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FLUID-DYNAMIC, COATING EFFICIENCY AND ADHESION OF AN AQUEOUS POLYMERIC SUSPENSION TO INERT PARTICLES IN A SPOUTED BED

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Key Words and Phrases: adhesion; contact angle; particle coating; spouted bed fluid dynamic.

Abstract: This work aimed at investigating the fluid dynamics of the conventional spouted bed during a coating process, using several inert particles such as polystyrene, polypropylene, polyethylene of low density, ABS®, placebo tablets and glass beads, analyzing the adhesion of the film on the particle and the influence of the physical properties of the particles and of an aqueous polymeric coating suspension on the fluid dynamics.

Nomenclature

d – diameter, mm;
M – mass, kg;
RH – relative humidity, %;
t – process time, min;
T – temperature, ºC;
V – velocity, cm/s;
X – moisture content, %;
Q – Flow rate, kg/s;
P – pressure, Pa;
C – concentration, %;
W – suspension flow rate, m^3/s;
H – height, cm;
n – behavior index;
m – consistency index, N/m^2;
δ – particle growth, %;
η – coating efficiency, %;
θ – contact angle, º;
ϕ – sphericity;
Φ – angle of repose, º;
ρ – density, kg/m^3;
ε – porosity, %;
τ – shear stress, N/m^2;
γ – rate of strain, s^{-1};
Δ – drop.

Subscripts

p – particle;
sol – solids;
S – stable;
M – maximum;
s – spout;
ms – minimum spout;
o – dry bed;
susp – suspension;
ap – apparent;
i – initial;
f – final;
an – annulus;
ft – fountain.

Introduction

Coating is an unit operation widely used in food, chemical, agricultural and pharmaceutical industries, as an effective technique for modifying the surface characteristics and protecting products, such as tablets, confects, fertilizers, seeds, etc.

The utilization of the spouted bed as an efficient equipment for coating of coarse particles (d_p > 1 mm) has been a subject of several studies [1 – 5].

The coating process in a spouted bed consists in dispersing of the coating suspension over the particles being in a cyclic motion in the bed. Heated air dries the suspension deposited on the particles surface. The formation of the film coat is realized by repeated passages of the particles in the spray zone [6, 7]. Thus, the fluid-dynamic pattern of the particles in the bed, during spraying of the suspension, is very important for the performance of the coating process and product quality. The bed fluid-dynamic stability is necessary to obtain a product with homogenous coating. In addition, the dynamic stability determines a reproducible process with economically viable conditions.

Since the fluid dynamic of the bed, during spraying of the suspension, is strongly influenced by inert particles and coating suspension characteristics, this work has the aim to analyze the adhesion of the film on the particle and the fluid dynamic of a conventional spouted bed during a film coating process, while spraying an aqueous polymeric suspension on the inert particles.

Materials characterization

Particles characterization

Polystirene (**PS**), polypropylene (**PP**), low-density polyethylene (**LDPE**), ABS®, placebo tablets (composed basically of lactose, starch and magnezium stearate) and glass beads were used as inert particles. They were analyzed and classified by size (sieve analysis) sphericity (the ratio of the biggest inscriptive diameter and the smallest circumscriptive diameter in the projected area of the particle on a stable plane [8]), true (Helium picnometry) and apparent (mercury porosimetry analysis) density, bulk density, porosity, angle of repose (rotator barrel method), and flowability (flow through an 80º funnel with 1,5 cm of orifice diameter). The particles characteristics are given in table 1.

Table 1

Inert particles characteristics

Material	Glass beads	PS	LDPE	PP	PlaceboTablets	ABS®
Shape	spherical	wedge	pellet	pellet	cylindrical	spherical
d_p, mm	2,77	4,58	3,56	2,91	5,54	2,90
ϕ	0,865 ± 0,037	0,880 ± 0,047	0,785 ± 0,050	0,750 ± 0,066	0,825	0,788 ± 0,045
Φ, º	33,0 ± 1,4	34,5 ± 1,9	35,5 ± 1,5	35,0 ± 1,0	43,8 ± 2,0	43,7 ± 1,9
ρ_{bulk}, kg/m^3	1562,5 ± 31,8	657,5 ± 4,1	595,4 ± 5,5	597,9 ± 6,8	862,9 ± 24,4	648,2 ± 6,0
ρ_{true}, kg/m^3	2491,5 ± 0,4	1060,0 ± 0,6	919,3 ± 0,2	905,3 ± 0,6	1657,1 ± 0,8	1022,1 ± 0,8
ρ_{ap}, kg/m^3	2491,7	1049,1	919,4	905,3	1348,2	1013,6
ε_p, %	0	1,03	0	0	18,64	0,83
Flowability, cm/s	18,86 ± 0,22	19,43 ± 0,20	22,92 ± 0,53	23,76 ±0,12	–	16,03 ± 0,54

Characteristics of the coating suspension

The formulation of the polymeric coating suspension (table 2) is based on previous studies by Ataíde [9] on coating of tablets in spouted beds and spout-fluid beds. The suspension properties include: density, surface tension and rheology (table 3). The density was determined by picnometry, the surface tension, by the du-Nouy tensiometer, and the rheological parameters were obtained by a capillary rheometer through the pseudo-rheologic curve. The contact angle between the suspension and the solid surface of inert particles was determined with a contact angle micrometer (Cole-Parmer®, USA). The results are show in table 4.

Table 2

Formulation of the polymeric coating suspension

Reagent	% mass
Hydroxyethylcelulose (HEC)	3,50
Polyethylene Glycol 6000	0,75
Magnesium Esthearate	1,00
Titanium Dioxide	1,25
Colorant	1,00
Talc	3,50
Water	89,00
C_{sol}, %	11,00

Table 3

Characteristics of the suspension

Density, kg/m^3	Surface Tension, 10^3 N/m	Rheology
1033 ± 3 (14…37 ºC)	63,37 ± 0,98 54,81 ± 0,15 (35 ºC)	Pseudo-Plastic Power-Law Fluid n = 0,8625 ± 0,0299 m = 0,1086 ± 0,0042 R^2 = 0,9964 (25 ºC)

Table 4

Contact angle of the suspension-inert material

Material	Glass beads	ABS®	Placebo tablets	PS	PP	LDPE
θ, º	40,3	71,5	74,0	79,9	81,0	92,4

The experimental set-up

The experimental system is shown in Fig. 1. The spouted bed was built from plexiglass and its dimensions are: column diameter of 20 cm, column height of 30 cm, orifice diameter of 5 cm and angular base of 60º.

Samples of particles were collected during the coating process at the annulus region, and the values of moisture content were obtained by the static gravimetric method, in a vacuum oven at 75 °C and 25 in Hg, during 24 hours. The inlet and outlet relative humidity of the air were measured by hygrometers at the specified locations.

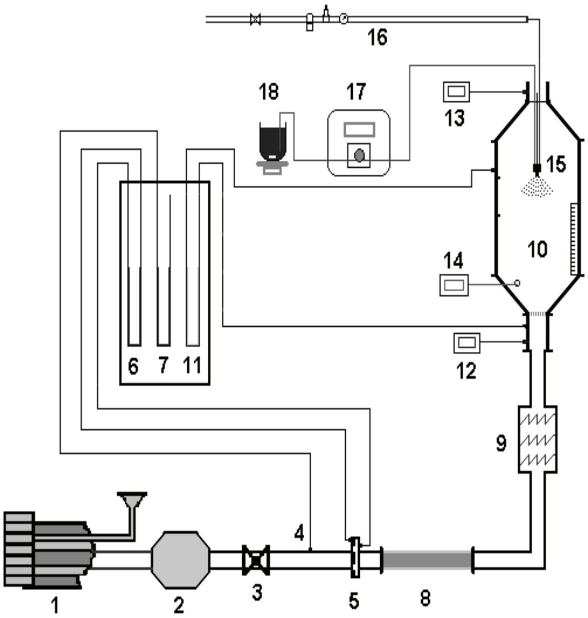

Fig. 1 Experimental Set-up:
1 – blower; *2* – cooler; *3* – control valve; *4* – pipeline; *5* – orifice plate; *6* – differential manometer; *7* – differential manometer; *8* – silica gel bed; *9* – resistances; *10* – bed; *11* – differential manometer; *12* – thermo-hygrometer; *13* – thermo-hygrometer; *14* – thermocouples; *15* – sprayer; *16* – compressed air line; *17* – peristaltic pump; *18* – coating suspension

Methods

During the coating process, the following measurements were taken: the bed pressure drop, the temperature and the relative humidity of inlet and outlet air, the temperature of the particles in the annulus, the fountain and the annulus height, and the circulation velocity of particles at the annulus wall. The experiments in duplicate showed appropriate reproducibility.

The circulation velocity was determined by measuring the time spent by a particle to pass a distance of 3,5 cm marked on the transparent wall of the conical part of the bed. Ten measurements were made for each time interval, during the coating process, and the mean value was adopted for the circulation velocity. The annulus and fountain heights were measured by a ruler attached to the bed wall.

The following operating conditions were maintained constant in all experiments. The gas temperature was 60 ºC, the coating suspension was sprayed at 137,89 kPa (20 psig) and the suspension flow rate was $2,10^{-7}$ m^3/s (12 ml/min). The values of the ratio of the air flow rate and the minimum spouting flow rate in a dry bed ($Q_s/Q_{ms,o}$), the process time and the load of particle utilized for the coating experiments are presented in table 5.

For the evaluation of the coating efficiency, η, the following definition used by several researchers [2, 10-13], was adopted in this work.

$$\eta = \frac{\text{mass of solid adhered to particles}}{\text{total mass of solid supplied to the bed}} = \frac{M_{\text{f}} - M_{\text{i}}}{W_{\text{susp}} C_{\text{sol}} t} . \qquad (1)$$

Table 5

Operating conditions

Material	$Q_s/Q_{ms,o}$	t, min	M, kg
Glass bead	1,08	30	3,3
PS	1,22	10	1,5
LDPE	1,34	5	1,4
PP	1,33	10	1,4
Placebo tablet	1,09	30	1,8
ABS®	1,09	30	1,4

The mass relative growth, δ, was calculated as:

$$\delta = \frac{M_f - M_i}{M_i} 100. \tag{2}$$

Results and discussion

The fluid-dynamic of a dry bed was analyzed previously to establish operating conditions for the coating experiments. Table 6 presents the minimum spouting flow rate (Q_{ms}), the maximum (ΔP_M) and the stable (ΔP_S) bed pressure drop determined for the dry bed.

Among all the particles tested, only three of them were coated by the suspension of HEC. Particles with a high contact angle ($\geq 80^o$) such as polystyrene (PS), polypropylene (PP) and polyethylene of low density (LDPE), revealed no adhesion of the suspension to the particle surface, and the dry powder was totally elutriated. For the other particles (ABS®, placebo tablets and glass beads), satisfactory values of the coating efficiency and particle growth were gotten in the experiments. Fig. 2 illustrates the coating and drying process as a function of the suspension-particle contact angle.

Table 7 shows the values for temperature and relative humidity of inlet and outlet air, temperature of particles in the annulus, moisture content of the coating film at the end of the process, coating efficiency and growth obtained from experiments. The coating efficiency and the relative mass growth were calculated only for the coated particles.

Table 6

Fluid-dynamic parameters

Material	$Q_{ms} \cdot 10^3$, kg/s	ΔP_M, kPa	ΔP_S, kPa
LDPE	7,35	1,057	0,732
PP	9,03	1,089	0,731
PS	11,07	1,371	0,546
ABS®	12,72	1,282	0,614
Placebo tablet	19,86	3,047	0,909
Glass bead	16,24	3,195	1,741

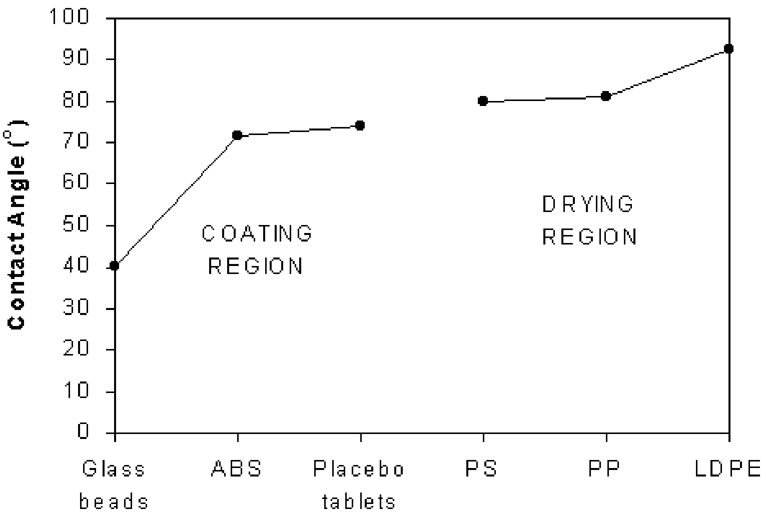

Fig. 2 Influence of the contact angle on the film adhesion

Table 7

Operating conditions and results from coating experiments

Material	T_{inlet}	T_{outlet}	T_{an}	RH_{inlet}	RH_{outlet}	X, db %	η, %	δ, %
LDPE	60,4	41,7	41,0	7,5	63,6	0,06	–	–
PP	60,0	46,1	44,7	11,2	35,0	0,11	–	–
PS	60,2	43,2	46,8	11,3	36,5	0,05	–	–
ABS®	60,0	37,1	38,0	13,1	53,8	0,37	84,04	2,79
Placebo tablet	60,3	45	48,0	11,3	33,3	8,58	90,37	1,97
Glass bead	60,4	45,6	49,0	17,4	38,5	0,03	76,33	0,97

The suspension-particle contact angle is a property that determines the wettability of the solid so it governs the coating process. However, the other particle properties such as apparent density, porosity, angle of repose and shape have a great influence on the bed fluid dynamics. Consequently, these properties influence the drying process, and the uniformity and growth of the coating film as well as the coating efficiency. Particles of PS and PP have close values of the suspension contact angle, porosity and apparent density, suggesting that both would present a similar behavior in the experiments. But, the coating film formed on the PP surface is not continuous and detaches more easily, whereas the film formed on the PS surface is more homogeneous. The drop spreading on the PS surface is more complete than the one on the PP, due to its plane surface. Therefore, the contact angle qualifies the wettability and the adhesion, thus determines whether the particle will be coated or it will serve as an inert carrier for drying of the suspension. The same behavior was observed by Pont et al. [14] in a fluidized bed coater. The geometric shape determines either the coating or the drying efficiency.

The particles that are coated by the suspension (ABS®, tablet and glass beads) have a lower contact angle and a uniform shape. For the glass beads and ABS® particles, their spherical shape minimizes the inter-particle attrition during the coating, helping the formation of a homogeneous film.

Table 8

Contact angle for uncoated and coated particles.

Material	Glass beads		ABS®		placebo tablets	
	uncoated	coated	uncoated	coated	uncoated	coated
θ, º	40,3	72,4	71,5	72,6	74,0	85,0

The first layer of coating was responsable by the film-particle adhesion, but after the first layer of coating, the interaction was between the suspension drop and the coating film adhered on particle. Table 8 presents the measurements of contact angle of the interfaces suspension-dry film for coated particles. It can be observed that for nonporous particles, the values of the suspension-dry film contact angles were similar-in magnitude, about 72 degrees. For the tablet, its high porosity (18,64 %) facilitates absorption of the suspension, in spite of the high contact angle of the coated placebo tablets. Due to its high porosity, there was neither free moisture on the particles surfaces nor liquid bridges linking the particles. Thus, the interparticle forces were not significant in this case, maintaining intense particle circulation in the bed, even with the increase of moisture inside the bed.

Fig. 3 shows the particle growth for coated particles, in terms of mass of the adhered film. The experimental data were fit by a linear model with satisfactory adjust, having a correlation coefficient higher than 0,99. The linear growth of particles indicates that the coating efficiency can be considered constant during the process.

The fluid-dynamic study for the three particles coated by the suspension can be visualized in Fig. 4, 5 and 6. Fig. 4 shows the values for the fountain and annulus heights and the bed pressure drop during the coating experiments, parameterized in relation to the dry bed. The fountain height obtained when coating glass beads and placebo tablets was practically the same as the dry bed value and for the ABS® wet bed, the fountain height even decreased. This behavior indicates dynamic stability of the spouted bed during coating of ABS®, glass beads and placebo tablets. The annulus height was practically maintained the same as for the dry bed.

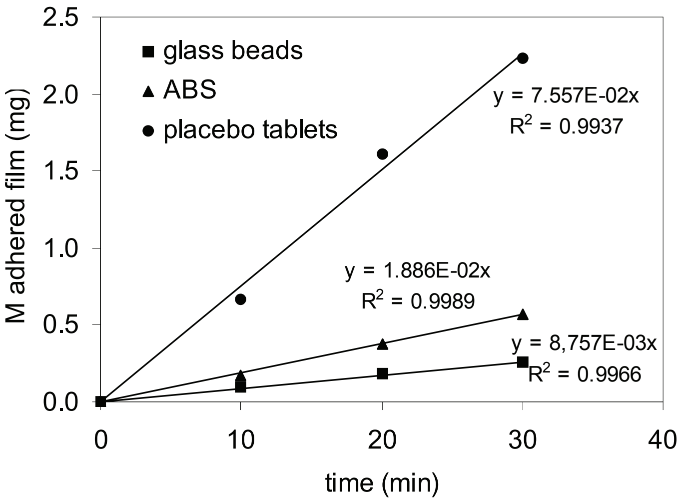

Fig. 3 Particle growth during coating

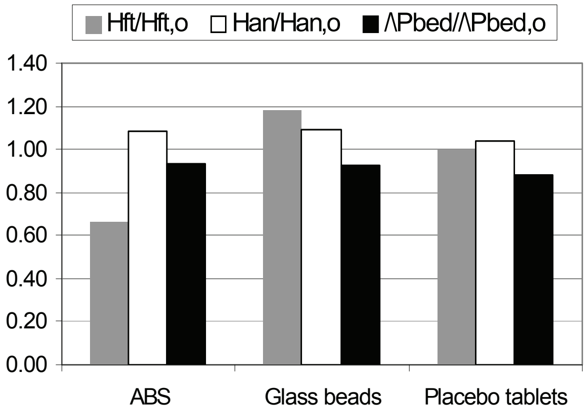

Fig. 4 Fountain and annulus height and bed pressure drop during the coating

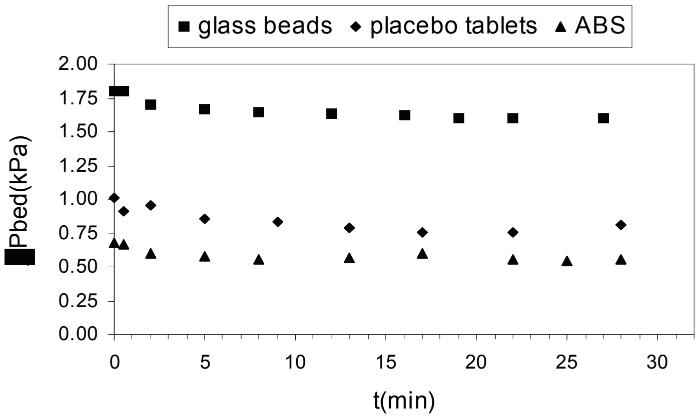

Fig. 5 Bed pressure drop for ABS®, placebo tablets and glass beads

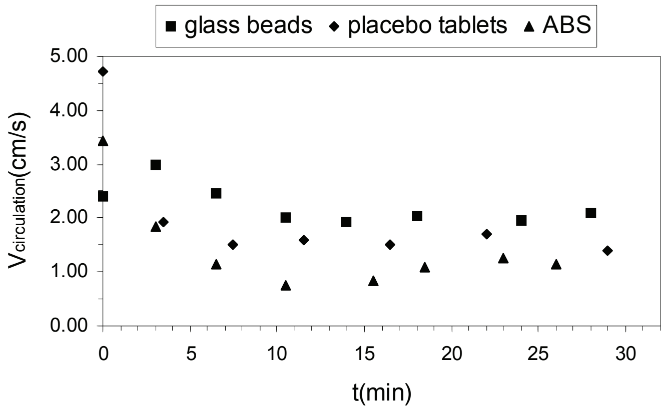

Fig. 6 Particles circulation velocity in the annulus during coating for ABS®, placebo tablets and glass beads

Fig. 5 presents the bed pressure drop during the coating process. For the coated particles tested, a reduction of the bed pressure drop was verified at the beginning of the experiments. After a short time (about 5 minutes), the pressure drop stabilized at a value bellow the value characteristic of a dry bed. The difference between the bed pressure drop in a dry bed and in a wet bed is more significant for particles having low apparent density. The glass beads, for example, as denser particles, presented a reduction by only 7,5 % of this parameter.

Other authors [15 – 18] also noted the reduction in the bed pressure drop with increasing moisture inside the bed due to spraying of the suspension (Fig. 5). A possible reason for this behavior is that with the increasing moisture content, the annulus becomes more packed and thus a larger fraction of the spout air passes through the preferential channel, the spout, making it more diluted and thus, decreasing the bed pressure drop.

The particle circulation velocity at the annulus wall during coating can be seen in the Fig. 6. As it was observed for the bed pressure drop, the circulation velocity decreases at the beginning of the process, and after 5 minutes it stabilizes and does not vary with time any more. The particles in a wet bed present a lower circulation velocity than in a dry bed, as the particles are heavier, besides the increasing of inter-particle forces, which hamper their mobility inside the bed. This behavior agrees with the reduction of the bed pressure drop, indicating a decrease in the number of particles with a consequent increase of bed voidage in the spout region.

The difference between the circulation velocity in a dry bed and in a wet bed is more significant for particles having a high contact angle and a low density. The glass beads presented a reduction on the circulation velocity in a wet bed by only 25 % whereas the ABS® presented a reduction by 75 %.

It can be also verified from Figure 6 that the time over which stabilization of the particle circulation velocity occurs in the wet bed is the same as the bed pressure drop stabilization. Therefore, the instant when the bed reaches a stable spout regime during a coating process can be obtained by monitoring the bed pressure drop.

Conclusions

Particles with a high contact angle of suspension, ≥ 80º, such as polystyrene (PS), polypropylene (PP) and low-density polyethylene (LDPE) are not coated by the polymeric suspension of HEC used in this work. There is no enough adhesion of the suspension to the particle surface and the dry powder is practically totally elutriated. For the other particles analyzed, namely ABS®, placebo tablets and glass beads that have the contact angle less than 74º, satisfactory values of the coating efficiency, from 76 to 90%, and particles growth, from 0,97 to 2,79 %, were gotten in the experiments.

The first layer of coating is responsible by adhesion of the film to the particle, indicating if coating or drying process will occur.

The uniformity of the film, the particle growth and the coating efficiency are influenced by the suspension-film interaction, the particle properties and the bed fluid-dynamic.

A reduction of the bed stable pressure drop was verified when moisture is present inside the bed for the particles tested. The spouted bed stable pressure drop in a dry bed is higher than in a wet bed, and the difference between the values is more significant for particles having low apparent density

The particle circulation velocity in the annulus decreases with increasing moisture inside the bed, keeping constant all the operating conditions during the coating experiment.

Based on the results presented in this work, the particles physical properties that influence the coating process are:

- contact angle of suspension: a low contact angle represents a higher adhesion of the suspension to the particle surface, thus improving the coating process and the bed dynamics stability.

- angle of repose: a low angle of repose indicates good flowability of the particle in the bed, and increase of the moisture inside the bed due to the spraying of suspension does not damage in a significant way the particle circulation in the annulus.

- apparent density: denser particles support higher moisture content in the bed without instability symptoms .

- shape: the particle geometric shape has a significant influence on the coating efficiency. Particles with a uniform surface and a spherical shape presented a higher coating efficiency.

- porosity: the particle porosity allows absorption of the sprayed suspension, also improving the coating process with no effect on particle circulation.

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Гидродинамика, интенсивность покрытия и адгезия водных полимерных суспензий к инертным частицам в псевдоожиженном слое

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Ключевые слова и фразы: адгезия; гидродинамика фонтанирующего слоя; покрытие частиц; угол контакта.

Аннотация: Цель данной работы – изучить гидродинамику в обычном фонтанирующем слое при нанесении покрытия, с использованием инертных частиц, таких как полистирол, полипропилен, полиэтилен низкой плотности, шарики АБС-пластика (сополимер акрилонитрила, бутадиена и стирола), фармацевтические таблетки плацебо и стеклянные шарики, и проанализировать адгезию пленки к частицам и влияние физических свойств частиц и водных полимерных покрывающих суспензий на гидродинамику слоя.

Hydrodynamik, Intensität der Deckung und Adhäsion der Wasserpolymeraufschlämmungen zu den Inertteilchen in der Fluidisationsschicht

Zusammenfassung: Das Ziel dieser Arbeit besteht darin, die Hydrodynamik in der üblichen Strahlschicht bei dem Deckungsauftragen mit Benutzung von einigen Inertteilchen solcher wie Polystyrol, Polypropylen, Polyäthylen von niedriger Dichte, Kügelchen vom ABC-Kunststoff (Kopolymer von Vinylzyanid, Butadien und Stirol), die pharmazeutischen Tabletten Plazebo und die Glaskügelchen zu erlernen und die Adhäsion der Folie zu den Teilchen und die Einwirkung der physikalischen Eigenschaften der Teilchen und der Wasserpolymeraufschlämmungen auf die Hydrodynamik der Schicht zu analysieren.

Hydrodynamique, intensité du revêtement et adhésion des suspensions polymères envers les particules inertes dans une couche pseudo-liquide

Résumé: Le but de cet article est d'étudier l'hydrodynamique dans une couche d'éruption au cours du revêtement avec l'utilisation de quelques particules inertes comme, par exemple, polystyrène, polypropylène, polyéthylène de la basse densité, des bulles du plastique ABC (copolymère d'acrylonitrile, butadiène et styrène), des tablettes pharmaceutiques placebo, des bulles de verre, et d'analyser l'adhésion du film envers les particules et l'influence des propriétés physiques des particules et des suspensions polymères aqueuses de revêtement sur l'hydrodynamique de la couche.