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European Journal of Pharmaceutics and Biopharmaceutics 44 (1997) 269–272

European Journal of Pharmaceutics and Biopharmaceutics

Paper

Percolation theory and physics of compression

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Received 3 July 1996; accepted 22 April 1997

Abstract

The concept of percolation theory is an excellent tool to elucidate the physics of compression. Earlier findings taking into account the percolation theory indicated that the formation of a tablet can be subdivided into a two-stage process with a 'weak-bond' percolation effect at a lower percolation threshold p_c corresponding to the relative tapped density ρ_r and a 'strong-bond/site' percolation effect at an upper percolation threshold p_c^*, i.e. at a relative density ρ_r^*, where brittle fracture and/or plastic flow starts to play an important role for the formation of a stable compact. The new findings which are now presented indicate that the uniaxial compression can be interpreted as a 2-dimensional percolation process where the stress is transmitted by the contact points of the particles. Thus the modified Young's elasticity modulus of the compact can be described by the fundamental equation of percolation theory with a critical exponent $q = 1.3$ (which is the conductivity exponent) and with a lower percolation threshold of the relative tapped density ρ_r. If the same equation is applied for the tensile strength, high values of the critical exponent result, indicating that a still unknown fractal dimension seems to play a key role. © 1997 Elsevier Science B.V.

Keywords: Percolation theory; Uniaxial compression; Elasticity; Tensile strength; Lactose-granules; STA-RX 1500®

1. Introduction

1.1. Percolation theory

Percolation theory [1] represents a novel powerful concept which covers a wide range of applications in pharmaceutical technology. It provides new insights into the physics of tablet compaction and the properties of compacts [2–7].

Different types of percolation can be distinguished: random-site, random-bond, random-site-bond, continuum, etc. Generally, percolation theory deals with the number and properties of clusters [1]. A percolation system is considered to consist of sites in an infinitely large real or virtual lattice.

At a percolation threshold p_c some property of a system may change abruptly or may suddenly become evident. Such an effect starts to occur at the percolation threshold p_c and is usually called a critical phenomenon. Table 1 shows critical volume-to-volume ratios for well-defined geometrical packing of monosized spherical particles. The percolation thresholds depend on the type of percolation and the type of lattice, i.e. the micro structure of the system [1].

A number of tablet properties are directly or indirectly related to the relative density ρ_r of the compact. Within the framework of percolation theory the relative density ρ_r represents the occupation probability p of a lattice spanning the volume of the tablet. The lattice sites which are not occupied represent the pore structure. According to percolation theory the following relationship holds for the tablet property X close to the percolation threshold p_c [1]:

$$X = S \cdot (p - p_c)^q \qquad (1)$$

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PII S0939-6411(97)00130-6

Where X is the tablet property, p the occupation probability, p_c the percolation threshold, S the scaling factor and q the critical exponent.

The critical exponent q depends only on the Euclidean or fractal dimension of the process (e.g. $q = 1.3$ for the conductivity in two dimensions) [1].

1.2. Process of uniaxial compression

A 3-dimensional lattice spanning the die volume is imagined. During compression the number of sites to be occupied is constantly reduced. According to the principle of uniaxial compression the mean particle–particle separation distance is more reduced in the z-direction than in the lateral directions. Thus, it can be assumed that in the beginning a 1-dimensional bond percolation is responsible for stress transmission. After the rearrangement of the particles an important build-up of stress occurs as particles can no longer be displaced easily. This situation is typical for a site percolation process. The stress transmission is mainly in the lateral direction. Thus, the original 3-dimensional problem can be split into a 1-dimensional and, subsequently, 2-dimensional percolation phenomenon [5–7]. A bond percolation effect could be identified at a lower percolation threshold p_c corresponding to the relative tapped density ρ_r. Thus the tensile strength of slightly compressed powder systems could be described by a straight line, i.e. $q = 1$ for relative densities close to the relative tapped density [5]. However, at a relative density ρ_r^* a site percolation effect seems to occur as above $p_c^* = \rho_r^*$ brittle fracture and/or plastic flow starts to play an important role for the formation of a stable tablet.

Thus the tensile strength of a tablet could be approximated by two linear relationships as a function of the relative density describing a two-stage process with a 'weak-bond' percolation effect close to the relative tapped density ρ_r and a 'strong-bond'/site percolation effect at an upper percolation threshold p_c^*. This model, which divides the compaction process into two completely different phases, needs the identification of two percolation thresholds: the lower one, which is close to the relative tapped density and an unknown upper one [5]. On the other hand, the modified Young modulus of elasticity could be approximated by a straight line for the whole range of relative densities assuming a single lower percolation threshold p_c at the relative tapped density.

Thus, the present study investigates the following questions and pursues the following goal:

1. Hypothesis I: The lower percolation threshold p_c equal to the relative tapped density is the single bond percolation threshold, where the stress is transmitted by the contact points of the particles.
2. Hypothesis II: The mechanical properties X (modified Young's modulus of elasticity, tensile strength) can be described by the fundamental Eq. (1) of the percolation theory for the whole range of the relative densities involved.
3. The experimental determination of the value of the critical exponent q needs a sufficient precision and tablets with a broad range of relative densities need to be produced.

Table 1
Selected percolation thresholds for various lattices

Lattice	Site	Bond
2-Dimensional		
Honeycomb	0.696	0.653
Square	0.593	0.500
Triangular	0.500	0.347
3-Dimensional		
Diamond	0.430	0.388
Simple cubic	0.312	0.249
Body-centered cubic	0.246	0.180
Face-centered cubic	0.198	0.119

'Site' refers to site percolation and 'bond' to bond percolation [1]

2. Theoretical model of the tablet mechanical properties: elasticity and tensile strength

Kirkpatrick [8] found that near the percolation threshold different 3-dimensional lattices have a similar behaviour. Thus, the dimensionality of the system is more important than the details of the lattice, i.e. no special assumptions have to be made for a random continuum system. Taking into account the hypotheses described in the previous section and the combination of the earlier developed mathematical model of Leuenberger and Leu [5,6] with the findings of Kirkpatrick leads to the following equations for the elasticity (Eq. (2)) and the tensile strength (Eq. (3)):

$$\frac{E^*}{E^*_{max}} = S \cdot (p - p_c)^q \quad (2)$$

$$\frac{\sigma_T}{\sigma_{T_{max}}} = S \cdot (p - p_c)^q \quad (3)$$

Where E^* is the modified Young's modulus of elasticity [3], E^*_{max} the extrapolated maximal modified Young's modulus of elasticity, σ_T the tensile strength and σ_{Tmax} the extrapolated maximal tensile strength [9].

3. Materials and methods

3.1. Materials

Tablets (round, flat, diameter 11 mm, weight 400 ± 1 mg) for the subsequent determination of the modified Young's modulus of elasticity and the tensile strength using the Zwick 1478 Universal Testing Instrument (Zwick GmbH, Ulm, Germany) were prepared. As starting material the following powder systems were used: α-lactose monohydate 200 mesh (De Melkindustrie Veghel, The Netherlands) batch No. 024341 (mean size: 84.3 μm, rel. tapped density: 0.54); batch No. 92882 (mean size: 95.2 μm, rel. tapped density: 0.50); lactose–polyvinylpyrrolidone (96 + 4 wt./wt.%) granulate (mean size: 266.8 μm, rel. tapped density: 0.51) and its sieve fractions fine: 500–710 μm (rel. tapped density: 0.40); medium: 710–1000 μm (rel. tapped density: 0.38), coarse: 1000–1200 μm (rel. tapped density: 0.42), STA-RX 1500® batch No. 12 (mean size: 94.7 μm, rel. tapped density: 0.56), STA-RX 1500® batch No. 811024 (mean size: 77.9 μm, rel. tapped density: 0.53). The method of preparation is described in detail in [5,9].

3.2. Methods

The Eq. (2) and Eq. (3) were tested with different critical exponents q, which were found in the literature [1,10] and also with an unknown exponent q to be determined, with a fixed p_c equal to the relative tapped density [9]. Due to the flip-flop property of Eq. (2) and Eq. (3) either p_c or q needs to be fixed. The data were evaluated using non-linear regression analysis described in detail in [5].

4. Results and discussion

4.1. Elasticity (modified Young's modulus)

A critical exponent q of 1.3 gives the best fit of the data with Eq. (2) (Fig. 1). This result is supported by the fact, that the determination of the critical exponent q of the series of investigated powders and granules ($n = 8$, i.e. six batches of lactose powder and granules and two batches of STA-RX 1500®) leads to a mean value of 1.305 ± 0.121 with a fixed p_c equal to the relative tapped density [9].

A critical exponent of $q = 1.3$ is found in the literature for the 2-dimensional problem of conductivity [1]. As we can assume, that the elasticity behaves similarly to the conductivity, i.e. the stress and the electricity are in both cases transmitted through the contact points of the particles, a critical exponent $q = 1.3$ is reasonable for the elasticity.

4.2. Tensile strength

In the case of the tensile strength the determination of q leads to rather high values of the critical exponent (lactose powder: $q = 4.6$, lactose granules: $q = 6.6$) yielding the best fit of Eq. (3), [9] (Fig. 2). Such high critical exponents are not expected for the case of Euclidean dimensions [1,10].

A possible explanation of these high critical exponents for the tensile strength is found in the work by Ehrburger and Lahaye [11]. They found critical exponents of $q = 5.92$ in two dimensions and $q = 3.89$ in three dimensions compacting colloidal silica. Ehrburger and Lahaye explain these high values for the critical exponents as a consequence of the fractal dimension of the agglomerates, which are compacted. Thus, it is

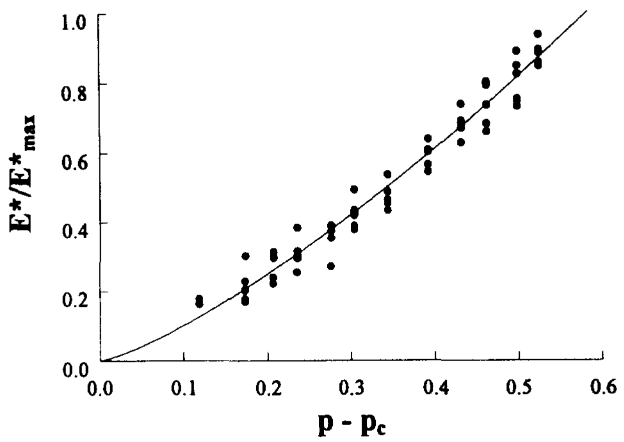

Fig. 1. Tablet property: normalised modified Young's modulus of elasticity (E^*/E^*_{max}) of lactose-granules compacted as a function of the relative density according to Eq. (2) with a critical exponent $q = 1.3$ and a fixed p_c equal to the relative tapped density [9].

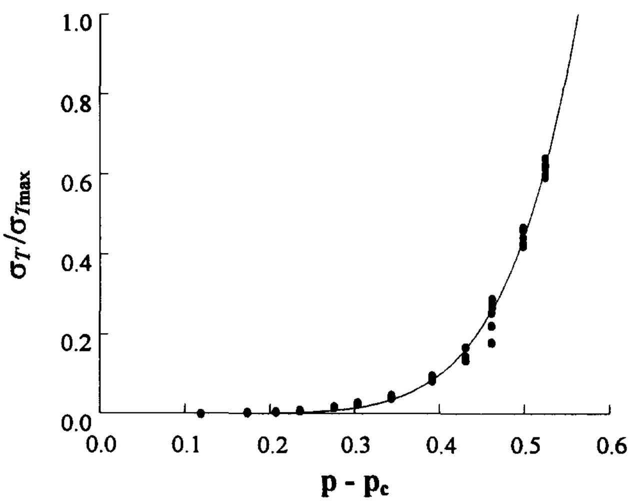

Fig. 2. Tablet property: normalised tensile strength ($\sigma_T/\sigma_{T_{max}}$) of lactose-granules compacted as a function of the relative density according to Eq. (3) with a critical exponent to be determined ($q = 6.6$) and a fixed p_c equal to the relative tapped density [9].

possible to use the fundamental Eq. (1) of percolation theory to describe the tensile strength for the whole range of the relative densities. However, a still unknown fractal dimension, which is related to the property of the powder or the granule has to be assumed to be responsible for the values of the critical exponent q.

5. Conclusions

From the results obtained with the modified Young's modulus of elasticity, where a critical exponent $q = 1.305 \pm 0.121$ was obtained, it can be concluded that the uniaxial compression process can be considered as a 2-dimensional percolation phenomenon, describing how the sites of the cross-sectional virtual 2D-lattice are occupied.

The fundamental equation of percolation theory can also be applied for the tensile strength of a lactose tablet for the whole range of relative densities yielding a high value of the critical exponent, which indicates that a still unknown fractal dimension may govern the process. Further investigations are necessary to decide unambiguously whether the model with a single percolation threshold and a high value of the critical exponent q assuming the involvement of a fractal dimension governing the compression process can be justified and is superior to the previously developed two-stage compression model assuming two different percolation thesholds [5].

Acknowledgements

We acknowledge the financial support given by the Swiss National Foundation (grant 20–34049.93).

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