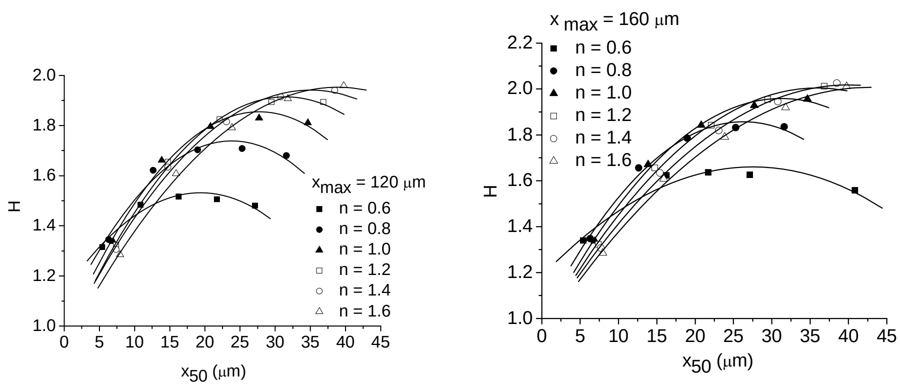


Fig. 6. Variation of the information entropy with the mean diameter, $\bar{x}$, the 18 cements

$(H = a + b\,\bar{x} + c\,\bar{x}^2)$



(a) (b)

Fig. 7. Variation of the information entropy with x_{50} diameter

$(H = a + b\,x_{50} + c\,x_{50}^2)$

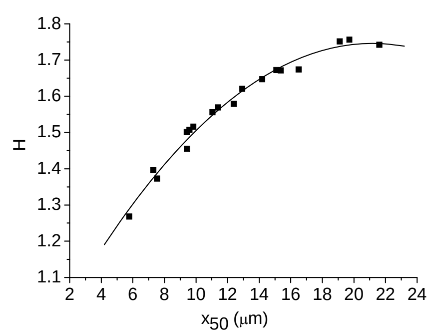
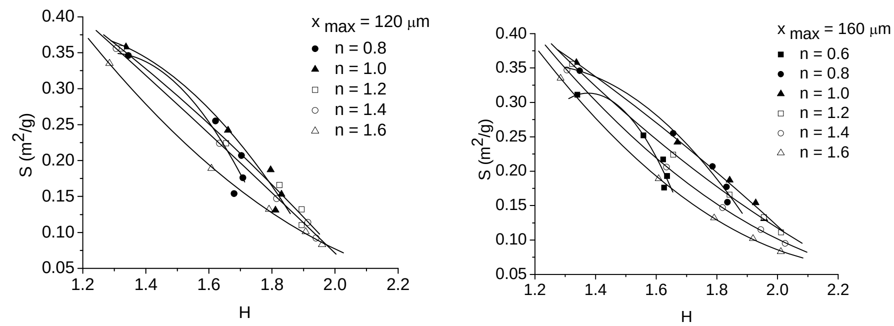


Fig. 8. Variation of the information entropy with with x_{50} diameter for the 18 cements

$(H = a + b\,x_{50} + c\,x_{50}^2)$



(a) (b)

Fig. 9. Variation of the specific surface with the information entropy
($S = a + b\,H + c\,H^2$)

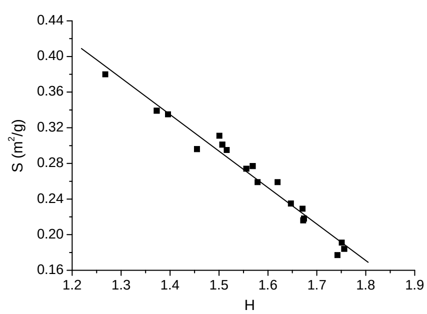


Fig. 10. Variation of the specific surface with the information entropy the 18 cements
($S = a + b\,H$)

From Figs. 3, 5, 6 it can be observed the information entropy (H) correlations with dimensional parameters (x', $\bar{x}$, and x_{50}). In all cases increasing values for maximum diameter and also for n leads to an increase in entropy, hence the disorder degree, and the heterogeneity for powder material. This is expected because likewise increases the number of granulometric classes, consequently spreading system.

The dependencies H - x', H - $\bar{x}$ and H - x_{50} are represented for x constant and a given uniformity index; graphs shows a monotonic variation for certain values of the considered granulometric parameters. Good results are obtained for parabolic function. A simple relationship of dependency was considered, but with high regression coefficient (R). Table 2 shows that in all cases the proposed dependencies are characterized by values of R above 0.90.

Similar conclusions are obtained from the correlation H - n in the distribution RRS when x_{max} = constant and x' = constant (Fig. 4);

The specific surface of a powder mixture is greater when non-uniformity degree, respectively entropy, have lower values (for x_{max} = constant şi n = constant), as shown in Fig. 10). In this case the dependence is also stronger, being given by the equation of a straight (in the most cases) line.

The results calculated the basis of the experimental data for 18 cements have shown the same type dependency as for the simulated powder mixtures.

Table 2

Equation coefficients (a, b, c) for the fitting curves shown in Figs. 3 -10, and their correlation coefficients (R)

n	x_{max}, μm	a	b	c	R	Fig. number
0.6		1.138	0.022	$-3.092 \cdot 10^{-4}$	0.941	
0.8		1.041	0.036	$-4.892 \cdot 10^{-4}$	0.971	
1.0	120	0.958	0.044	$-5.621 \cdot 10^{-4}$	0.989	Fig. 3a
1.2		0.922	0.046	$-5.45 \cdot 10^{-4}$	0.996	
1.4		0.914	0.044	$-4.9 \cdot 10^{-4}$	0.997	
1.6		0.911	0.042	$-4.35 \cdot 10^{-4}$	0.997	
0.6		1.107	0.028	$-3.628 \cdot 10^{-4}$	0.971	
0.8		1.004	0.040	$-4.892 \cdot 10^{-4}$	0.985	
1.0	160	0.949	0.044	$-4.971 \cdot 10^{-4}$	0.995	Fig. 3b
1.2		0.942	0.043	$-4.464 \cdot 10^{-4}$	0.997	
1.4		0.943	0.041	$-3.964 \cdot 10^{-4}$	0.996	
1.6		0.935	0.039	$-3.692 \cdot 10^{-4}$	0.996	
10		1.220	0.245	-0.129	0.906	
20		0.994	1.127	-0.470	0.917	
30	120	0.753	1.685	-0.653	0.981	Fig. 4a
40		0.661	1.810	-0.648	0.995	
50		0.674	1.688	-0.555	0.998	
10		1.305	0.106	-0.075	0.985	
20		1.230	0.774	-0.341	0.852	
30	160	1.015	1.382	- 0.567	0.938	Fig. 4b
40		0.840	1.761	-0.684	0.980	
50		0.754	1.883	-0.690	0.993	
0.6		0.298	0.112	-0.002	0.984	
0.8		0.686	0.073	-0.001	0.995	
1.0	120	0.809	0.062	$-9.629 \cdot 10^{-4}$	0.999	Fig. 5a
1.2		0.866	0.056	$-7.623 \cdot 10^{-4}$	0.999	
1.4		0.878	0.051	$-6.328 \cdot 10^{-4}$	0.999	
1.6		0.907	0.047	$-5.492 \cdot 10^{-4}$	0.998	
0.6		0.641	0.065	-0.001	0.998	
0.8		0.842	0.052	$-7.019 \cdot 10^{-4}$	0.999	
1.0	160	0.933	0.049	$-6.011 \cdot 10^{-4}$	0.997	Fig. 5b
1.2		0.934	0.047	$-5.188 \cdot 10^{-4}$	0.997	
1.4		0.943	0.045	$-4.766 \cdot 10^{-4}$	0.996	
1.6		0.936	0.044	$-4.582 \cdot 10^{-4}$	0.996	
		0.823	0.059	$-9.094 \cdot 10^{-4}$	0.973	Fig. 6
0.6		1.138	0.040	-0.001	0.943	
0.8		1.041	0.058	-0.001	0.971	

1.0 1.2 1.4 1.6	120	0.958 0.922 0.942 0.911	0.064 0.063 0.057 0.053	-0.001 -0.001 $-8.143 \cdot 10^{-4}$ $- 6.880 \cdot 10^{-4}$	0.989 0.996 0.995 0.997	Fig. 7a
0.6 0.8 1.0 1.2 1.4 1.6	160	1.185 0.999 0.949 0.941 0.943 0.935	0.034 0.065 0.064 0.059 0.053 0.050	$-6.279 \cdot 10^{-4}$ -0.001 -0.001 $-8.226 \cdot 10^{-4}$ $-6.694 \cdot 10^{-4}$ $-5.840 \cdot 10^{-4}$	0.948 0.988 0.995 0.997 0.996 0.996	Fig. 7b
0.6 0.8 1.0 1.2 1.4 1.6	120	0.883 - 1.323 - 0.191 0.652 0.842 1.342	0.081 2.591 1.026 -0.098 -0.347 -1.056	-0.001 1.003 -0.460 -0.095 -0.019 0.212	0.985 0.875 0.964 0.990 0.998 0.999	Fig. 8 Fig. 9a
0.6 0.8 1.0 1.2 1.4 1.6	160	- 3.190 - 0.319 0.651 1.101 1.331 1.438	5.094 1.122 -0.127 -0.703 -1.015 -1.187	-1.851 -0.466 -0.68 -0.105 0.200 0.255	0.933 0.984 0.996 0.999 0.999 0.999	Fig. 9b
		0.908	-0.409		-0.985	Fig. 10

4. Conclusion

The correlations between the uniformity degree and different granulometric characteristics calculated for a Rosin-Rammler-Sperling type has show that informational entropy can be an overall and general statistical indicator for the evaluation ofe particle size uniformit powdery mixtures.

The informational entropy can be used as basic tehnological parameter in a series of operations which involves suspensions hydrodynamics, mixing or materials grinding.

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