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CHARACTERIZATION OF GRANULATED AND POWDERED SOLIDS

BY

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Abstract. The objective of this study is to investigate the physical properties of urea, granules and powder, for characterizing the material behavior during storage, handling and processing. The particle size distribution, bulk density, Carr’s Index, Hausner ratio, porosity and specific surface area of the bed, angle of repose and flow rate were determined for granular and powdered urea. Also, the influence of the particle size on the physical and flowing properties was investigated. Experimental results are in good concordance with those of the literature.

Key words: particle size distribution, bulk density, angle of repose, bed porosity.

1. Introduction

Physical properties of granular and powdered solids plays a significant role in their storage, handling and processing with application in agriculture, food processing, chemical industry (processing of polymers, fertilizers), construction materials industry (glass, ceramics, cement), varnish and paint industry, metallurgy, cosmetics, pharmaceuticals (Barbosa-Canovas *et al.*,

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2005; Degouet *et al*., 2007; Ganesan *et al*., 2008; Holdich, 2002; Schulze, 2008). Also, physical and flow properties of solid-fluid systems are extremely important for research, development and quality control for several operations and are essential to design efficient and economic equipments.

Urea is mainly used in agriculture as fertilizer, but has other applications in the chemical, pharmaceutical and food industries and in cosmetics. Urea is used in the form of granules, powder and solutions and it is important to know its physical properties in various presentations.

This paper presents some physical properties of urea granules and powder, experimentally determined, their influence on the flow characteristics of material.

2. Experimental

2.1. Materials

The following materials were used in this study: urea granules (from Azomureş plants), four types of granules with different sizes (1.25 mm, 1.8 mm, 2.25 mm and 2.85 mm) and urea powder obtained from granules by grinding.

2.2. Methods

Main features of granular solids and powders include particle size, particle size distribution, real density, bulk density and tapped density, porosity and specific surface area of the bed, angle of repose, flow rate and so on.

Particle size distribution. Granular or powdered substances are often made up of elements with various shape and size. Particle size and particle size distribution has a direct influence on material properties such as: reactivity or dissolution rate (*e.g*. catalysts, tablets), stability in suspension (*e.g*. sediments, paints), texture and feel (*e.g*. food ingredients), appearance (*e.g*. powder coatings and inks), flowability and handling (*e.g*. granules), packing density and porosity (*e.g*. ceramics) (Benkovic *et al*., 2009; Brittain, 2001; Degouet *et al*., 2007; Manickam *et al*., 2011; Rawle, 1993).

Particle size distribution may be determined by several methods: sieving, optical and electronic microscopy, sedimentation, gas adsorption, Fraunhofer diffraction and so on.

The size distribution of the granular and powdered solids was determined by sieving, the simplest method for particle size analysis based on the size of the particles and independent of other particle properties (Fan *et al*., 1998; Folk, 1968; Holdich, 2002; Rawle, 1993).

Two sets of sieves were used with mesh size of 3.35–0.5 mm, for the initial material, and 1.6-0.05 mm, for crushed urea, according with International test sieve series (*** 2000).

Sieves were arranged vertically in order of decreasing mesh size in a vibration device (Fig. 1). The sample of the material was weighed and placed on the top sieve and the nest of test sieves was mechanically vibrated 10 min to ensure a good separation. After separation, the fractions retained on all sieves were weighed one by one.

The arithmetic mean of three repeated measurements was taken into account in each case.

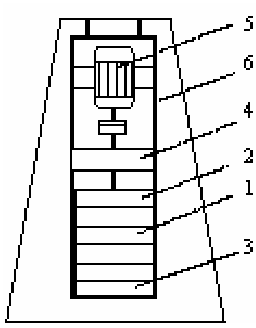


Fig. 1 − Screening device scheme.
1 - sieve set, 2 - cover, 3 - pan, 4 - vibration device, 5 - electric motor, 6 - support.

Bulk and tapped densities. The bulk density of granular and powdered solids is important to the design of the equipment for processing, handling and storage and can be determined by various methods (Agubata *et al*., 2012; Apeji *et al*., 2010; *** 2009; Hausner, 1967): by measuring the volume of a known mass of sample, by measuring the mass of a known volume of powder into a cup or a measuring vessel. The bulk density of a granular urea and powder were determined by measuring the volume of a known mass of material in a graduated cylinder.

The tapped density was obtained by mechanical means by tapping the cylinder containing the sample on a platform until the volume occupied by the solid remains constant.

The bulk and tapped densities were calculated using eqs. (1) and (2).

$$\rho_L = \frac{m}{V} \tag{1}$$

$$\rho_T = \frac{m}{V_T} \tag{2}$$

where: ρ_L and ρ_T are the bulk and tapped densities, [kg·m^{-3}]; m − the weight of the material, [kg]; V and V_T − the volumes of the material and tapped material respectively, [m^3].

Densities for urea were determined for the original material (unclassified), four types of urea granules with different particle size (1.25 mm, 1.8 mm, 2.25 mm and 2.8 mm) and urea powder.

To determine the bulk density and the tapped density of the material were used a graduated cylinder and a balance type Partner WPS 1200/C/2. In each case there were determined the amount of material in the cylinder, the volume occupied by the material before and after compaction. The mean of three determinations was recorded.

For processing the material in fixed bed, porosity and specific surface area have a particular importance.

Porosity of the bed. Material porosity is the fraction of the bulk volume of the sample that is occupied by void space (Geankoplis, 2003) and for granular and powder urea, was calculated from the true and tapped densities:

$$\varepsilon = 1 - \frac{\rho_T}{\rho} \tag{3}$$

where: ε is the bed porosity, [%]; ρ and ρ_T - the true and the tapped densities, [kg m^{-3}]. The true density of the material (1330 kg·m^{-3}) was determined from literature (Meessen, 2012).

Specific surface area. The specific surface area is defined as the total surface area of material per unit of mass solid or bulk volume.

In this study, only the external specific surface based on volume was considered. Because urea particles are nearly spherical shape, the external specific surface per unit bed volume was calculated from equation:

$$a = \frac{6 \cdot (1-\varepsilon)}{d} \tag{4}$$

where: a is the specific surface area, [m^2 m^{-3}]; ε − the external porosity, d − the spherical particle diameter [m].

Flowability of the granular or powdery material can be characterized by several properties: *i.e.* Carr’s index, Hausner ratio, angle of repose, flow rate (Carr, 1965; Holdich, 2002; Schulze, 2008).

Hausner ratio and Carr’s Index may be calculated from eqs. (5) and (6) (Barbosa-Canovas *et al*., 2005; Carr, 1965; Ganesan *et al*., 2009; Hausner, 1967):

$$\mathrm{HR} = \frac{\rho_T}{\rho_L} \tag{5}$$

$$\mathrm{CI} = \frac{\rho_T - \rho_L}{\rho_T} \cdot 100 \tag{6}$$

According with Hausner and others (Apeji *et al*., 2010; Azubuike *et al*., 2012; Hausner, 1967) the Hausner ratio less than 1.25 indicates a good flowability while a value of 1.25 or higher suggests a poor flow of the material. Also, the Carr Index values are used to characterize the flow properties of the material (excellent and good flow properties for Carr's index less 16 and fair or poor flowability for values greater 18 (Carr, 1965).

Angle of repose. Angle of repose is defined as the angle between the horizontal and the slope of a heap of granular material and is strongly affected by size and shape of the particles, degree of sorting of the material, measuring method (Miller *et al*., 1999).

The measurement of the static angle of repose was carried out using a draining method (Barbosa-Canovas *et al*., 2005; Carr, 1965).

The device consists of a cylindrical stainless steel container with a funnel at the bottom. Inside the cylinder is mounted a horizontal disc with adjustable position (Fig. 2).

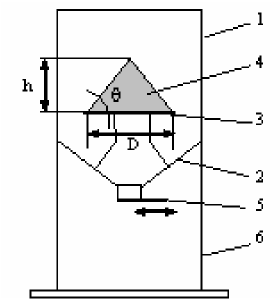


Fig. 2 − The device for determining the drained angle of repose.
1 - cylindrical container; 2 - funnel; 3 - disc; 4 - heap of material; 5 - flap; 6 - support.

Granular or powdery material is poured into the container, excess material is removed through the containers funnel and on the disc remains a heap of material. The angle of repose was calculated from equation:

$$\theta = \mathrm{tg}^{-1}\frac{2 \cdot h}{D} \tag{7}$$

where: θ is the angle of repose, [º]; h − the height of the conical heap of the material remained on the disc, [m]; D − the diameter of the disc, [m].

Angles of up to 35º indicate reasonable flow potential of the solid, whereas those greater than 45º exhibit poor flow (Apeji *et al*., 2010; Azubuike *et al*., 2012; Carr, 1967).

Determinations were done in triplicate and the mean was taken.

Flow rate. Flow rate was determined in the same apparatus as the angle of repose using the funnel method (Agubata, 2012).

A weighed amount of the material to be analyzed was poured into the recipient. The material was allowed to flow through the orifice of the funnel and the time required for complete emptying of the container was noted. The flow rate was calculated using the eq. (8):

$$v_m = \frac{m}{t} \quad (8)$$

where: v_m is the flow rate of the material, [m·s^{-1}]; m − the weight of solid [g]; t − the time for emptying the recipe [s].

3. Results and Discussions

The results of the study are presented in tables and figures.

Particle size distribution. With the experimental data were calculated the fractions retained on each of sieves used in the test as percentages of the starting sample weight, the diameter of each particle size fraction (as arithmetical mean of the dimensions of the two sieves that define the fraction), the cumulative percentages of oversize and undersize materials (Table 1).

Table 1
Data of Size Distribution

Size range [mm]	Size class [mm]	Mass frequency [%]	Cumulative oversize [%]	Cumulative undersize [%]
Granular urea				
3.35 – 3.15	3.250	0.914727	0.914727	99.08527
3.15 – 2.5	2.825	7.309143	8.22387	91.77613
2.5 – 2.0	2.250	83.89474	92.11861	7.881389
2.0 – 1.6	1.800	0.528894	92.64751	7.352495
1.6 – 1.25	1.425	5.93055	98.57806	1.421945
1.25 – 0.5	0.875	1.421945	100	0
Urea powder				
1.6 – 1.25	1.425	1.566408	1.566408	98.43359
1.25 – 0.5	0.875	10.60945	12.17586	87.82414
0.5 – 0.25	0.375	16.70396	28.87982	71.12018
0.25 – 0.1	0.175	64.03844	92.91826	7.081743
0.1- pan	0.050	7.081743	100	0

The results were used to construct the histogram (the mass fraction percent retained as a function of sieves size, Fig. 3) and the cumulative distribution curves (the cumulative percentages of oversize and undersize material respectively as a function of particles size, Fig. 4).

Granular urea has a wide particle size distribution, while powdered urea has a narrow particle size distribution.

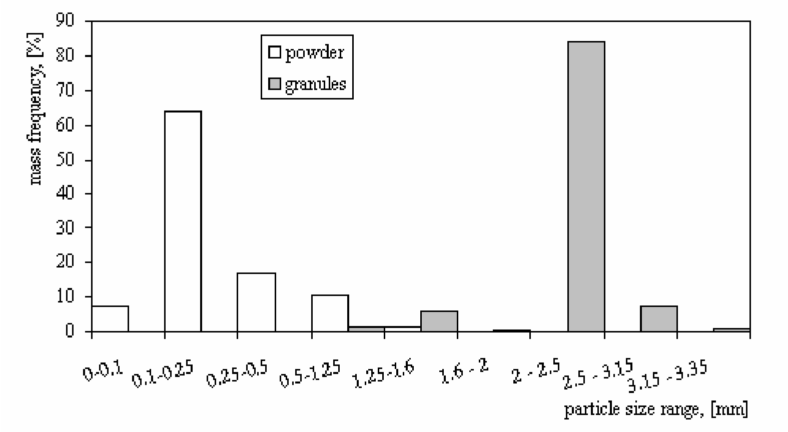


Fig. 3 − Particle size presentations in normal distributions for granular and powdered urea: histograms of frequency *versus* range size.

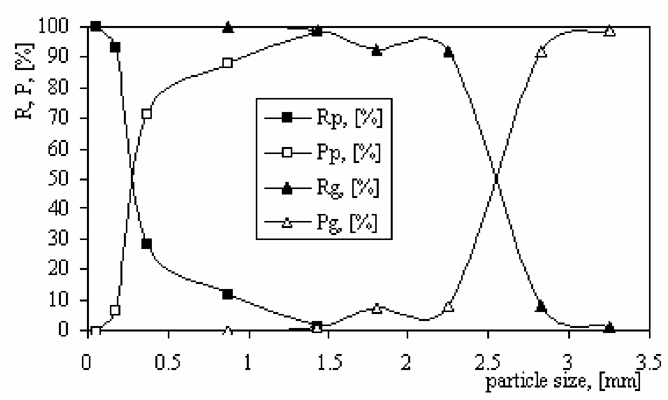


Fig. 4 − Particle size presentations in normal distributions for granular (g) and powdered (p) urea: the cumulative distributions (R – oversize, P – undersize).

The particle sizes of the studied materials suggests that the granular urea has excellent flow properties while the powder urea has fair flow properties - is considered that the materials with particle sizes larger than 0.2 mm are free flow, while fine powders are poor flow (Teunou *et al*., 1999).

For a polydisperse system, according to industrial applications, several values of mean diameter can be defined. These values can be calculated or determined from the graphs. For example:

− the *mode* of the material (the size corresponding to a highest mass frequency) can be determined from the histogram or from the frequency distribution,

− the *median* size of the material from the cumulative curves (the value corresponding to 50% oversize and undersize respectively),

− the *mass mean diameter* based on the mass fraction of the sample is calculated using the relation (Fan *et al*., 1998; Rawle, 1993):

$$d_m = \sum_{i=1}^{n} x_i \cdot d_i \tag{9}$$

where: d_m is the mass mean diameter of material, [m]; x_i − the mass fraction of particles having a diameter equal to d_i.

The mean, median and mode depend on the symmetry of the distribution and can be different. From experiments and calculated results and from graphs were determined the characteristic size of the materials (Table 3).

Table 3
Characteristic Size of the Materials

Characteristic size	Granular urea	Powder urea	Source
mass mean diameter, [mm]	2.23	0.293	eq. 9
mode, [mm]	2.25	0.175	Histogram
median diameter, [mm]	2.54	0.275	Cumulative distribution

Bulk and tapped density. Bulk and tapped densities for all materials studied were calculated with the eqs. (1) and (2) and results presented in Table 4 and Fig. 5 are in good concordance with the values from literature (Manickam *et al*., 2011; *** 2006).

Bulk and tapped densities variation as function as particle size are plotted in Fig. 5. As shown, both densities decrease with increasing particle size.

Bed porosity and specific surface area. The bed porosity and the specific surface area for granular urea were calculated with eqs. (3) and (4) and Fig. 6 present the influence of the particle size on these properties. The porosity of the granular urea increased with the particle sizes increased because the large particles do not pack as closely as small particles. The specific surface area of all particles in a unit volume decreased with the increase in the particle size.

Table 4
Characteristics of the Materials Tested

Properties	Urea	
	Unclassified granule	Powder
Bulk density, [kg·m^{-3}]	753.448	584.198
Tapped density, [kg·m^{-3}]	765.621	747.523
Carr's index, [%]	1.59	21.849
Hausner ratio	1.016	1.279
Angle of repose, [º]	32.85	45.59
Flow rate, [g·s^{-1}]	59.46	11.33

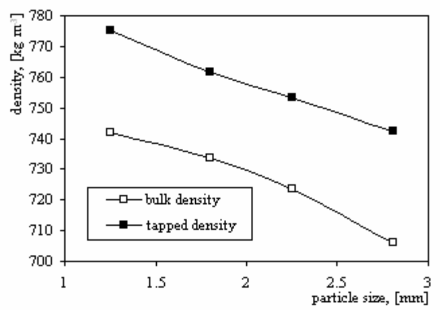
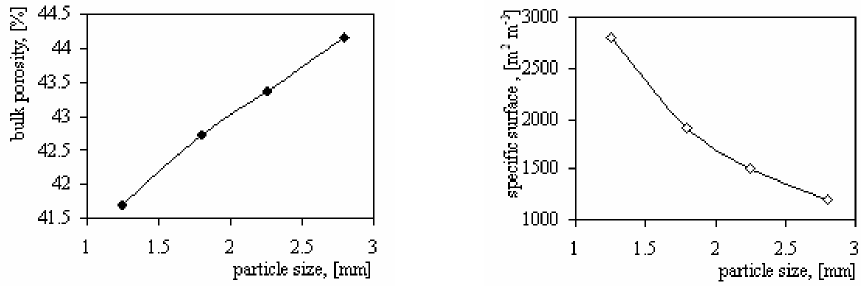


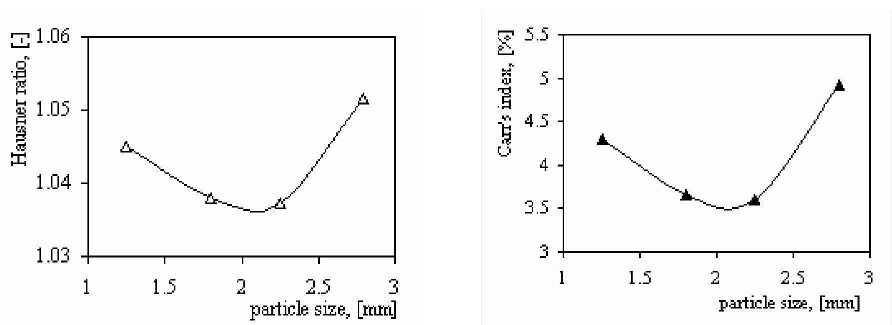
Fig. 5 − Effect of urea particle size on bulk and tapped density.



a *b*

Fig. 6 − Effect of urea particle size on bed porosity (*a*) and specific surface area (*b*).

The Carr's index and Hausner ratio calculated for urea granules and powder with eqs. (5) and (6) are presented in Table 4 and suggest a good flowability for granular urea (values for Carr's index less than 16 and Hausner ration less than 1.25) and a poor flowability for powdered urea (Carr's index value in range 23-28 and Hausner ratio greater than 1.25). Also, the values of the Carr's index and Hausner ratio shows that particles with sizes in the range 1.5 - 2.5 mm have the best flow properties (Fig. 7).



a *b*

Fig. 7 − Effect of urea particle size on Hausner ratio (*a*) and Carr's index (*b*).

Angle of repose and flow rate. The angle of repose of less than 35° indicate the free flow for all kinds of granular urea while the angle of repose for the powder has a value greater than 45° and shows a poor flowing (Table 4). The experimental values of the flow rate are in concordance with the angle of repose values (Table 4).

Figs. 8 present the particle size influence on angle of repose and flow rate of the material. The angle of repose of the sorted materials increases with decreasing particle size because smaller particles tend to adhere much more strongly to each other. The flow rate decrease with decreasing particle size.

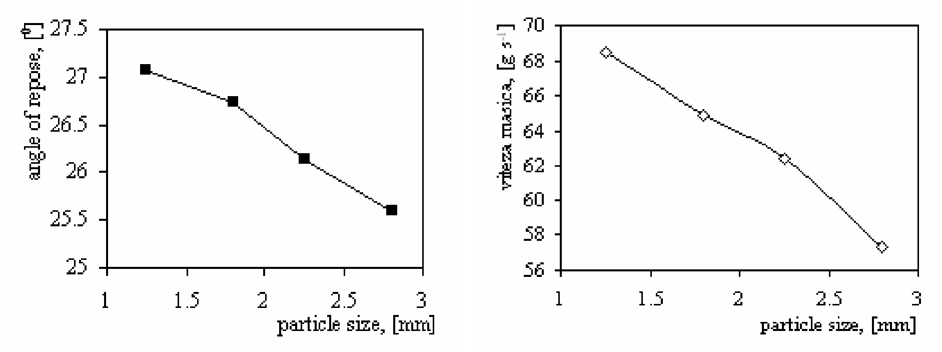


Fig. 8 − Effect of urea particle size on angle of repose (*a*) and flow rate (*b*).

4. Conclusions

Physical and flow properties of urea, granules and powder, were investigated experimentally: bulk and tapped densities, Carr's index, Hausner ratio, angle of repose, flow rate, bed porosity and specific surface area.

When the particle size of urea was reduced by milling, the granulometric distribution and mean diameter of the material was modified. The particle size distribution was reduced for powdered urea in comparison with granular urea. Also, the mass mean diameter, mode and median diameter have smaller values for urea powder.

Carr's index, Hausner ratio, angle of repose and flow rate suggest a good flowability for granular urea and a poor flow for powdered urea.

For the granular urea, bulk density, tapped density, specific surface area, angle of repose and flow rate varies inversely with size of particles while bed porosity varies directly with size of particles.

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CARACTERIZAREA MATERIALELOR GRANULARE ŞI PULVERULENTE

(Rezumat)

Lucrarea a urmărit determinarea proprietăților fizice şi de curgere ale ureei în formă granulară şi pulverulentă. A fost realizată analiza granulometrică a ureei înainte şi după măcinare şi au fost determinate moda, mediana şi diametrul mediu al materialului din histograme, curbele de distribuție cumulativă şi relații de calcul. Au fost determinate proprietățile fizice şi de curgere pentru materialul granular, materialul măcinat şi patru clase granulometrice. Rezultatele experimentale au evidențiat dependența proprietăților ureei de dimensiunile particulelor materialului şi sunt în concordanță cu cele din literatura de specialitate.