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The influence of particle size and shape of components of binary powder mixtures on the maximum volume reduction due to packing

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Abstract

The influence of particle size and shape of components of binary powder mixtures on the maximum volume reduction due to packing has been investigated in a systematic manner using the Kawakita-equation. Different grades of microcrystalline cellulose were mixed with defined size fractions of angular particles (lactose monohydrate), spherical particles (Elcema G250) and needle-shaped particles (acetylsalicylic acid). The particle shape of the mixture components clearly influenced the maximum volume reduction due to packing, represented by the Kawakita constant *a*. While angular particles improved the packing properties of the microcrystalline celluloses, the addition of spherical and needle-shaped particles generally resulted in lower values for *a* at equivalent concentrations. The addition of both spherical or needle-shaped particles in concentrations between 25% and 75% to fine grade microcrystalline cellulose led to a steady decrease in volume reduction ability, whereas for medium or coarse grade microcrystalline cellulose the graphical presentation of *a* as a function of concentration of the second mixture component always showed a maximum value. The particle size of both powders contained in the binary mixtures influenced the values of the Kawakita constant *a* with the exception of angular particles. The finer the particle size of each component, the larger became the value of *a*.

Keywords: Binary powder mixtures; Kawakita-equation; Particle shape; Particle size; Powder packing

1. Introduction

The packing properties of powders are very important for the production of solid dosage forms such as tablets or capsules. For example, in capsule filling it is desirable to be able to fill powders or granules into capsules of a smaller size such as size 2 or 3, even if the required fill weight of the formulation is rather large. Hence, in addition to the flow properties of powder mixtures their packing properties need to be optimized.

In a powder bed of monosized spherical particles a variation in the packing structure can arise, so that the interparticulate porosity reaches its extremes of 26% for a tight rhombohedral, or 48% for a loose cubical packing (Martin et al., 1983). If spherical particles of a coarse and a fine

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monosized fraction are mixed, ideal bimodal packing (Furnas, 1931) will occur, because the fine particles can occupy the space left between the coarse particles. By adding fine monosized spherical particles to coarse monosized spherical particles so that the volume fraction of the coarse powder is between 0 and 0.735, the packing density of the powders will increase proportionally to the increase in the coarse powder volume fraction, and will reach its maximum at a coarse powder volume fraction of 0.735. Above this value, the packing density will drop sharply due to a lack in fine particles to fill the interparticulate pores. However, in practical binary mixtures, particles are usually not monosized. Lam and Nakagawa (1994) have shown that in this case the shape of the particle size distributions of the fine and coarse powder fractions will influence the value of the coarse powder volume fraction, at which maximum packing occurs, and therefore a prediction of the composite packing density of binary mixtures based on Furnas' model will be erroneous. Westman and Hugill (1932) proposed a different model for predicting composite packing densities, which could be extended for mixtures comprising more than two particle fractions. Again, spherical particle shape is required for its validity. While this model appears to successfully predict the composite packing density of binary mixtures composed of different but narrow particle size fractions (Newton and Bader, 1981), it loses power with each additional powder component (Staple, 1975). This suggests that it will also fail if the powder components in a binary mixture comprise a wider size distribution, or if the size distributions are truncated or largely skewed.

None of the models discussed above is able to predict a maximum in volume reduction achieved by application of vibration, small pressure or simply tapping. However, such a prediction would be as desirable in capsule filling as the prediction of the composite packing density. A possible method to be used for this purpose is the Kawakita model (Kawakita, 1956). The model has been extensively reviewed by Kawakita and Lüdde (1970/71) for use in connection with compaction under high loads, but it can also be used to study the volume reduction of a powder due to tapping (Lüdde and Kawakita, 1966):

$$\frac{N}{c} = \frac{1}{ab} + \frac{1}{a}N \tag{1.1}$$

$$c = \frac{V_o - V_N}{V_o} \tag{1.2}$$

$$a = \frac{V_o - V_\infty}{V_o} \tag{1.3}$$

where a and b are constants characterizing the powder, N is the tapping number, c represents the degree of volume reduction achieved after N tappings, V_o is the maximum bulk volume of the powder, V_N is the bulk volume of the powder after N tappings, and V_∞ is the minimum bulk volume. If the minimum bulk volume can be approached by a finite number of tappings, i.e. the tapped powder volume equals the minimum bulk volume, the constant a, which describes the maximal possible relative decrease of the initial (maximum) bulk volume due to tapping, can be approximated by:

$$a = 1 - \frac{V_{tapped}}{V_{maximum}} \tag{2}$$

However, the accurate determination of a requires measurement of the maximum bulk volume and the bulk volume after a defined number of tappings, repeating the last until no further change in bulk volume can be achieved by tapping. The constant a will then be calculated from the slope of the experimentally assessed Kawakita function according to Equation 1.1.

There exist no general rules about the influence of particle size and shape on packing and densification of powders, and both parameters appear to interact (Shotton and Obiorah, 1973). An irregular particle shape appears to contradict dense packing (Ridgway and Rupp, 1969). The possibility in reducing the powder bulk volume for example by tapping is also dependent on particle size and shape. Lüdde and Kawakita (1966) found that the constant a decreased with an increase in particle size. The value of a was smallest for spherical and largest for irregular particles. However, all observations discussed were made on single powders, and not on powder mixtures, which can comprise several particle sizes and shapes in combination.

The aim of this study was to assess the influence of particle size and shape of components of binary powder mixtures on the maximum volume reduction due to packing in a systematic manner using the Kawakita model.

2. Materials and methods

2.1. Materials

Three grades of microcrystalline cellulose were used: Avicel® PH105 ('MCC105'), Avicel® PH102 ('MCC102', FMC Corporation, Philadelphia, USA), Emcocel® ('MCC101', Edward Mendell, New York, USA). Lactose monohydrate BP ('LM', Bletchley, Amsterdam, Netherlands), Elcema® G250 ('G250', Rettenmaier and Sons, Ellwangen-Holzmühle, Germany), and acetylsalicylic acid ('ASS', Rhone Poulenc Ltd., Dagenham, UK) were also used.

2.2. Methods

A particle size analysis was made for LM, G250 and ASS by sieving (Endecotts Ltd., London, UK) using sieves of 500 μm, 250 μm, 125 μm, 90 μm and 38 μm aperture size. With respect to the particle size distributions obtained, the following sieve fractions were used in the experiments: LM: 125–250 μm; 90–125 μm; 38–90 μm;G250 and ASS: 250–500 μm; 125–250 μm; 90–125 μm.

The microcrystalline celluloses were used unsieved. They can be classified as fine grade (MCC105, average particle size 20 μm), medium grade (MCC101, average particle size 50 μm) and coarse grade (MCC102, average particle size 90 μm).

The particle shape was assessed by visual inspection under a microscope (BH-2, Olympus Co., Tokyo, Japan). MCC105 and MCC101 provided fibrous particles, whereas MCC102 appeared as large, spherical agglomerates. The shape of LM was angular, that of G250 was spherical, and ASS proved to be needle-shaped.

Binary powder mixtures were prepared using a Turbula mixer (Type Schatz T2C, Willy A. Bachofen AG Maschinenfabrik, Basel, Switzerland). Seventy-five g of powder were mixed for 10 min and the mixtures were stored in plastic bags. Each binary mixture contained one grade of microcrystalline cellulose and one particle size fraction of LM, G250 or ASS. The concentration of the microcrystalline cellulose component was 25% w/w, 50% w/w or 75% w/w.

The initial (maximum) bulk volume of all single powder components and each binary mixture using a defined mass was determined in a 100.0 ml measuring cylinder. The powders were then subjected to tapping using a tapped density volumeter (Copley Instruments, Nottingham, UK). Volume readings were undertaken first in intervals of 10 tappings, and depending on the progress of volume reduction later after consecutive tappings of 20, 50 or 100. It was found that even after 50 000 tappings the decrease in volume had not finished. Thus, the minimum bulk density could not be achieved in a reasonable time. Therefore, the Kawakita constant a was calculated from the slope of the volume-decrease function given in Equation 1.1. The tapped density was defined as the value where three consecutive readings did apparently not lead to a further decrease in powder bulk volume. In this respect, about 2500–3000 tappings were always necessary.

3. Results and discussion

An attempt to predict the tapped density of the binary mixtures from the values of the single powder components using Furnas' and Staples' equations failed completely. This is due to the fact that the tapped density volumeter used is not able to provide successful packing conditions. The tapping speed is too high and after an initial densification the machine vibrations lead to redispersion of the powder particles instead of a closer packing. The process of volume reduction varies from powder to powder due to differences in particle properties such as shape or size, and due to a different degree of interparticulate friction. Thus, the relative degree of packing achieved after about 3000 tappings varies from powder mixture to powder mixture, and cannot be predicted from the packing properties of the single components.

Table 1
Kawakita constant a [%] of single components and binary mixtures of microcrystalline cellulose and LM, G250 or ASS

MCC	2nd component		0%	35%	55%	75%	100%
MCC105	LM	(38–90 μm)	34.8	36.5	33.0	32.4	45.8
		(90–125 μm)	34.8	34.6	35.1	34.1	43.3
		(125–250 μm)	34.8	32.7	30.7	37.0	43.3
MCC101	LM	(38–90 μm)	22.5	26.1	29.1	32.9	45.8
		(90–125 μm)	22.5	24.7	28.2	36.0	43.3
		(125–250 μm)	22.5	20.1	31.0	31.9	43.3
MCC102	LM	(38–90 μm)	22.2	19.9	27.4	41.3	45.8
		(90–125 μm)	22.2	26.0	27.0	37.5	43.3
		(125–250 μm)	22.2	22.0	28.6	35.4	43.3
MCC105	G250	(90–125 μm)	34.8	32.5	27.6	27.9	23.7
		(125–250 μm)	34.8	29.9	24.1	20.0	21.9
		(250–500 μm)	34.8	28.0	25.3	23.3	21.9
MCC101	G250	(90–125 μm)	22.5	31.2	28.2	29.0	23.7
		(125–250 μm)	22.5	28.9	25.7	20.8	21.9
		(250–500 μm)	22.5	25.6	27.2	19.4	21.9
MCC102	G250	(90–125 μm)	22.2	27.8	29.3	28.4	23.7
		(125–250 μm)	22.2	24.9	24.6	22.6	21.9
		(250–500 μm)	22.2	27.7	26.8	20.8	21.9
MCC105	ASS	(90–125 μm)	34.8	37.8	31.2	33.4	15.3
		(125–250 μm)	34.8	30.1	24.9	22.3	12.7
		(250–500 μm)	34.8	28.7	28.1	20.1	10.3
MCC101	ASS	(90–125 μm)	22.5	28.7	23.3	24.8	15.3
		(125–250 μm)	22.5	28.0	22.8	20.9	12.7
		(250–500 μm)	22.5	25.1	24.4	22.3	10.3
MCC102	ASS	(90–125 μm)	22.2	24.8	25.6	23.6	15.3
		(125–250 μm)	22.2	23.3	24.5	22.9	12.7
		(250–500 μm)	22.2	24.7	24.9	24.2	10.3

Therefore, only the maximal possible relative decrease of the initial bulk volume due to tapping (Kawakita constant a) will be discussed, and the values of a for the single powder components and the binary powder mixtures are listed in Table 1. The values of the constant a are not dependent on reaching the minimum bulk volume, because the Kawakita function in the form presented as Equation 1.1 is linear, and the slope of the function will therefore not change.

Fig. 1 compares the change in the value of constant a as a function of the LM content in the binary mixture. Overall trends can be observed neglecting the use of different particle size fractions of LM: first, an addition of LM up to 75% w/w to MCC105 does not significantly alter the value of a. Thus in the case of angular shaped particles between 38 and 250 μm being the second component of a binary mixture with MCC105, the bulk properties of the cellulose product determine the bulk properties of the binary mixtures. Secondly, using MCC101, an addition of LM increases the value of a polynomially, i.e. with increased concentration of the LM particles their bulk properties become more important for the behaviour of the binary mixtures. Thirdly, this is also partly true if MCC102 is used instead of MCC101. However, the sigmoidal shape of the trend function suggests that at high concentrations of LM above about 70% w/w the bulk properties of this excipient are predictive for the bulk properties of the binary mixtures.

In Fig. 2, the influence of the particle size of the LM component in binary mixtures containing microcrystalline cellulose is studied. Generally, for all three particle size fractions it can be seen, that up to a concentration of 50% w/w LM the value of the constant a is more or less similar, and

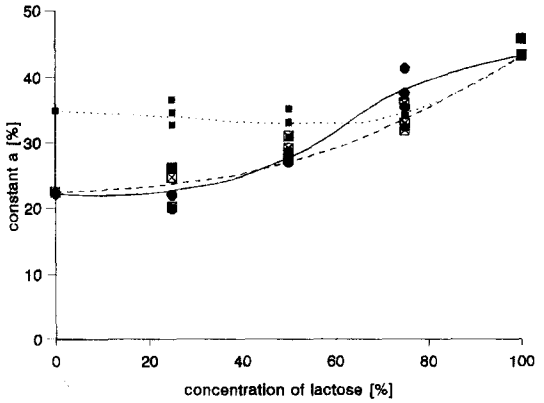

Fig. 1. Kawakita constant a [%] as a function of the concentration of LM irrespective of particle size in binary mixtures with microcrystalline cellulose. ■, MCC105; ⊠, MCC101; ●, MCC102; trend function based on MCC105: dotted line; trend function based on MCC101: dashed line; trend function based on MCC102: full line.

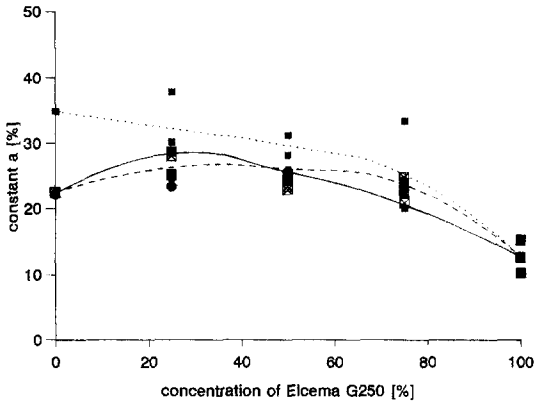

Fig. 3. Kawakita constant a [%] as a function of the concentration of G250 irrespective of particle size in binary mixtures with microcrystalline cellulose. ■, MCC105; ⊠, MCC101; ●, MCC102; trend function based on MCC105: dotted line; trend function based on MCC101: dashed line; trend function based on MCC102: full line.

above this concentration the value of a increases proportionally to the further amount of LM incorporated. For the largest particle size fraction of LM, the values of a are initially smaller than for the two finer powders. This suggests that the large difference in particle size between the two components of the binary mixtures and the spherical shape of the MCC102 particles lead to additional pore volume as reported by Lam and Nakagawa (1994), and thus resistance to close packing. Combining the results obtained from Figs. 1 and 2, it appears that for binary mixtures of angular and fibrous powders only the particle size of the fibrous component affects the properties of the powder mixtures.

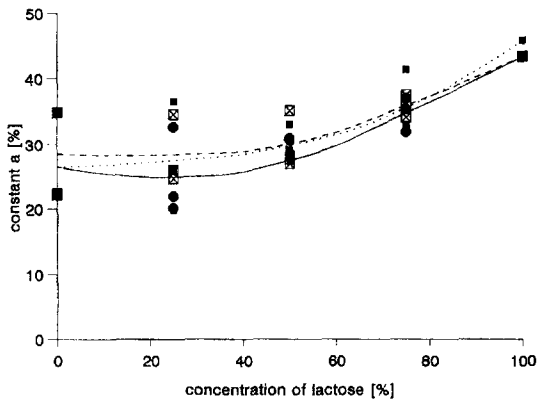

Fig. 2. Kawakita constant a [%] as a function of the concentration of LM in binary mixtures with microcrystalline cellulose, irrespective of grade of microcrystalline cellulose. ■, LM 38–90 μm; ⊠, LM 90–125 μm; ●, LM 125–250 μm; trend function based on LM 38–90 μm: dotted line; trend function based on LM 90–125 μm: dashed line; trend function based on LM 125–250 μm: full line.

Fig. 3 compares how the value of the constant a changes if drawn as a function of the G250 concentration in the binary mixtures. Using MCC105, the value of the constant a decreases, following a slightly polynomial function, with an increase in G250 content. Due to the spherical shape of the G250 particles, the pore space between these particles is generally larger, but high concentrations of the fine MCC105 component can fill these pores and thus increase the packing as reflected in higher values of a. Using either MCC101 or MCC102, the values of a show a maximum between 25% and 50% w/w of G250 added. Above 50% w/w of G250, the value of the constant a decreases proportionally to the increase in G250 concentration. However, the initial values of a are lower if the spherical MCC102 has been used. Packing of binary mixtures of monosized spherical particles, of which one fraction is coarse and the other is fine, should always result in a maximum in packing at about 70% to 80%

w/w of the coarser powder in the mixture, but with broadening particle size distributions this maximum is usually distorted to higher concentrations of the finer powder component (Lam and Nakagawa, 1994), as this can be seen from the graph.

The influence of particle size of the G250 component on the change in the value of the constant a obtained for binary mixtures with microcrystalline cellulose is investigated in Fig. 4. For the two smaller size fractions of G250, there exists a maximum at 25% w/w of G250 in the binary mixtures. Above this concentration, the values of a decrease proportionally with the increased concentration of G250. Furthermore, for the medium size fraction of G250, the packing properties of a binary mixture containing 75% w/w of this powder are similar to those of the powder component itself. This is also valid for the largest size fraction of G250. However, in the latter case the value of the constant a does not change up to 50% w/w of G250, thus implying that up to this concentration the packing properties of MCC102 dominate the packing behaviour of the binary mixtures. Thus, combining the informations obtained from Figs. 3 and 4, it appears that for binary mixtures composed of spherical and fibrous or only spherical particles the packing properties depend on the particle size of either component of the mixture.

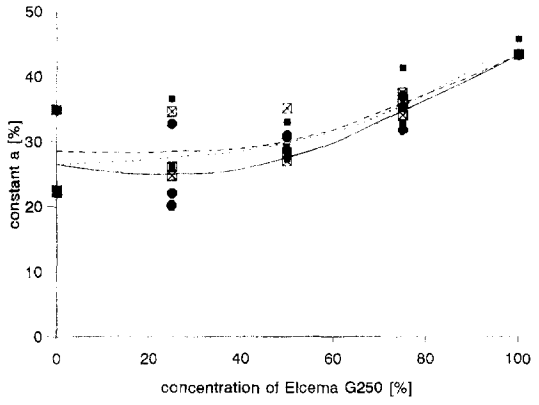

Fig. 4. Kawakita constant a [%] as a function of the concentration of G250 in binary mixtures with microcrystalline cellulose, irrespective of grade of microcrystalline cellulose. ■, G250 90–125 μm; ⊠, G250 125–250 μm; ●, G250 250–500 μm; trend function based on G250 90–125 μm: dotted line; trend function based on G250 125–250 μm: dashed line; trend function based on G250 250–500 μm: full line.

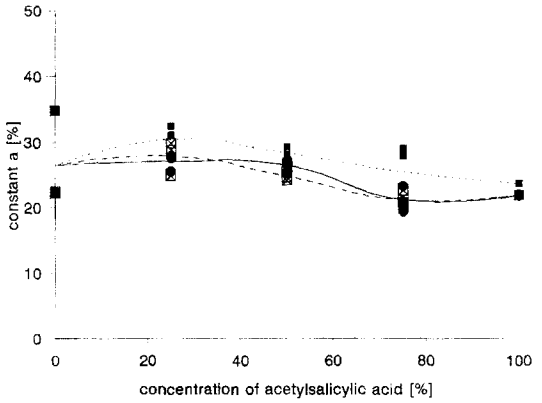

Fig. 5. Kawakita constant a [%] as a function of the concentration of ASS irrespective of particle size in binary mixtures with microcrystalline cellulose. ■, MCC105; ⊠, MCC101; ●, MCC102; trend function based on MCC105: dotted line; trend function based on MCC101: dashed line; trend function based on MCC102: full line.

Fig. 5 compares the values of the constant a obtained for binary mixtures containing ASS and different grades of MCC. Using MCC105, the addition of ASS decreases the value of a proportionally. However, the values determined are not related to the constant a obtained for ASS alone, represented by a significant drop of the trend function above 75% w/w ASS. This suggests that the packing properties of MCC105 are more reflected in the binary mixtures than those of ASS. Using MCC101, the values of the constant a are similar over the whole range of mixture concentrations tested, but they are higher than the values of a of both single components. Compared with the results determined using MCC105, this suggests that the fibrous or needle shape of particles of both components in a mixture is only a hindrance for packing above a certain particle size, and if the mean particle size and size distribution of the two components of the mixture is very different. As soon as one of the needle-shaped or fibrous components is replaced by a powder of different shape, the second particle shape determines the packing properties, as can be seen from Fig. 5 for MCC102.

From Fig. 6 it can be seen that the packing properties of the needle shaped ASS dominate the bulk behaviour of the binary mixtures more, the larger the particle size of the second powder component is. Hence, the particle size of both mixture components proved an important property in these binary mixtures.

4. Conclusions

The particle shape of each single component of binary powder mixtures influences the maximum volume reduction due to packing, represented by the Kawakita constant a. While angular particles improve the packing properties of microcrystalline celluloses, the addition of spherical and needle-shaped particles generally results in lower values for a at equivalent concentrations. The addition of both spherical or needle-shaped particles in concentrations between 25% and 75% to fine grade microcrystalline cellulose leads to a steady decrease in volume reduction ability, whereas for medium or coarse grade microcrystalline cellulose the value of a as a function of concentration of the second mixture component first increases and at higher concentrations decreases, thus showing a maximum value. The particle size of both powders contained in a binary mixture influences the values of the Kawakita constant a with the exception of angular particles. The finer the particle size of each component, the larger is the value of a.

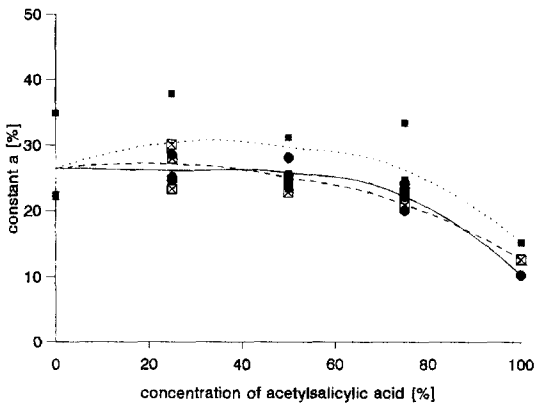

Fig. 6. Kawakita constant a [%] as a function of the concentration of ASS in binary mixtures with microcrystalline cellulose, irrespective of grade of microcrystalline cellulose. ■, ASS 90–125 μm; ⊠, ASS 125–250 μm; ●, ASS 250–500 μm; trend function based on ASS 90–125 μm: dotted line; trend function based on ASS 125–250 μm: dashed line; trend function based on ASS 250–500 μm: full line.

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