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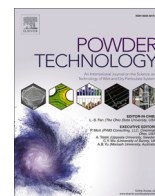
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Review

## A century of granular packing models

G. Roquier\*

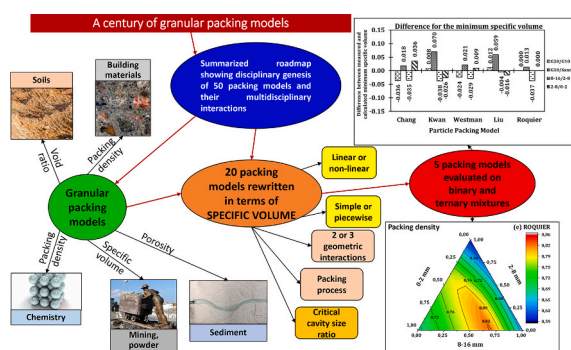
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## HIGHLIGHTS

- Granular packing models developed over the last 100 years reviewed.
- Roadmap summarising the shaping of models using interdisciplinary approaches.
- Main models rewritten in terms of specific volumes for easier comparison.
- Agreement with certain packing mechanisms discussed.
- Five models analyzed on binary and ternary mixtures of coarse and fine aggregates.

## GRAPHICAL ABSTRACT



## ARTICLE INFO

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## ABSTRACT

This article provides a review of particle packing models. A summary roadmap shows their disciplinary genesis and the multidisciplinary interactions that have shaped them over a 100-year period. Described by field, their development has led to models that can be simple or piecewise, linear or non-linear, involving two or three geometric interactions, and possibly the packing process and a critical cavity size ratio. To facilitate comparisons, around twenty of them are rewritten in terms of specific volume. Their agreement with certain packing mechanisms and their extension to multi-sized mixtures are discussed. Finally, five of these non-linear packing models are applied to binary mixtures of aggregates. The analysis shows that they perform well, both in terms of estimating minimum specific volumes and the combinations needed to achieve the optimum. Two of them stand out slightly from the rest. The comparison is then extended to ternary combinations of aggregates or spheres.

## 1. 100 years of history

The volume occupied by a material composed of several granular classes is of interest to a large number of sectors: ceramics, powders, chemistry, soils, sediment research, superalloys, construction materials. To characterize it, increasingly sophisticated packing models have been developed for nearly 100 years, spurred on by the pioneering work of

Furnas in 1928 [1]. Granular packing models have reached such a degree of maturity that they have contributed to the emergence of new composite materials, such as ultra-high performance concretes [2], ceramic additive manufacturing [3] or new superalloys [4]. In the context of optimization, as is the case for these materials, it is rather the packing density that is used. But other disciplines often involve void ratio, porosity or specific volume as part of their specific developments.

Since the need to accurately estimate the volume occupied by a

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Nomenclature			
<i>List of main symbols</i>		$S(\mathbb{V})$	Configurational entropy in a given total volume
$a$	Loosening effect coefficient	$V$	Specific volume
$B$	Filling coefficient in Han's and Wu's models	$V_1$	Specific volume of a binary mixture when the coarse granular class is dominant
$b$	Wall effect coefficient	$V_2$	Specific volume of a binary mixture when the fine granular class is dominant
$c$	Wedging effect coefficient in Kwan's model	$X$	Macroscopic characterization of the state of compaction of a system
$D_r$	Relative density	$y_1$	Volume fraction of the coarse class by reference of the total solid volume
$\bar{d}$	Effective dominant size in Chang's model	$y_2$	Volume fraction of the fine class by reference of the total solid volume
$d_1$	Diameter of a coarse particle in a binary mixture	$z$	Parameter in Toufar's and Goltermann's models
$d_2$	Diameter of a fine particle in a binary mixture	$z_0$	Parameter in Goltermann's model
$e$	Void ratio	$\beta_{12}$	Quadratic coefficient of the binary synergism in Yu's model
$e_{min}$	Minimum void ratio	$\gamma_{12}$	Cubic coefficient of the binary synergism in Yu's model
$e_{max}$	Maximum void ratio	$\eta$	Granular activity coefficient in Chang's model
$f(r)$	Interaction function describing the loosening effect or the filling coefficient depending on the model	$\lambda_{12}, \lambda_{21}$	Crowding factors in Mooney's model
$g(r)$	Interaction function describing the wall effect or the embedding coefficient depending on the model	$v_0$	Initial specific volume in Yu's model
$G$	Parameter in the conic equation of Westman	$v_1$	Specific volume of the coarse class
$K$	Compaction index describing the packing process efficiency in de Larrard's and Roquier's models	$v_2$	Specific volume of the fine class
$k_d, k_s$	Factors in Toufar's and Goltermann's models	$v'$	Virtual specific volume of the coarse class in de Larrard's and Roquier's models
$\ell$	Rod length in Farr's model	$v''$	Virtual specific volume of the fine class in de Larrard's and Roquier's models
$m$	Number of layers of fine particles required to fill the voids of the coarse particles in Han's and Wu's models	$\mathcal{V}'_1$	Virtual specific volume of a binary mixture when the coarse class is dominant in de Larrard's and Roquier's models
$m$	Spacing factor in Dewar's model	$\mathcal{V}'_2$	Virtual specific volume of a binary mixture when the fine class is dominant in de Larrard's and Roquier's models
$n$	Porosity	$\phi$	Packing density
$P_2$	Probability for a coarse particle to be in contact with a fine particle in Han's and Wu's models		
$R_N$	Ratio of the numbers of fine and coarse particles in Wu's model		
$r$	Fine/coarse diameter ratio		
$r_0$	Critical ratio of entrance or critical cavity size ratio		

granular material is a problem common to several sectors of activity, two major consequences have emerged over time:

- i. These models have points in common, because the problem to be solved is not very different from one discipline to another.
- ii. These common points are partially hidden because each sector uses its own language and notations to characterize the packing (packing density, void ratio, porosity, specific volume) and granular classes.

The aim of this paper lies in reviewing the mathematical models proposed in the literature for granular packing models within a common framework using specific volume as a reference, thereby highlighting their assumptions, similarities and differences, strengths and weaknesses. A summarized roadmap, presented in Fig. 1, illustrates the disciplinary genesis of the different models and their multidisciplinary interactions.

Several types of models have been proposed over the decades: basic models illustrating certain boundary conditions, linear packing models with respect to the solid volume fractions, non-linear packing models, piecewise models combining linear and non-linear, models based on a statistical or thermodynamic approach and mineralogical-inspired models based on elementary reference cells with the existence of cavities. A careful review of all these modeling approaches is proposed in this study.

However, a particular emphasis will be placed on models with granular interaction functions as this is the most developed version at present. The first models were relatively basic, involving for each granular class its volume fraction and a characteristic value of its packing. Next, the geometric interactions existing between granular

classes were highlighted and taken into account as a function of the diameter ratio. In the case of binary mixtures, most of these models agree to distinguish the case of dominant fine particles and the case of dominant coarse particles, which leads to consider two different types of interaction functions. Finally, a further step has been taken by integrating the particle shape, the packing process or an additional granular interaction in order to improve the estimation of packing density, porosity, void ratio or specific volume of the mixture.

A table will then summarize the general organization of the main models presented, including their linearity or non-linearity, their compliance with three boundary conditions relating to binary mixtures of very contrasting sizes or of the same size, their parameters and their extension or non-extension to multi-sized mixtures.

Finally, based on a reference publication already used twenty years ago for a similar purpose, five of the models meeting the above-mentioned boundary conditions (some under certain conditions) and which have not yet been evaluated on these data will be.

The remainder of this paper is organized as follows. In section 2, the approaches adopted in around fifty different models are described by disciplinary field. The assumptions put forward are in fact closely linked to the history of research carried out in the same branch. However, transversal approaches are also favored by certain authors who thus scientifically enrich their work. They will be highlighted in this paragraph. In sections 3, 4, 5, the equations of the main packing models in the case of binary mixtures will be presented, thus making it possible to clearly distinguish linear from non-linear models with respect to the solid volume fractions. All equations are rewritten in terms of specific volume in order to be able to compare them. Section 3 concerns limiting cases, section 4 linear packing models, section 5 non-linear packing

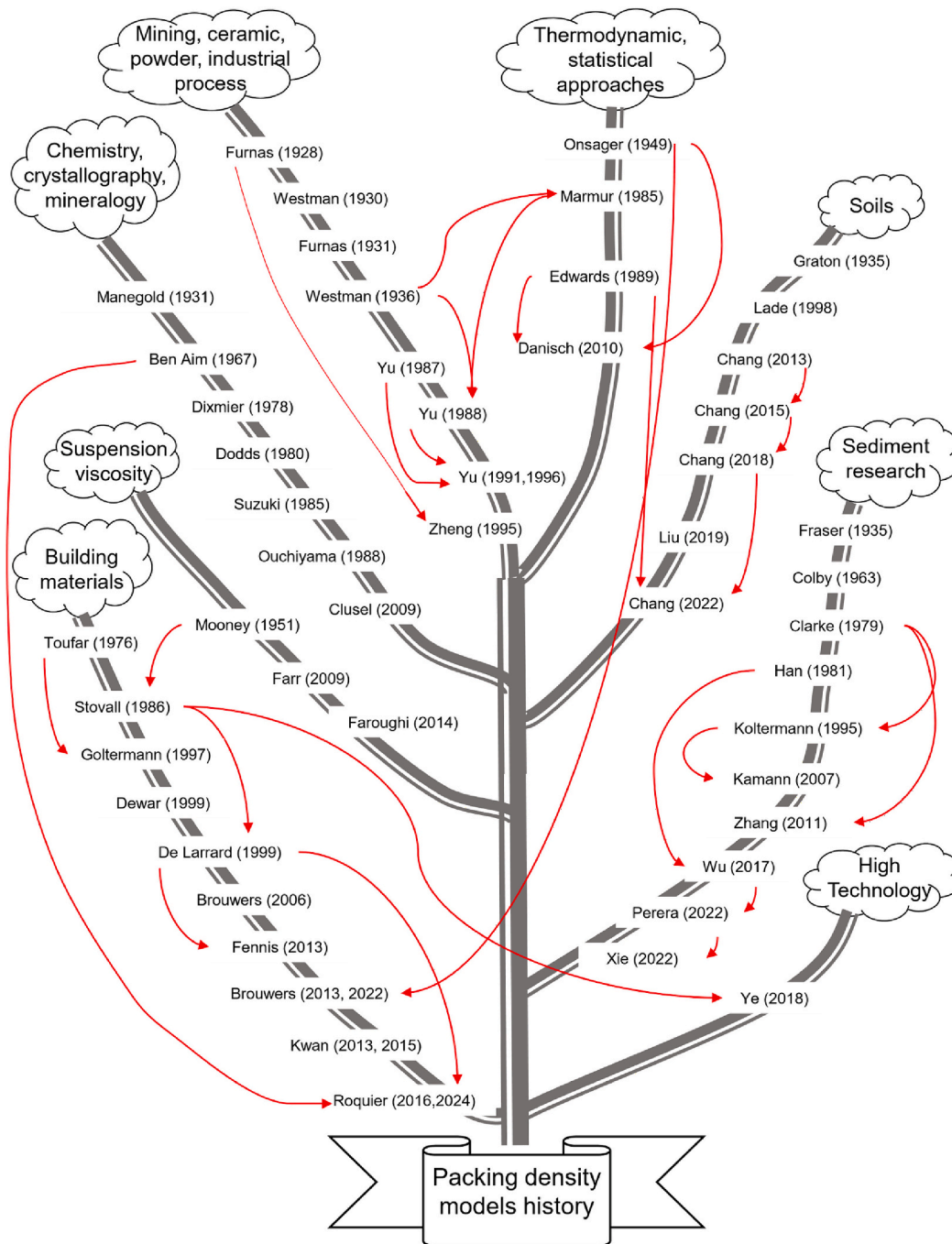


Fig. 1. Summarized roadmap showing the disciplinary genesis of the different packing density models and their multidisciplinary interactions over the years.

models. In section 6, a categorization of these mathematical models is provided. In section 7, five of these particle packing models will be compared on binary and ternary mixtures of aggregates. In section 8, a conclusion will be made.

## 2. Development of packing models according to disciplinary fields

### 2.1. Definitions

Considering a total volume  $V_t$  that contains solid particles of volume  $V_s$  and void volume  $V_v$ , such as  $V_t = V_s + V_v$ , the packing density  $\phi$ , the specific volume  $V$ , the void ratio  $e$  and the porosity  $n$  are given as:

$$\phi = \frac{V_s}{V_t} \quad (1)$$

$$V = \frac{V_t}{V_s} \quad (2)$$

$$e = \frac{V_v}{V_s} \quad (3)$$

$$n = \frac{V_v}{V_t} \quad (4)$$

In addition, three concepts are simply introduced on a graph representative of the specific volume of a binary mixture  $V$  as a function of the volume fraction of the fine class  $y_2$  (Fig. 2): the *filling mechanism*, the

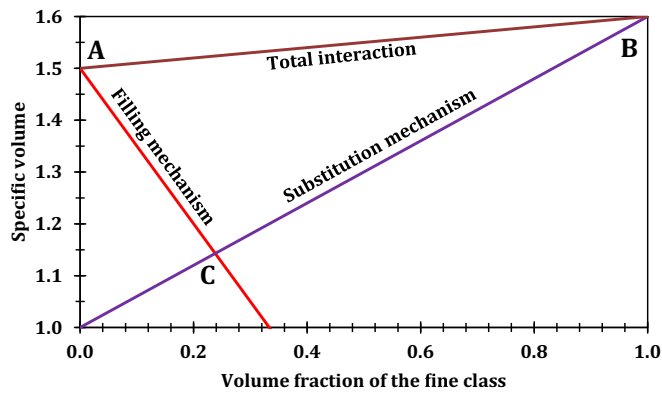


Fig. 2. Illustration of the *filling mechanism*, the *substitution mechanism*, the *total interaction* on a representative diagram of the specific volume for a binary mixture.

*substitution mechanism*, the *total interaction*. They will be explained later.

Finally, it should be noted that the assumptions of constitution of the various models are provided in section 2, but that the most representative equations are approached only in section 3 for the *total interaction* and the *filling and substitution mechanisms*, in section 4 for linear packing models and in section 5 for non-linear packing models.

## 2.2. Mining, ceramic, powder, industrial process

Modern packing density models owe much of their development to the early work of Furnas [1,5], and Westman [6,7], which later led to the development of linear and non-linear packing models [8–14], the two being sometimes associated like that of Yu [9].

Furnas, who was a physical chemist with the US Bureau of Mines in Minneapolis, worked out mathematical relations between the specific volume and the size composition in systems of broken solids. The first of his papers deals mainly with binary mixtures while the second extends the proposed theory to multicomponent systems. Furnas observes that a mixture of spherical particles passes through a minimum of porosity when the composition of the mixture varies: the minimum is all the more pronounced as the size contrast is important. In this case, he establishes that the small particles fill the cavities of the coarse particles such as the total volume of the mixture does not increase. In the case of the dominant fine particles, the principle consists in substituting some of them, separated by voids, by a small amount of spread coarse particles consisting of matter. The specific volume curve of the mixture  $V$  expressed as a function of the volume fraction of one of the two classes then breaks down into two straight lines (Fig. 2): one corresponds to the *filling mechanism* (segment AC) when the coarse particles are dominant, the other corresponds to the *substitution mechanism* (segment CB) when the small particles are dominant. Note that the line AC, once extended, would pass through the point of coordinates (1, 0). Without being aware of Furnas' publication, Westman & Hugill [6] offer an analogous interpretation of the observed minimum porosity. They called the segment AB (Fig. 2) the specific volume of the constituents before mixing: the *interaction* is said to be *total*. A few years later, Westman [7] proposed a very clever conic equation intended for binary mixtures allowing, thanks to a single parameter  $G$ , to cover all possible intermediate diameter ratios. The latter being defined by  $r = d_2/d_1$  ( $d_2 \leq d_1$ ),  $G$  indeed evolves between  $G = 1$  for  $r = 1$  and  $G \rightarrow \infty$  for  $1/r \rightarrow \infty$ . In the first case, the specific volume of the mixture is represented by a straight line connecting the points representative of the specific volumes of each component in mono-dispersed packing state. In the second case, the specific volume of the mixture is represented by the straight lines corresponding to the *substitution mechanism* or the *filling mechanism*. Many years later, Yu et al. [8] will propose a general relationship between  $G$  and  $r$ , applicable to both binary mixtures of spherical and non-spherical

particles.

Nevertheless, it is another model which will constitute the most important contribution of Yu et al. in the field of packings: the “linear-mixture packing model” [9], which is a combination of a “linear packing model” [10] and a “mixture model” [11]. A critical cavity size ratio  $r_0$  (called the critical ratio of entrance by Yu et al.) allows selection of the appropriate model. For spherical particles,  $r_0$  given experimentally by McGeary [12] has the value of  $1/6.5 \approx 0.154$ . If  $r \leq r_0 = 0.154$ , Yu et al. consider that the packing is formed almost exclusively by the *filling mechanism*: the “linear packing model” is then applied. Otherwise, the “mixture model” is used. The “linear packing model” [10] assumes that the specific volume of the mixture is calculated as a linear function of the solid volume fractions of each size class. To each granular class considered dominant in turn, corresponds the calculation of a specific volume of the mixture, the highest value obtained being the real. The “mixture model” [11] includes a term called synergism of the mixture and two coefficients which are respectively the quadratic coefficient and the cubic coefficient of the binary synergism. This model is inspired by an original approach of Marmur [13] who applied to mixtures of solid particles a modified and enriched thermodynamic concept of partial molar volume. The “mixture model” is therefore a cubic “non-linear packing model” with respect to the solid volume fractions.

In 1995, Zheng et al. [14] will update the Furnas model by approaching the calculation of the packing density of a binary mixture from a new angle. Furnas indeed considers that the theoretical maximum packing density is the sum of two terms. The first one corresponds to the packing density obtained when the container is stuffed with the coarse class alone. The second one corresponds to the increase of packing density obtained with the addition of the fine particles between the large ones. It is in this second term that Zheng et al propose to take into account the influence of the volume fraction of fine particles  $y_2$  and of the size ratio  $r$  via two empirical functions.

## 2.3. Chemistry, crystallography, mineralogy

Because the scientific community of chemists, crystallographers and mineralogists focused very early on the spatial arrangement of atoms with a periodic and ordered character, studies focused on reference geometric cells in which it was possible to carry out porosity and coordination calculations. The cells are mostly spherical and sometimes tetrahedral.

Manegold et al. [15] were pioneers to applying classical calculations of crystallography to regular stacks of spheres. Now well-known results on the porosity and coordination of regular packings are set out there. In particular, Manegold et al. highlight a different critical cavity size ratio than that used by Yu et al. [9] to delineate the application conditions of their “linear packing model” and their “mixture model”. If Yu et al. favor  $r_0 = 0.154$  indeed, Manegold et al. willingly refer to:  $r_0 = 0.225$ . The explanation is as follows [16]. In the case of the two regular stacks of coordination 12, the twelve spheres are not arranged regularly around the central sphere. Two kinds of cavities exist: the ones delimited by four, the other delimited by six spherical surfaces. In the first case, the diameter  $d_2$  of the largest sphere that can pass through the restraint between 3 spheres of diameter  $d_1$  is characterized by  $r_0 = 0.154$ . In the second case, the diameter of the largest sphere that can fit into a tetrahedral cavity is characterized by  $r_0 = 0.225$ .

Inspired by the work of Manegold, Ben Aim [16] stated in 1970 that the realization of a binary mixture whose diameter ratio  $r$  is lower than 0.22 provides the possibility to the small grains to lodge in all the cavities formed by the coarse grains. In this situation, he highlights the notion of *eutectic* by establishing a formal analogy with molecular structures and, in particular, with the thermodynamics of binary solutions. When  $r$  is between 0.22 and 0.5, Ben Aim estimates that the distinction between the “dominant coarse particles” and the “dominant fine particles” domains is no longer so clear. However, he believes that the curve  $V$  vs  $y_2$  can be likened to two straight lines with an acceptable



error. In the field of dominant fine grains, Ben Aim then introduced the notion of *wall effect*. He assumes that when a sphere of diameter  $d_1$  is immersed in a bed of spheres of diameter  $d_2$ , the disturbance caused by the wall of the coarse sphere is limited inside a spherical crown comprised between the spheres of diameter  $d_1$  and  $d_1\sqrt{1+2r}$ : this last limit corresponds to the contact points of the first layer of small spheres surrounding the coarse one. If  $d_2 \ll d_1$ , the disturbed zone by the wall effect has a thickness close to  $d_2/2$ . Ben Aim then carries out his reasoning by considering that the packing of small spheres presents two different porosities: the first one in the spherical crown logically higher than the second, the bulk porosity. To summarize, Ben Aim curve  $V$  vs  $y_2$  breaks down into two straight lines: the first one corresponds to the *filling mechanism* when the coarse particles are dominant, the second one takes into account the wall effect when the small particles are dominant.

Ben Aim's thesis also makes the link between porosity and coordination number. This is indeed a recurring concern among chemists. In 1978, Dixmier [17] proposed, in a very rich article, a new description of random packings, based on the Dirac's chord method. By applying it to packings of equal hard spheres, he establishes a one-to-one relation between the packing density and the average coordination number. In 1980, Dodds [18] took a further step in addressing the issue by considering multicomponent random sphere packings. He developed a simple "statistical geometric model" by using tetrahedra subunits whose vertices correspond to the center of each spherical particle, considered in contact with its neighbour. However, Dodds' gapless packing assumption has three undesirable consequences: too high coordination number for identical spheres (13.4 instead of 12 in reality), too low porosity in a regular tetrahedron (0.2204 while it should not be on average <0.2595), a model inapplicable for diameter ratios <0.154 (restraint between three identical spheres).

In 1985, Suzuki & Oshima [19] established a theory to determine the coordination number which will then lead to a model of porosity. They consider four cases of contact for a 2-component mixture consisting of coarse particles 1 and fine particles 2: 1-1, 1-2, 2-1, 2-2. Their approach first uses a coarse sphere of diameter  $d_1$  surrounded by small ones of diameter  $d_2$ . They then determine the coordination number and the porosity in a spherical cell whose diameter,  $d_1\sqrt{1+2r}$ , corresponds to that chosen by Ben Aim to delimit the exterior of his spherical crown. The porosity associated with each granular class is then calculated according to its fractional area in the granular assembly. By multiplying it by the associated volume fraction and by performing the sum for all the granular classes, the porosity of the mixture is finally predicted. During the different stages, the authors take into account one parameter which makes it possible to calibrate the theoretical approach on experimental results: this is the coordination number of a monodisperse medium created under the same conditions of packing process. It should also be noted that where Ben Aim considers a wall effect in a spherical crown, Suzuki & Oshima perform their reasoning in a spherical cell without referring to any particular geometric interaction.

Like Suzuki & Oshima [19], Ouchiyama & Tanaka's considerations [20] began with a simplified model for the coordination number. They put forward two fundamental assumptions called uniformity assumptions. The first one considers that a sphere of diameter  $d$  is in contact with its neighbours having the average diameter  $\bar{d}$ . The second one estimates that the part of the volume shared by several hypothetical spheres is allocated equally to each sphere, regardless of their size. Unfortunately, uniformization makes the overall average porosity too high [21].

In the manner of Ben Aim [16] on the one hand and Suzuki & Oshima [19] on the other, Clusel's "granocentric model" (2009) [22] examines the complexity of the global packing from the point of view of a single particle, whose field of vision would be blocked by the first layer of particles surrounding it. In his statistical approach, Clusel is mainly inspired by the Dodds' model [18] without making the restrictive assumption that all particles are in contact with their neighbours. The

formation of the assumed homogeneous and isotropic packing is described by random processes. A maximum solid angle available around the central particle is introduced as an effective parameter to describe the geometrical congestion prevailing around it. Its value is not simply  $4\pi$  but depends on the polydispersity and the local packing structure. The formation of the neighbour shell is then modeled by the first passage of a one-dimensional random walk, where the steps are the solid angle subtended by the incoming particles on the central particle. As in any effective model, limitations to the applicability of the granocentric theory have been identified. Especially, the polydispersity being the dominant source of disorder in the packing, the model can not be applied to highly monodisperse packings or to bidisperse packings in the "rattler" regime, where particles smaller than the cavities formed by the coarse spheres become mobile inside them.

#### 2.4. Thermodynamic and statistical approaches

Very quickly, chemists who were interested in molecular packings established the link with the thermodynamics of binary solutions. Some researchers have therefore sought to transpose this powerful tool, which highlights the equilibrium states of matter, from physical chemistry to granular media. This transversal approach has become widespread between these two sectors and has given rise to clever and promising theories with the introduction of new concepts in granular packing such as those of excluded volume, eutectic, compactivity, partial particle volume, etc.

Onsager [23] is an illustration of this. The 1968 Nobel Laureate in chemistry made a deep contribution to thermodynamics thanks to his entropic theory of transition to nematic order for rod-shaped colloids. He based his reasoning on two-particle interactions by introducing the notion of excluded volume around them. If two rods approach in the same plane in a non-parallel way, there is a minimum distance between their center of gravity below which the two particles cannot approach without changing their mutual orientation. A phase separation must then take place above a critical volume fraction. These are Onsager's expressions for excluded volume which will later be applied to the study of the packing of binary particles by Brouwers.

Ben Aim [16] has also made a noteworthy contribution to the use of thermodynamics concept in the field of packing. Indeed, he calls *eutectic* the cusp point corresponding to the minimum of porosity according to Furnas' assumptions. He asserts that this phenomenon is analogous to molecular eutectics when the small molecules become lodged inside the pseudo-crystalline network of the coarse molecules. Ben Aim then undertakes to lay rigorous mathematical foundations by defining a partial particle volume by analogy with the notion of partial molar volume of a solution.

Marmur [13] took up this idea. He specifies the physical meaning of the partial particle volume: it corresponds to the increase in the total volume of the packing when one particle is added among many others. This increase in volume is, in general, different of the particle volume itself, since the latter either penetrates into a void or moves other particles to position itself. He also distinguishes between partial particle volume which counts individual particles and partial molar volume which relates to numbers of moles. The only unknown function in the exposed approach being that of the specific volume of the binary mixture, Marmur ends up concluding that the conical equation of Westman [7] is the most appropriate.

The seminal work of Edwards & Oakeshott [24] is referenced now as it will be the inspiration for two packing models which will be described later. Based on statistical mechanics, it concerns the existence of thermodynamic states in a granular system. When the latter is subject to extensive manipulations such as vibration or shear in a given total volume  $\mathbb{V}$ , the microscopic dynamics is controlled by its mechanically stable configurations which are then equiprobable according to the authors. The logarithm of their number is used to define a configurational entropy  $S(\mathbb{V})$ . Edwards & Oakeshott then refer to an associated

state variable  $X$ , naturally defined by  $X^{-1} = \frac{\partial S(V)}{\partial V}$ , by analogy with the canonical definition of temperature in systems at thermodynamic equilibrium. It follows that  $X$  is a macroscopic characterization of the state of compaction of the system. Some successes have been recorded by Edwards' theory, especially in the description of properties of a number of granular systems. The establishment of a phase diagram for jammed matter spanning from the RLP for which  $X \rightarrow \infty$  to the RCP for which  $X = 0$  testifies to this [25]. However, Edwards' proposal requires several clarifications. First, the equiprobability assumption on which this theory is based has already been put in doubt on small systems of bidisperse discs [26] and generally fails at higher densities than that corresponding to the unjamming point for polydisperse soft repulsive discs [27]. Second, the construction of the statistical physics of the problem can be strongly affected by the presence of spatial correlations, sometimes extended over distances comparable to the size of the system [28].

Under the umbrella of Edwards' statistical approach, Danisch et al. [29] also addressed the description of the properties of random assemblies of polydisperse hard spheres. Their analytical process brings together different observations integrating packings from RLP to RCP, the coordination number and the friction. The key aspect seems to be the dependence of the coordination numbers between the different granular species and their concentrations. This result is then incorporated into a statistical theory of volume fluctuations which calculates the free volume of a particle in terms of the coordination number. The volume associated with each particle is provided from a Voronoi tessellation. The mean Voronoi volume is calculated by involving the inverse cumulative distribution of each species which is separated into two contributions: a term taking into account the contact spheres and a bulk term. By working in the limit of a large number of particles and with reference to Onsager [23], Danisch et al. include an excluded volume for the bulk term and an excluded surface for the contact term. Their theory captures well the behavior of bidisperse sphere packings for size ratios  $r$  between 0.5 and 1. However, for  $r < 0.5$ , deviations are found indicating the limit of their approach, which is otherwise relatively difficult to grasp.

## 2.5. Soils

Whether for the study of soil settlements or to better understand certain mechanisms at the origin of disasters such as liquefaction phenomena, the void ratio is a key parameter to be determined, in particular because the contracting or dilating character of a fine natural geomaterial is directly linked to its initial state. This is why soil specialists have deployed great efforts over the years to estimate this quantity, especially in the case of pure sands or sand-fines mixtures. However, in the field of soils, the void ratio is not unique and often refers to an interval defined by a maximum and a minimum, corresponding respectively to a loose or dense state. In this respect, the grain deposition method plays an important role, as many experts have pointed out. They have therefore set up elaborate experimental or simulation protocols, such as the controlled pluviation method, to reconstitute granular samples in well-defined states with the highest level of homogeneity. Unfortunately, packing models are not yet sufficiently refined to incorporate the associated controlled parameters such as the pluviation flow rate or the free-fall height of the grains. While they often refer to the deposition method, model designers have in fact preferred, over the course of developments, to focus on moving from linear to non-linear packing models to improve estimates.

A few years after Manegold et al. [15], Gratton & Fraser [30] presented an excellent review of the geometric principles related to the packing of spheres by defining different arrangements in strict crystallographic terminology. They also meticulously analyzed the many pore structures to study their effects on porosity and permeability.

Lade et al. [31], for their part, emphasized the importance of relative

density  $D_r$ , on the properties and characteristic behavior of granular material,  $D_r$  involving  $e_{min}$ ,  $e_{max}$  and  $e$ , respectively the minimum, the maximum and the void ratio of the soil studied. After specifying that  $e_{min}$  and  $e_{max}$  should depend on the shape of the grain-size distribution curve, the grain shape and the deposition method, they then demonstrated experimentally the important role played by fines content on the structure of a sand, affecting its compressibility and its static liquefaction potential. Referring to the idealized packings of Manegold et al. [15], they carried out an analytical method for estimating the minimum void ratio for spheres of different sizes. Lastly, they took advantage of McGeary's study [12] to underline that a binary mixture is all the more interesting when the diameter ratio  $r$  remains lower than 0.154. This is why the theory they proposed is essentially adapted to mixtures with strong size contrast.

From 2013 and probably well before, Chang will undertake long-term work on granular packings and especially sand-silt mixtures. Very quickly, he will focus his efforts on the notion of dominant grains network (2013,2015) [32,33] and on the concept of dominant size approached as a continuous variable (2018) [34]. Finally, he will choose to draw inspiration from equilibrium thermodynamics by introducing the concepts of "effective particle size" of the packing and "excess free volume potential" by considering the "interaction activity" between the granular species of a packing (2022) [35]. The path followed by Chang is described below.

In 2013, after showing that the mechanical behavior of sand-silt mixtures is mainly governed by the initial void ratio, the way the soil was deposited and the level of effective stress applied, Chang & Meidani [32] distinguished three cases in the inter-granular soil mix classification: fines content  $< 25\%$  where soil matrix is coarse grain dominated, fines content higher than  $35\%$  where soil matrix is fine grain dominated and an intermediate case. Expressed in terms of specific volume in a graph  $V$  vs  $y_2$ , their approach leads to a "linear packing model" made up of two straight lines located respectively above AC and CB (Fig. 2).

In 2015, Chang et al. [33] proposed a slight variation on the previous theory. The packing model is always linear but the analysis of the two equations obtained indicates that they are made up of a common trunk constituted of the terms of the equation representing the upper limit called the *total interaction* (AB, Fig. 2). On the coarse grain dominant side, a term proportional to a parameter called the filling coefficient is then subtracted. On the fine grain dominant side, a term proportional to a parameter called the embedment coefficient is subtracted. The novelty lies mainly in the use of two power functions expressed as a function of the size ratio  $r$  to describe the granular interaction coefficients. The filling exponent  $p$  and the embedment exponent  $s$  are determined from test results.

In 2018, Chang & Deng [34] reached a new milestone by introducing the concept of effective dominant skeleton. In a "linear packing model" for binary mixture, the dominant packing skeleton is composed of particles of size  $d_1$  on the side of the dominant coarse grains and particles of size  $d_2$  on the side of the dominant fine grains. In their "non-linear packing model", Chang & Deng assumed that the mixture is composed of a multitude of particle classes ranging in diameter between  $d_2$  and  $d_1$ . Only these two extreme sizes correspond to real granular classes. The others are fictitious, which means that their solid volume fraction is zero, but they nevertheless interact with each other via the granular interaction coefficients always in power-law. The dominant size  $\bar{d}$  is then effective and can be treated as a continuous variable.  $\bar{d}$  is determined by writing that the derivative of the specific volume or the void ratio of the mixture is zero with respect to this variable.

In 2019, Liu et al. [36] developed an alternative "non-linear packing model". In reality, it consists in a piecewise model based on an ideal scenario and a general scenario. The ideal scenario corresponds to that of Furnas [1]. The general scenario considers that the specific volume is a quadratic function of the solid volume fraction of each granular class. For a multi-sized mixture, each component successively acts as the

dominant size class while the others are non-dominant. To each attempt corresponds a specific volume of the mixture, the real value being the highest. Like Chang et al. [33,34], the granular interaction coefficients are termed as filling and embedment coefficients. They are depicted by a power function of the size ratio  $r$  but they require the calibration of four parameters:  $A$ ,  $B$ ,  $p$ ,  $s$ , the last two being respectively the filling and embedment exponents.

Finally, in 2022, Chang & Deng [35] completely redefined their 2018 packing model [34] on a thermodynamic basis, limiting themselves however to binary mixtures. They used the partial particle volume introduced by Ben Aim [16] and taken up by Marmur [13], that they called volume potential. An “excess free volume potential” is defined for each granular species and makes it possible to deduce that of the bi-dispersed packing. The granular interaction coefficients and the filling and embedment exponents  $p$  and  $s$  of the 2018 packing model are respectively replaced by granular activity coefficients and material exponents  $\eta_1$  and  $\eta_2$  in the 2022 packing model. The *effective particle size*  $\bar{d}$  is considered as an internal state variable representative of the packing configuration. By considering  $\eta = \eta_1 = \eta_2$ , the principle of minimum excess free volume potential is used to determine the activity coefficients. As for the 2018 packing model, the approach amounts to finding the value of  $\bar{d}$  for which the derivative of the specific volume of the mixture is zero.

## 2.6. Sediment research

The reasons that led to the need to estimate the porosity of sediments have evolved over the years. Historically, it was directly linked to the sedimentation engineering which had to convert mass to volume for sediment deposits. At the time of energy challenges, it can also have a direct relationship with the extraction and/or storage of fluid or gas in carbonated sediments.

As Manegold [15] four years before him, Fraser [37] began by studying the ideal packings of spheres. He refers in particular to the critical ratio of entrance, which will then be taken up by Yu & Standish [9,11], before focusing experimentally in two-component and in multi-component systems. Above all, he is one of the first to study the impact on porosity of the deposition method with or without compaction.

In 1963, Colby [38] took another step by proposing a method for calculating the porosity of sediment mixture by dividing it into multiple granular classes. Expressed in terms of specific volume, it corresponds to the upper limit eq. AB (Fig. 2), already described by Westman & Hugill [6].

In 1979, Clarke [39] developed a reasoning based on porosities varying linearly with solid volume fractions and therefore on specific volumes varying non-linearly with them. However, a comparison of experimental data to estimations shows an underestimation of porosity for all sediment mixtures [40].

This is why Koltermann & Gorelick [40] developed the “Fractional Packing Model”. When the fine volume fraction is lower than the coarse class porosity, they consider that a fraction of fine grains are distributed in the pore space of the coarse skeleton, while the other fraction displaces the coarse particles. Similarly, when the fine volume fraction is greater than the coarse class porosity, the mixture porosity value is greater than that estimated by the ideal model [39]. In both cases, they include a new coefficient called  $y$  which translates the relative proportions of coarse and fine packings.  $y$  depends of  $y_{min}$  which is the  $y$  value occurring when the fine volume fraction equals the porosity of the coarse component. Values of  $y_{min}$  are provided in a table by the authors as a function of a confining pressure. However, the “Fractional Packing Model” has three drawbacks. First, it assumes that the fines grains are smaller than the cavities of the coarse skeleton, which is too restrictive as assumption for most natural sediments. Second, the physical meaning of  $y$  is not very clearly spelled out. Third, the minimum porosity does not occur at  $y_{min}$  as it should, when the latter is very small [41].

Kamann et al. [41] proposed an evolution of the previous model which led to a piecewise-linear function for the porosity. This one involves the porosities of the coarse and the fine components, the pre-mixed fine volume fraction, the fine volume fraction which coincides with the porosity of the coarse component and the corresponding minimum porosity. However, the “piecewise-linear model” for porosity does not perform better than the “Fractional Packing Model” for mixtures with low fine fractions [42].

Zhang et al. [43], meanwhile, chose to use Clarke’s ideal-mixture equations [39] as a starting point. They completed their reasoning by defining two additional types of mixtures based on porosity: zero-mixing when the two components are packed without mixing with each other and partial mixing which is an intermediate between zero-mixing and Clarke’s ideal mixing. They then introduced a mixing coefficient  $\lambda$  which varies between 0 for zero-mixing and 1 for ideal mixing. The value of  $\lambda$ , determined experimentally, is dependent on the mixing condition. In terms of porosity, each curve provided by the model is composed of two segment lines, one decreasing and one increasing, separated by a point representative of the minimum porosity. Zhang’s model was validated on binary mixtures composed of a filtration sand as fine component and glass beads as coarse component. The size ratio range tested [43] varies between  $r = 0.0035$  and  $r = 0.3$ , which means that the span between 0.3 and 1 is not explored.

Only two years after Clarke [39], Han et al. [44] opened a new original path. They developed a “random filling theory”, based on configurations in 2D and in 3D. The equation proposed by Han involves a filling coefficient  $B$  which represents the proportion of the coarse class participating in the increase of packing density.  $B$  is expressed as a function of two terms:  $P_2$  raised to the power  $m + 1$ .  $P_2$  corresponds to the probability for a coarse particle to be in contact with a fine particle in 3D.  $m$  corresponds to the number of layers of fine particles required to fill the voids enveloping the coarse particles in 2D.

In 2017, Wu & Li [42] opted for Han’s “random filling theory” framework [44]. They adapted the  $B$  term to a 3D configuration, the centers of the coarse spheres being located at the vertices of tetrahedrons.  $m$  is expressed as an affine function of the size ratio  $r$ . The filling condition is respected when  $r \leq 0.225$  which is consistent with the concept of tetrahedral cavity. According to the authors, the covering probability of a coarse particle by finer ones is expressed as the ratio of the number of small particles available to the required one. However, when the first number is greater than the second, an upper bound  $B_{max}$  is set for  $B$ . In this case indeed, the coarse particles begin to be dispersed among the small ones. The theory would like  $B$  to reach the maximum value of 1 but the authors prefer to set a limit located around  $B_{max} = 0.85$ . Wu & Li [42] applied their model to sand-clay and pebble-sand mixtures, each of them presenting very contrasting sizes since  $r$  varies between 0.007 and 0.06.

In 2022, Perera et al. [45] proposed to improve the previous model by extending it to more general packings, without limiting them to the concept of filling. In particular, they consider that the model must include rearrangement of all the particles in the mixture and must be extended to trimodal mixtures, by focusing on a semi-empirical approach. The filling coefficient  $B$  and the packing model become respectively the packing coefficient and the “random particle packing model”.  $B$  involves now  $P_2$ ,  $m$ , the fraction of coarse particles and two coefficients  $\lambda$  and  $\mu$ , related to the size ratio  $r$ .  $m$  is determined as the same way as [42], but now has a lower bound at 0.5 when  $r > 0.225$ .  $\lambda$  and  $\mu$  are respectively expressed as a function of  $a_i$ ,  $b_i$  and  $a_\mu$ ,  $b_\mu$ . These four coefficients are obtained by regression analysis on different types of particles. The analysis of the packing function  $B$  as a function of  $r$  reveals three categories of filling efficiency:  $r \leq 0.1$ : filling without loosening of the coarse skeleton;  $r > 0.225$ : no filling;  $0.1 < r \leq 0.225$ : transition between filling and no filling. Trimodal mixtures consisting of fine F, medium M, coarse C particles are treated using three approaches. The first two consider that two classes form clusters before interacting with the third: CM-F and FM-C approaches. The third one considers that all



classes randomly interact with each other: F-M-C approach. It is found that the F-M-C and the FM-C approaches are the better.

Xie & Hu [46] then extended the “random particle packing model” to multi-sized mixtures by developing a multi-stage normalization. The basic idea consists in combining multiple particle size groups into binary and ternary subsystems. Each of them is then converted into a set of representative data including fraction, particle size and porosity. This representative information is then used as initial data for the next stage. The particle size groups are continuously reduced in number until the target porosity is achieved.

### 2.7. Suspension viscosity

The rheological modeling of concentrated suspensions often calls upon the concept of relative viscosity in a Newtonian solvent. Its mathematical expression takes into account the divergence of the viscosity which occurs when the solid volume concentration reaches the most compact packing. A liquid – solid transition occurs and the suspension becomes a packing. This is why specialists in the viscosity of concentrated suspensions were very early confronted with the problem of estimating the packing density of mixtures with several particle sizes. Some models even conceal little treasures.

It seems the case with Mooney’s viscosity model (1951) [47] which contains a packing model that went unnoticed at first sight. Mooney introduced crowding factors in a self-consistent model. They are denoted:  $\lambda_{12}$  and  $\lambda_{21}$ . For spheres of very contrasting sizes  $d_1 \gg d_2$ , the suspension of the small spheres in the liquid between the coarse ones behaves towards these latter as a homogeneous liquid of increasing viscosity. Therefore, the coarse elements are not disturbed by the small ones. Mooney therefore writes  $\lambda_{12} = 0$ . By elsewhere, by reasoning in a total volume unity, the volume accessible to the fine fraction  $\phi_2$  is equal to:  $1 - \phi_1$ , where  $\phi_1$  corresponds to the volume fraction of the coarse class. Mooney then deduces:  $\lambda_{21} = 1$ . He makes no attempt to determine the functions  $\lambda_{12}(r)$  and  $\lambda_{21}(r)$  but he proposes a curve shape representative of  $\lambda$  as a function of the logarithm of the size ratio  $r$ . In terms of specific volume of a binary mixture, Mooney’s theory provides a curve  $V$  vs  $y_2$  which breaks down into two straight lines : the equations involve  $\lambda_{12}$  and  $\lambda_{21}$  respectively.

Sixty years later after Mooney, Faroughi & Huber [48] took up the crowding factor concept in order to study interfering size ratios. They define power-law interaction functions of particle size ratio  $r$ , that they called contracting factors. Without necessarily establishing a direct link, functions of the same type will be taken over by Chang et al. [33–35].

Farr & Groot [49], for their part, developed a theory for the close packing density of hard spheres of arbitrary size distribution, based on a clever mapping onto a 1D problem. They constructed a normalized distribution  $P_{1D}(\ell)$  of rod lengths  $\ell$  from any number-weighted diameter distribution  $P_{3D}(d)$ .  $P_{1D}(\ell) d\ell$  represents the number fraction of rods with lengths between  $\ell$  and  $\ell + d\ell$ . From this distribution, a set of rods are placed sequentially on a line, from the longest on the left to the shortest on the right. They do not touch each other, but are positioned so as to provide a space whose length is at least equal to a fraction  $f = 0.7654$  of the shorter of the two rods concerned. This value has been selected because it leads to a packing density of 0.6435 for monodisperse spheres. In the sequence, the longest remaining rod is inserted into the largest gap available, which may require widening the latter by moving all the rods to the right of the space. If more than one placement option is available, the position chosen is the leftmost one. In this process, periodic boundary conditions in 1D are maintained. At the end, the rods occupy a length fraction  $\phi_{RP}$  on the line. The RP estimate is considered to be the actual RCP volume fraction of the original spheres in space. This model was then tested on extensive simulations for mixtures of elastic spheres with hydrodynamic friction. For the distributions studied, the authors obtained an excellent agreement between theory and simulation. The fact remains that it is above all an algorithm reserved for spherical particles.

### 2.8. Building materials

The construction sector has contributed to the evolution of packing density models thanks in particular to the appearance of high-performance concrete. Until then, concretes were formulated using reference granular curves. But the addition of a supplementary constituent, silica fume, made some of the latter obsolete and was at the origin of new studies on granular packings.

Toufar et al. [50] were pioneers in the field of non-linear packing models. They consider the following three limiting cases:  $r \rightarrow 1$ ,  $r \rightarrow 0$  on the side of the dominant coarse grains,  $r \rightarrow 0$  on the side of the dominant fine grains, before using a single specific volume equation which is suitable for diameter ratios  $r > 0.22$ . This formula is that of the *total interaction* reduced by a term taking into account two parameters. The first  $k_d$  takes into account the fact that fine particles are too large to fit into the available caverns without loosening the coarse particles:  $k_d$  must therefore depend on the particle size ratio. The second  $k_s$  is a statistical factor taking into account the probability linked to the amount of interstitial sites available in the coarse granular skeleton and by considering that each fine particle is surrounded by four coarse particles.

In 1997, Goltermann et al. [51] proposed to make a minor correction to the previous model in the  $k_s$  expression after noting that the packing density of a sample composed of coarse particles does not increase sufficiently when a small quantity of fine particles is added.

Dewar [52], for his part, focused on the formulation of concretes by developing a general theory of particle mixtures involving the analysis of void ratios. His approach encompasses five main steps. First, he creates a 3D composite element consisting of a particle and a void associated with it, both of which are modeled by cubes. Second, by keeping the same pattern, he considers that a binary mixture of fine (dimension  $d_2$ ) and coarse (dimension  $d_1$ ) particles dilates the structure of the latter, each coarse particle being separated from a congener by a distance equal to  $m d_2$  where  $m$  is a spacing factor. Third, he assumes that the evolution of the void ratio of a binary mixture as a function of the volume fraction of the fine particles  $y_2$  can be represented by six points, denoted from A to F, connected by a series of straight lines with change of slopes rather than with a curve. Fourth, each of the six points is associated with a particular value of  $m$ . Fifth, besides the available space within the dilated structure of the coarse particles, an additional space due to particle interference is added and calculated thanks to a factor  $Z$ . The latter is a function of the size ratio, of the void ratio of the coarser material and of two empirical constants,  $k_{int}$  and  $k_p$ , the values of which depend upon the change point B, C, D or E under consideration.

In parallel with Dewar in the United Kingdom, de Larrard in France developed a scientific approach of the concrete mixture proportioning [53] based on packing density models: firstly the Linear Packing Density Model (LPDM) designed in collaboration with Stovall and Bui [54], and then the Compressible Packing Model (CPM, 1999) [53]. In 1986, Stovall et al. [54] elaborated a linear model partially contained in the Mooney’s viscosity model and based on crowding restrictions. Indeed, certain mathematical quantities involving geometric interaction functions  $f_{STO}(r)$  and  $g_{STO}(r)$  in the LPDM can be assimilated to the crowding factors  $\lambda_{12}(r)$  and  $\lambda_{21}(r)$  defined by Mooney. But the latter does not give them an explicit form while Stovall et al. provide mathematical expressions for  $f_{STO}(r)$  and  $g_{STO}(r)$ , respectively in the large sphere packed region and in the small sphere packed region. It can also be noted that the linear packing models of Stovall et al. [54] on the one hand and of Yu & Standish [10] on the other hand have the same form with different interaction functions. However, they have the disadvantage of estimating, near the optimum, too high packing density. This is why, from 1999, de Larrard [53] chose to consider the latter as a virtual packing density, intrinsic to the mixture, accessible only by placing each grain in its ideal location, but inaccessible to experimentation which depends on the chosen packing process. Thus, the Compressible Packing Model (CPM) makes it possible to determine the real packing density of the mixture thanks to an implicit equation involving a compaction index  $K$

which is a scalar representative of the packing process. By analogy with certain viscosity models including that of Mooney, the compaction index is obtained by the sum of the partial indices relating to each granular class. Each of them involves the virtual packing density of the mixture when the considered class is dominant and the real packing density of the mixture sought. To each value of  $K$  can only correspond one and only one real packing density of the mixture. In the CPM, the interaction coefficients linked to the wall effect and the loosening effect were calibrated by de Larrard on crushed and rounded aggregates.

In 2012, Fennis [55] developed an extension of the CPM to optimize the particle packing of powders in ecological concrete: the Compaction-Interaction Packing Model (CIPM). The presence of surface forces indeed requires advanced interaction equations including the additional effects of agglomerating particles on the wall and loosening effects. Indeed, due to agglomeration, the loosening effect increases and the wall effect decreases when the small particles start to stick to the coarser particles. The new interaction equations involve the size ratio and a maximum range of loosening effect and of wall effect.

As Fennis' work illustrates, the Dutch concrete school has greatly contributed to the evolution of this material towards a more ecological design [55]. Since 2006, Brouwers [56–58] gave it considerable impetus by carrying out studies on the packing density of polydisperse particles [56], of binary crystalline structures, of bimodal randomly placed spheres [57], of trimodal spheres and of binary similar particles with size ratios from 0.4 to 1 [58]. In [56], he demonstrated that a bimodal discrete packing contains important information in regard to the discrete and continuous geometric distributions of the power-law type with a distribution modulus  $q$ . According to Brouwers, in the limit  $r \rightarrow 1$  implying a continuous distribution, the maximum packing density is obtained for  $q = 0$ . For discretely sized particles, the distribution modulus is positive and varies between  $0 < q < 0.37$ . In [57], his paper reveals that the packing density of bimodal random arrangements of spheres with a size ratio  $r \uparrow 1$  can be described with a similar model as for crystalline arrangements. The closed-form equation contains the solid volume fraction of one of the two components, the size ratio, and depends on the monosized packing density and on a packing parameter with a prescribed, therefore non-adjustable, value. Finally, in [58], Brouwers revisited the excluded volume model for spherocylinders and cylinders by Onsager [23] and employed it to derive an asymptotically expression for two equally shaped (similar) particles of different sizes, i. e. with the same length/diameter ratio. From a second order perturbation, he shows that the variation from the monosized packing density amounts a quantity which is proportional to the square of the relative size difference  $1/r - 1$ . No adjustable parameter has been introduced into the equations, the governing parameters being namely all physically defined.

If the advent of high-performance concretes, their formulation and the influence of the latter on their ecological impact constituted major research themes for de Larrard on the one hand, Fennis and Brouwers on the other, the same applies for Kwan who undertook a scientific and pragmatic approach to the “concrete-system”. Kwan et al. [59,60] emphasize that, depending on the objectives, packing density is a key concept that must be maximized or optimized. They postulate that the overestimation, by linear models, of the packing density near the optimum is due to an additional geometric interaction, the wedging effect, which would not be taken into account in the calculations. The wedging effect would occur both in the domain of dominant coarse grains and in the domain of dominant fine grains. In the first case, fine particles would be trapped between coarse particles which would move apart: the latter would interact with the finer ones through loosening (coefficient  $a$ ) and wedging (coefficient  $c$ ) effects. In the second case, the fine particles would not be able to completely envelop the coarse particles: the first ones would interact with the second ones through wall (coefficient  $b$ ) and wedging (coefficient  $c$ ) effects. The packing model thus developed is a non-linear model called the 3-parameter particle packing model (3PPM). The interaction coefficients  $a, b, c$  are expressed as a function of

the size ratio  $r$  and are calibrated for spherical particles, uncompacted angular particles and compacted angular particles. Where the CPM considers that an ideal packing density is not achieved because the packing process resulting from an action external to the mixture (vibration, compaction, etc) is not optimal, the 3PPM considers a geometric interaction intrinsic to the mixture influenced by the packing process only for angular particles. One of the particularities of the 3PPM is nevertheless that the granular interactions are less strong in “uncompacted” conditions than in “compacted” conditions, which can generate higher packing densities in the first case. The strategy adopted in the CPM and in the 3PPM, regarding the number of interaction functions to adopt depending on the type of materials encountered, also differs. Either the model is adapted for a particular type of material, requires few interaction functions but lacks versatility: this is the case of the CPM with 2 interaction functions adapted for aggregates. Either the model is versatile but requires a large number of interaction functions: this is the case of the 3PPM with 9 interaction functions.

In relation to this situation, another option chosen by Roquier [61,62] consists of individualizing the quantification of the granular interactions characterized by  $a$  and  $b$ . The framework of the model, called the Theoretical Packing Density Model (TPDM), consists of the CPM with theoretical loosening effect and wall effect coefficients. Each calculation of  $a$  and  $b$  is a particular case which takes into account: the size ratio  $r$ , the shape and surface roughness of the particles via the critical cavity size ratio  $r_0$  from which the loosening effect occurs, the packing density or the specific volume of each granular class and the packing process via the compaction index  $K$ . Inspired by the work of Ben Aim [16], the methodology is based on the study of a sphere belonging to the dominated class (the intruding sphere), surrounded by spheres belonging to the dominant class. For both types of interactions, a spherical reference cell, concentric with the intruding sphere, makes it possible to calculate the packing density in the disturbed volume by the wall effect or the loosening effect and to deduce  $a$  and  $b$ . The use of the compaction index  $K$  ensures that a more efficient packing process results in an increase in the packing density of the mixture. The use of the critical cavity size ratio  $r_0$  guarantees the taking into account of an insertion mechanism even if the size ratio does not tend towards 0, for both disordered or ordered packings. Thus, by revisiting the CPM and by exploiting a modified Ben Aim's cell, the TPDM offers a generic and accurate packing model, applicable to size ratios between 0 and 1, to all packing processes (from pouring to perfectly ordered) and all particle shapes (from spherical particles to rough and angular particles). It has been validated for a wide range of particle varieties (glass beads, steel balls, micropowders, soils, rounded sand, crushed aggregates) [61–64], for mixtures with a number of granular classes between two and ten, for various particle-size distributions (power-laws, truncated power-laws, uniform by volume, fractal models) [62], for high performance concrete [63] and for asphalt mixtures [64]. Despite a larger number of equations making it possible to individualize the estimation of the interaction coefficients, the TPDM is based on a scientific corpus and is entirely characterized by parameters with physical significance.  $r_0$  and  $K$  are now calibrated and are no longer considered adjustable: the TPDM became a closed-form packing model.

## 2.9. High technology

Some materials used for very specific applications like gas turbine discs require mechanical properties far superior to those obtained by conventional industrial processes. These specifications are a combination of both high strength and high creep resistance at high temperatures [4]. When using powder metallurgy, the sintering ability of the latter is closely linked to its packing density, the estimation of which is then necessary to optimize the particle size distribution of the powder before sintering.

In this context, Ye et al. [4] developed a second-order packing density theory based on the Linear Packing Density Model (LPDM) [54]. In

the domain of dominant fine grains, the authors consider that the void caused by the wedging effect occurs when two coarse grains contact and that this probability is proportional to the square of the solid volume fraction of coarse particles. Likewise, in the domain of dominant coarse grains, they consider that the additional void is proportional to the square of the solid volume fraction of fine particles. Finally, the peak observed at the junction between the two domains on the curve representing the packing density as a function of the solid volume fraction of one of the two components is attenuated.

### 3. Limiting cases: Filling mechanism, substitution mechanism, total interaction

The following equations relate to the specific volume of a binary mixture made up of coarse particles 1 and fine particles 2. The volume fractions by reference of the total solid volume are respectively noted:  $y_1$  and  $y_2$ , the specific volumes of granular classes are:  $v_1$  and  $v_2$ .

#### 3.1. Furnas (1928)

In 1928, Furnas [1] provided the mathematical expressions for the *filling mechanism* (Eq. (5)) and the *substitution mechanism* (Eq. (6)). They are materialized by the segments AC and CB in Fig. 2.

$$V_1 = v_1 y_1 \quad (5)$$

$$V_2 = y_1 + v_2 y_2 \quad (6)$$

Both cases are characterized by an *absence of interaction*.

#### 3.2. Westman & Hugill (1930), Colby (1963)

Westman & Hugill [6] connect the points A and B (Fig. 2) by a straight line which they describe as the specific volume of the constituents before mixing, without giving its equation. It was not until 1963 that Colby [38] introduced the following formula:

$$V = v_1 y_1 + v_2 y_2 \quad (7)$$

The expression proposed to qualify this equation is that of *total interaction* as opposed to the *absence of interaction*.

### 4. Linear packing models with respect to the solid volume fractions

The linear packing models are presented by chronological order. The equations are expressed in terms of specific volume to facilitate comparisons. The size ratio is defined by  $r = d_2/d_1$  ( $d_2 \leq d_1$ ). The critical cavity size ratio from which the loosening effect occurs or the critical ratio of entrance is noted:  $r_0$ .

#### 4.1. Mooney (1951)

Mooney [47] indirectly developed a packing model to study the relative viscosity of a concentrated suspension in a Newtonian solvent. When the coarse and the fine particles are respectively dominant, the equations are the following:

$$V_1 = v_1 y_1 + \lambda_{12}(r) y_2 \quad (8)$$

$$V_2 = \lambda_{21}(r) y_1 + v_2 y_2 \quad (9)$$

Mooney did not provide equations for  $\lambda_{12}(r)$  and  $\lambda_{21}(r)$  but he specifies the following limits:  $\lim_{r \rightarrow 0} \lambda_{12}(r) = 0$ ,  $\lim_{r \rightarrow 0} \lambda_{21}(r) = 1$ , which allow to find Eqs. (5) and (6) when they are verified. Moreover, he highlights that:  $\lim_{r \rightarrow 1} \lambda_{12}(r) = \lim_{r \rightarrow 1} \lambda_{21}(r) = 1/\alpha$ ,  $\alpha$  being the packing density of spherical particles in a monodispersed packing state. One therefore find Eq. (7) when  $r \rightarrow 1$  only if the condition  $v_1 = v_2 = 1/\alpha$  is satisfied.

#### 4.2. Ben Aim (1970)

Ben Aim [16] distinguishes two cases: the mixture comprises either an excess of coarse spheres (Eq. (10)), either an excess of small spheres (Eq. (11)), compared to the *eutectic*:

$$V_1 = v_1 y_1 \quad (10)$$

$$V_2 = \left\{ 1 + \frac{5}{16} \left[ (1 + 2r)^{(3/2)} - 1 \right] \right\} y_1 + v_2 y_2 \quad (11)$$

For  $r \leq 0.2$ , Ben Aim proposes to replace Eq. (11) with Eq. (12):

$$V_2 = (1 + 0.9r) y_1 + v_2 y_2 \quad (12)$$

For  $r = 0$ , Eqs. (5) and (6) are respected. On the other hand, Eq. (7) is not satisfied for  $r = 1$ . It should however be remembered that Ben Aim has primarily focused on mixtures with high size contrast.

#### 4.3. Stovall, de Larrard, Buil (1986)

In 1986, Stovall et al. [54] proposed the linear packing density model of grain mixtures. The established formulas are as follows, respectively for the dominant coarse and fine particles:

$$V_1 = v_1 y_1 + v_1 y_2 (1 - f_{STO}(r)) \quad (13)$$

$$V_2 = [v_2 - (v_2 - 1) g_{STO}(r)] y_1 + v_2 y_2 \quad (14)$$

The interaction functions in the coarse sphere packed region and in the small sphere packed region are respectively:

$$f_{STO}(r) = \begin{cases} 1 - \frac{\left[ 1 - \left( \frac{r_0}{r} \right)^3 \right]}{\left\{ \left[ \frac{(v_1 - 1)}{v_1} [1 - r_0^3] - 3r_0^3 \right] (1 - r) + (1 - r_0^3) \right\}} & r > r_0 \\ 1 & r \leq r_0 \end{cases} \quad (15)$$

$$g_{STO}(r) = 1 - r \quad (16)$$

where  $r_0 \approx 0.2$  for spheres [54]. It can be noticed that:  $f_{STO}(r \leq r_0) = g_{STO}(0) = 1$ : the *filling and substitution mechanisms* are then active. However, it can be observed that:  $f_{STO}(1) = g_{STO}(1) = 0$ , which generates respectively  $V_1 = v_1$  in the coarse dominant zone and  $V_2 = v_2$  in the fine dominant zone: the *total interaction* condition is only satisfied when  $v_1 = v_2$ . The notion of dominant granular class is thus particularly highlighted in the case  $r = 1$ .

#### 4.4. Yu, Standish (1987)

Yu & Standish [10] resulted in the following two equations in the domains of coarse dominants and fine dominants for their "linear packing model":

$$V_1 = v_1 y_1 + \left( v_1 - \frac{(v_1 - v_{12}(r))}{y_{12}(r)} \right) y_2 \quad (17)$$

$$V_2 = \left( v_2 - \frac{(v_2 - v_{21}(r))}{y_{21}(r)} \right) y_1 + v_2 y_2 \quad (18)$$

where  $v_{12} = v_{21}$  is the minimum specific volume of components 1 and 2 and  $y_{12}$  or  $y_{21}$  is the corresponding solid volume fraction:

$$y_{12}(r) = 1 - \frac{(1 - r^2)}{\left( 2 - \frac{1}{v_0} \right)} \text{ and } y_{21}(r) = \frac{(1 - r^2)}{\left( 2 - \frac{1}{v_0} \right)} \quad (19)$$

$$v_{12}(r) = v_{21}(r) = \begin{cases} \frac{v_0}{\left\{1 + \left(1 - \frac{1}{v_0}\right) (1 - 2.35 r + 1.35 r^2)\right\}} & r \leq 0.741 \\ v_0 & r > 0.741 \end{cases} \quad (20)$$

$v_0$  is called the initial specific volume. By considering  $v_0 = v_1 y_1 + v_2 y_2$ , the *filling and substitution mechanisms* and the *total interaction condition* are satisfied when  $v_1 = v_2$  and almost respected when  $v_1 \neq v_2$ .

#### 4.5. Yu, Zou, Standish (1996)

In 1996, the precedent version has been improved to fit both spherical and non-spherical particles [65], respectively for the dominant coarse particles and the dominant fine particles:

$$V_1 = v_1 y_1 + v_2 y_2 (1 - f_{YU}(r)) \quad (21)$$

$$V_2 = [v_1 - (v_1 - 1) g_{YU}(r)] y_1 + v_2 y_2 \quad (22)$$

The interaction functions  $f_{YU}(r)$  and  $g_{YU}(r)$  are respectively, for spherical particles only:

$$f_{YU}(r) = (1 - r)^{3.3} + 2.8 r (1 - r)^{2.7} \quad (23)$$

$$g_{YU}(r) = (1 - r)^{2.0} + 0.4 r (1 - r)^{3.7} \quad (24)$$

Note that the linear model equations (Eqs. (21) and (22)) of Yu et al. [65] are not identical to those (Eqs. (13) and (14)) of Stovall et al. [54]. Indeed, certain indices related to the specific volumes of the granular classes have been modified by Yu et al. Thanks to the chosen interaction functions, the *filling and substitution mechanisms* and the *total interaction criterion* are satisfied.

#### 4.6. Chang, Wang, Ge (2015)

The formulas in the dominant coarse and dominant fine domains are as follows [33]:

$$V_1 = v_1 y_1 + v_2 y_2 (1 - f_{CHA}(r)) \quad (25)$$

$$V_2 = (v_1 - (v_1 - 1) g_{CHA}(r)) y_1 + v_2 y_2 \quad (26)$$

The filling coefficient  $f_{CHA}(r)$  and the embedding coefficient  $g_{CHA}(r)$  are respectively:

$$f_{CHA}(r) = (1 - r)^p \quad (27)$$

$$g_{CHA}(r) = (1 - r)^s \quad (28)$$

It can be noticed that:

- $f_{CHA}(0) = g_{CHA}(0) = 1$  and  $f_{CHA}(1) = g_{CHA}(1) = 0$  which allows to check Eqs. (5) to (7) respectively.
- Eqs. (25) and (26) are similar to Eqs. (21) and (22) respectively. Only the interaction functions change.
- The filling coefficient and the embedding coefficient are a power function of size ratio  $r$ .
- Two parameters,  $p$  and  $s$ , are required to estimate the specific volume of the binary mixture.
- $p$  evolves between 1.20 for steel shots and 3.86 for a sand-silt mixture called Ottawa mixture [33].
- $s$  evolves between 1.40 for concrete mixes and 2.89 for Ottawa mixture [33].

### 5. Non-linear packing models with respect to the solid volume fractions

The non-linear packing models are also presented by chronological order and are written in terms of specific volume to facilitate

comparisons.

#### 5.1. Westman (1936)

Westman [7] developed the following very clever conic equation:

$$\left\{ \frac{(V - v_1 y_1)}{v_2} \right\}^2 + 2G \left\{ \frac{(V - v_1 y_1)}{v_2} \right\} \left\{ \frac{(V - y_1 - v_2 y_2)}{(v_1 - 1)} \right\} + \left\{ \frac{(V - y_1 - v_2 y_2)}{(v_1 - 1)} \right\}^2 = 1 \quad (29)$$

For  $G \rightarrow \infty$ , Eq. (29) takes the form of Eq. (30). It means that  $V = v_1 y_1$  in the coarse dominant zone and  $V = y_1 + v_2 y_2$  in the fine dominant zone. The *filling and substitution mechanisms* are thus satisfied.

$$\left\{ \frac{(V - v_1 y_1)}{v_2} \right\} \left\{ \frac{(V - y_1 - v_2 y_2)}{(v_1 - 1)} \right\} = 0 \quad (30)$$

For  $G = 1$ , Eq. (29) resolves itself into the linear equation of Eq. (31). The solution is:  $V = v_1 y_1 + v_2 y_2$  which means that the *total interaction* occurs.

$$\frac{(V - v_1 y_1)}{v_2} + \frac{(V - y_1 - v_2 y_2)}{(v_1 - 1)} = 1 \quad (31)$$

Yu et al. [8] proposed, for the coefficient  $G$ , a general relation whis is given by the following equation:

$$G^{-1} = \begin{cases} 1.355 r^{1.566} & r \leq 0.824 \\ 1 & r > 0.824 \end{cases} \quad (32)$$

Marmur [13], for his part, recommends the following formula:

$$G^{-1} = 0.585 r \quad (33)$$

The condition  $G = 1$  for  $r = 1$  is therefore not satisfied with Eq. (33).

#### 5.2. Toufar, Born, Klose (1976)

In 1976, the model of Toufar et al. [50] estimates the specific volume of a binary mixture as:

$$V = v_1 y_1 + v_2 y_2 - y_1 (v_1 - 1) k_d k_s \quad (34)$$

Eq. (34) is therefore established from Eq. (7), representative of the *total interaction*, with an additional term taking into account two factors.  $k_d$  determines the influence of the size ratio while  $k_s$  is a statistical factor involving the volume fractions and the specific volumes of each granular class:

$$k_d = \frac{(1 - r)}{(1 + r)} \quad (35)$$

$$k_s = 1 - \frac{(1 + 4z)}{(1 + z)^4} \text{ with } z = \left( \frac{y_2}{y_1} \right) \frac{v_2}{(v_1 - 1)} \quad (36)$$

When  $r = 1$ , Eq. (7) holds. When  $r = 0$ , Eqs. (5) and (6) are not satisfied. In the latter case however, as the volume fraction of fine particles increases,  $k_s$  gradually tends towards 1 and since  $k_d = 1$ , the *substitution mechanism* becomes active.

#### 5.3. Han, Wang, Xiang (1981)

The main equation (Eq. (37)) proposed by Han et al. [44] is relatively close to that established by Toufar et al. (Eq. (34)):

$$V = v_1 y_1 + v_2 y_2 - y_1 (v_1 - 1) B \quad (37)$$

However, where Toufar et al. [50] separate the effect of the size ratio and the effect of the volume fractions of each component into two terms ( $k_d$  and  $k_s$ ), multiplied by each other, Han et al. [44] associate them more intimately. The corresponding equation (Eq. (38)) is based on geometric



considerations in 3D concerning the probability  $P_2$  for one coarse particle to come into contact with a fine particle and in 2D for the number of layers  $m$  (Eq. (39)) of fine particles needed to fill the void corresponding to each coarse particle (Fig. 2 in [42]).

$$B = P_2^{m+1} = \left( \frac{y_2}{y_1 r + y_2} \right)^{m+1} \quad (38)$$

$$m = \frac{1}{2} + \frac{(2 - \sqrt{3})(1+r)}{2r\sqrt{3}} \quad (39)$$

When  $B = 0$ , Eq. (37) reduces to Eq. (7). This case corresponds to  $y_2 = 0$ , that is to say a monodisperse packing. However, in the case of a binary mixture consisting of two granular classes with the same diameter ( $r = 1$ ) but with different specific volumes ( $v_1 \neq v_2$ ), the condition of *total interaction* is not satisfied, that is to say that the representative points are not located on the segment AB (Fig. 2).

When  $B = 1$ , Eq. (37) reduces to Eq. (6). However, this case is incompatible with Eq. (38). Indeed,  $r \rightarrow 0$  generates  $P_2 \rightarrow 1$  and  $m \rightarrow \infty$ , which leads to an indeterminate form for  $B$ . Concretely, the *substitution mechanism* tends to be active only if the volume fraction of fine particles  $y_2$  becomes relatively large when  $r \rightarrow 0$ .

When  $B = z = \left( \frac{y_2}{y_1} \right) \frac{v_2}{(v_1-1)}$ , Eq. (37) reduces to Eq. (5). The *filling mechanism* is active only in this particular case when  $r \rightarrow 0$  and therefore cannot be generalized to other volume fractions of the components. It should also be noted that for very strong size contrasts and for low volume fractions of fine particles, the specific volume of the mixture may be less than the lower limit provided by Eq. (5).

#### 5.4. Yu, Standish (1988)

As mentioned above, the “mixture model” [11] proposed by Yu & Standish is a cubic “non-linear packing model” with respect to the solid volume fractions:

$$V = v_1 y_1 + v_2 y_2 + \beta_{12} y_1 y_2 + \gamma_{12} y_1 y_2 (y_1 - y_2) \quad (40)$$

$\beta_{12}$  and  $\gamma_{12}$  are respectively the quadratic and the cubic coefficients of the binary synergism.  $v_0$  is the initial specific volume.

$$\beta_{12} = \begin{cases} 0 & r \geq 0.741 \\ 10.288 \times 10^{-\frac{1.4566}{v_0}} \{ -1.0002 + 0.1126 r + 5.8455 r^2 - 7.9488 r^3 + 3.1222 r^4 \} & r < 0.741 \end{cases} \quad (41)$$

$$\gamma_{12} = \begin{cases} 0 & r \geq 0.741 \\ \left\{ -1.3092 + \frac{15.039}{v_0} - \frac{37.453}{v_0^2} + \frac{40.869}{v_0^3} - \frac{17.110}{v_0^4} \right\} \{ -1.0029 + 0.3589 r + 10.97 r^2 - 22.197 r^3 + 12.434 r^4 \} & r < 0.741 \end{cases} \quad (42)$$

Compliance with the principle of *total interaction* is easily verifiable insofar as, for  $r \geq 0.741$ ,  $\beta_{12} = \gamma_{12} = 0$  which makes it possible to find Eq. (7) from Eq. (40). As Yu & Standish reserve this model for mixtures with a size ratio  $r > 0.154$ , verification of the *insertion* and *substitution mechanisms* is not of great interest.

#### 5.5. Yu, Standish (1991), Yu, Zou, Standish (1996)

The “linear-mixture packing model” proposed by Yu et al. [9,65] combines their “linear packing model” and their cubic non-linear “mixture model”. When binary mixtures of particles are considered, two packing effects are observed according to Yu et al. (1991) [9]: the *filling mechanism* and the *occupation mechanism*. The critical cavity size ratio  $r_0 = 0.154$  may be used to specify whether a binary mixture of particles is mainly characterized by *filling* (unmixing effect) or by *occupation* (mixing effect).

If  $r \leq r_0 = 0.154$ , the packing is predominantly characterized by *filling* and Yu et al. (1996) [65] call upon their “linear packing model”:

$$V = \max\{v_1 y_1 + v_2 y_2 (1 - f_{YU}(r)); [v_1 - (v_1 - 1) g_{YU}(r)] y_1 + v_2 y_2\} \quad (43)$$

The interaction functions are given by Eqs. (23) and (24).

If  $r > r_0 = 0.154$ , the packing is predominantly characterized by *occupation* and Yu et al. call upon their “mixture model” (1991) [9]:

$$V = v_0 + \beta_{12} y_1 y_2 + \gamma_{12} y_1 y_2 (y_1 - y_2) \quad (44)$$

where  $v_0$  is a weighted-average initial specific volume of the two components, provided by:

$$v_0 = v_1 y_1 + v_2 y_2 \quad (45)$$

The combination of Eqs. (44) and (45) gives Eq. (40).  $\beta_{12}$  and  $\gamma_{12}$  are provided by Eqs. (41) and (42). Thanks to the “linear-mixture packing model”, the *filling mechanism* (Eq. (5)), the *substitution mechanism* (Eq. (6)) and the *total interaction* (Eq. (7)) criteria are all met. However, there may be a slight discontinuity in the estimated packing densities when switching from the linear model to the non-linear model for the size ratio of 0.154.

#### 5.6. Zheng, Carlson, Reed (1995)

Inspired by the ideal particle packing of Furnas, Zheng et al. (1995) [14] developed the following empirical equation:

$$V = v_1 \left\{ 1 + \frac{(v_1 - 1)}{v_2} (e y_2 \ln(y_2))^{1.25 v_1} \exp(-4 r) \right\}^{-1} \quad (46)$$

where  $e$  is the base of a natural logarithm. The volume fractions of the constituents and the size ratio are therefore taken into account separately. The authors mainly use their model for  $r$  between 0.1 and 0.5 and did not attach particular importance to mixtures with high size contrast or single size for which Eqs. (5) to (7) are not satisfied. In particular, when  $r \rightarrow 0$ , the representative points of the specific volume of the mixture can be located below the segments AC and CB (Fig. 2).

### 5.7. Goltermann, Johansen, Palbøl (1997)

In 1997, Goltermann et al. [51] proposed to make a minor correction to the Toufar's model [50] by introducing a modification to the  $k_s$  expression (Eq. (36)):

$$k_s = \begin{cases} \frac{z}{z_0} k_0 & z < z_0 \\ 1 - \frac{(1+4z)}{(1+z)^4} & z \geq z_0 \end{cases} \quad (47)$$

where  $z_0 = 0.4753$  and  $k_0 = 0.3881$ . If  $z < z_0$ , Eq. (34) becomes linear:

$$V = v_1 y_1 + v_2 y_2 \left( 1 - k_d \frac{k_0}{z_0} \right) \quad (48)$$

The conclusions stated for the Toufar's model remain valid for the Goltermann's model.

### 5.8. De Larrard (1999)

The Compressible Packing Model (CPM) of de Larrard [53] is an evolution of the Stovall's model (1986) [54]. Designed for granular mixtures, the CPM takes into account the packing process via a compaction index  $K$ . When the packing process is perfect, i.e. when each particle is positioned at its ideal location, the packing is said to be virtual,  $K \rightarrow \infty$ , and the specific volume of the binary mixture ( $\mathcal{V}_1$  in the field of dominant coarse grains and  $\mathcal{V}_2$  in the field of dominant fine grains) is calculated using the improved Stovall's model so that the *total interaction* condition is satisfied.

$$\mathcal{V}_1 = \epsilon_1 y_1 + \epsilon_2 y_2 a_{LAR}(r) \quad (49)$$

$$\mathcal{V}_2 = [1 + (\epsilon_1 - 1) b_{LAR}(r)] y_1 + \epsilon_2 y_2 \quad (50)$$

where  $\epsilon_1$  and  $\epsilon_2$  are the virtual specific volumes of the coarse and fine granular classes respectively. Calibrated on crushed and rounded aggregates, the loosening effect coefficient  $a_{LAR}(r)$  and the wall effect coefficient  $b_{LAR}(r)$  are respectively:

$$a_{LAR}(r) = \sqrt{1 - (1-r)^{1.02}} \quad (51)$$

$$b_{LAR}(r) = 1 - (1-r)^{1.5} \quad (52)$$

The compaction index  $K$  has a mathematical structure analogous to certain viscosity models, such as that of Mooney [47]. Its function is determined by writing the self-consistency of the system which leads to:

$$K = K_1 + K_2 = \frac{\epsilon_1 y_1}{(V - \mathcal{V}_1)} + \frac{\epsilon_2 y_2}{(V - \mathcal{V}_2)} \quad (53)$$

The specific volume of the binary mixture  $V$  is determined implicitly by Eq. (53) in which  $\mathcal{V}_1$  and  $\mathcal{V}_2$  are calculated by Eqs. (49) and (50).  $K$  is a strictly decreasing function of  $V$  which means there is a unique value of  $V$  that satisfies Eq. (53) for any positive value of  $K$ . For a single grain size,  $K$  becomes:

$$K = \frac{1}{\left(\frac{v}{\epsilon} - 1\right)} \quad (54)$$

where  $\epsilon$  and  $v$  are respectively the virtual and the real specific volumes of the single granular class. The virtual specific volumes of the coarse and

**Table 1**  
Summary of  $K$  values in the CPM.

Packing Process	Pouring	Sticking with a rod	Vibration	Vibration + compression	Perfect
K	4.1	4.5	4.75	9	$\infty$

fine granular classes, respectively  $\epsilon_1$  and  $\epsilon_2$  (Eq. (55)), are obtained from their real specific volume, respectively  $v_1$  and  $v_2$ , from Eq. (54):

$$\epsilon_1 = v_1 \frac{K}{(1+K)}, \epsilon_2 = v_2 \frac{K}{(1+K)} \quad (55)$$

The calibration procedures carried out with the CPM on different packing processes have led to the values of  $K$  given in Table 1.

It can be noticed that:

- $a(r) = 1 - f(r)$  and  $b(r) = 1 - g(r)$ ,  $f(r)$  and  $g(r)$  being interaction functions used in other models. De Larrard indeed prefers to consider that the interaction coefficients are equal to 0 in the absence of interaction ( $d_1 \gg d_2$ ) and to 1 in the case of a total interaction ( $d_1 = d_2$ ).
- The structure of Eq. (49) is therefore identical to that of Eq. (21) (Yu et al. [65]) and Eq. (25) (Chang et al. [33]). Likewise, the structure of Eq. (50) is identical to that of Eqs. (22) and (26).
- The formulas  $a_{LAR}(r)$  and  $b_{LAR}(r)$  have been chosen so as to verify the continuity of the functions representative of the specific volume of the binary mixture and of their derivatives when  $d_1 = d_2 = d$ , which implies:  $a_{LAR}(1) = b_{LAR}(1) = 1$  and  $\left. \frac{\partial a_{LAR}(r)}{\partial r} \right|_{r=1} = \left. \frac{\partial b_{LAR}(r)}{\partial r} \right|_{r=1} = 0$ .

The *total interaction* (Eq. (7)) criterion is satisfied. The *filling mechanism* (Eq. (5)) and the *substitution mechanism* (Eq. (6)) are active when  $K \rightarrow \infty$ .

### 5.9. Kwan, Chan, Wong (2013), Kwan, Wong, Fung (2015)

The 3-parameter particle packing model (3PPM), developed by Kwan et al. [59,60], incorporates an additional geometric interaction. Besides the loosening effect coefficient  $a$  and the wall effect coefficient  $b$ , the authors indeed consider the wedging effect coefficient  $c$  both when coarse particles (Eq. (56)) and fine particles (Eq. (57)) are dominant.

$$V_1 = v_1 y_1 + v_2 y_2 \{ 1 - (1 - a_{KWA}(r)) (1 - c_{KWA}(r) (3.8^{y_2} - 1)) \} \quad (56)$$

$$V_2 = v_1 y_1 \left\{ 1 - (1 - b_{KWA}(r)) \left( \frac{v_1 - 1}{v_1} \right) (1 - c_{KWA}(r) (2.6^{y_1} - 1)) \right\} + v_2 y_2 \quad (57)$$

The mathematical expressions of the three interaction parameters are as follows:

$$a_{KWA}(r) = 1 - (1-r)^{a_0} - a_1 r (1-r)^{a_2} \quad (58)$$

$$b_{KWA}(r) = 1 - (1-r)^{b_0} - b_1 r (1-r)^{b_2} - b_3 (1-r)^{b_4} \quad (59)$$

$$c_{KWA}(r) = c_0 \tanh(c_1 r) \quad (60)$$

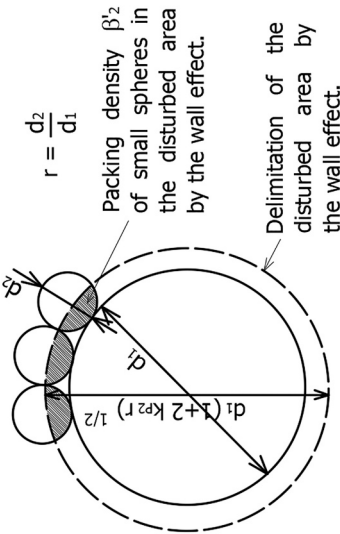
The coefficients involved in the formulas of the interaction parameters are provided in Table 2.

It can be noticed that:

**Table 2**  
Summary of coefficients involved in the 3PPM.

Subscript	0	1	2	3	4
Spheres	a	3.3	2.6	3.6	
	b	1.9	2	6	0
	c	0.322	11.9		
Uncompacted angular particles	a	5	1.9	3.1	
	b	1.9	2.1	10.5	0.2
	c	0.335	26.9		
Compacted angular particles	a	7.1	1.9	3.1	
	b	2.2	0.7	9.3	0.2
	c	0.335	26.9		

**Table 3**  
Determination of the wall effect coefficient  $b_{ROQ}$ .



$$\beta_2 = \frac{1}{2} = \frac{1}{2} \frac{(1+K)}{v_2} \quad (65)$$

$$d_{hyp} = d_1(1+2k_{p2}r)^{(1/2)} \quad (66)$$

$$N_{12}^{dense}(r) = \frac{\pi(1+r)}{r \arcsin\left(\frac{r}{1+r}\right)} \quad (67)$$

$$(\beta_2 - 6)(1+2k_{p2})^{(3/2)} + \frac{9}{2}k_{p2}^2 + 18k_{p2} + 5 = 0 \quad (68)$$

$$\beta_2(r) = \frac{N_{12}^{dense}(r)}{4(1+2k_{p2}r)^{(3/2)} - 1} \times \left( 2(1+2k_{p2}r)^{(3/2)} - 3(1+2k_{p2}r) \left( 1 + \frac{k_{p2}r}{1+r} \right) + \left( 1 + \frac{k_{p2}r}{1+r} \right)^3 - r^3 + \frac{3k_{p2}r^3}{1+r} \left( 1 - \frac{k_{p2}r}{1+r} \right)^3 \right) \quad (69)$$

$$b_{ROQ}(r) = \frac{(\beta_2 - \beta_2^i(r))}{(1 - \beta_2)} \left[ (1+2k_{p2}r)^{(3/2)} - 1 \right] \quad (70)$$

- The packing process is taken into account directly in the interaction functions since Kwan et al. distinguish compacted and uncompact angular particles in Table 2.
- $a_{KWA}(1) = b_{KWA}(1) = 1$  which allows to verify Eq. (7): the *total interaction* condition is respected.
- $a_{KWA}(0) = c_{KWA}(0) = 0$  which allows to verify Eq. (5): the *filling mechanism* is satisfied.
- For spheres,  $b_{KWA}(0) = c_{KWA}(0) = 0$  which allows to verify Eq. (6): the *substitution mechanism* is satisfied.
- For angular particles,  $b_{KWA}(0) = -0.2$  which does not allow to verify Eq. (6): the *substitution mechanism* is not satisfied. On the side of the dominant fine grains, the curve representing the specific volume of the binary mixture for strong size contrasts is even below the straight line provided by Eq. (6).

### 5.10. Roquier (2016,2024)

The Theoretical Packing Density Model (TPDM) [61,62] takes root in the Compressible Packing Model (CPM) [53] with innovative wall effect and loosening effect coefficients  $b_{ROQ}(r)$  (Table 3) and  $a_{ROQ}(r)$  (Table 4).

$$\mathcal{V}'_1 = v_1 y_1 + v_2 y_2 a_{ROQ}(r) \quad (61)$$

$$\mathcal{V}'_2 = [1 + (v_1 - 1) b_{ROQ}(r)] y_1 + v_2 y_2 \quad (62)$$

$$v_1 = v_1 \frac{K}{(1+K)}; v_2 = v_2 \frac{K}{(1+K)} \quad (63)$$

$$K = K_1 + K_2 = \frac{v_1 y_1}{(V - \mathcal{V}'_1)} + \frac{v_2 y_2}{(V - \mathcal{V}'_2)} \quad (64)$$

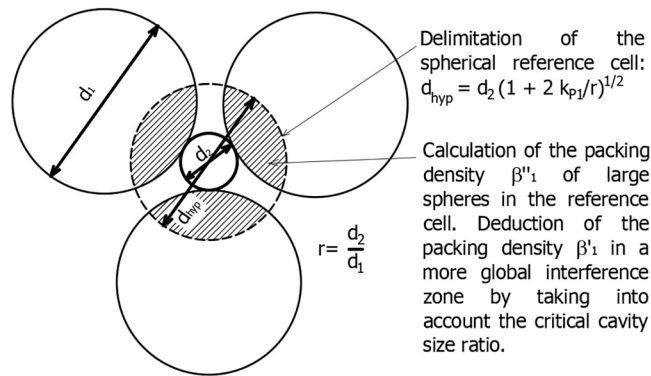
$v_1$  and  $v_1$  are respectively the real and the virtual specific volumes of the coarse class,  $v_2$  and  $v_2$  the real and the virtual specific volumes of the fine class,  $\mathcal{V}'_1$  and  $\mathcal{V}'_2$  the virtual specific volumes of the binary mixture in the field of dominant coarse grains and in the field of dominant fine grains,  $K$  the compaction index. The real specific volume of the binary mixture  $V$  is calculated thanks to the implicit Eq. (64).

The calibration procedures carried out with the TPDM on different packing processes have led to the values of  $K$  given in Table 5. The interaction coefficients  $a_{ROQ}(r)$  and  $b_{ROQ}(r)$  are determined in such a way as to involve not only the size ratio  $r$  but also the real specific volumes of each granular class, the compaction index  $K$  and the possibility, or not, for a fine particle to fit into an available cavity without modifying the skeleton of coarse particles thanks to the critical cavity size ratio  $r_0$ .

It can be noticed that:

- The wall effect (Table 3) is considered located in a spherical crown delimited by two spheres of diameters  $d_1$  and  $d_{hyp}$ . The latter (Eq. (66)) is calculated for respecting the boundary condition  $b_{ROQ}(1) = 1$ .
- The loosening effect occurs if  $r > r_0$  (Table 4). In this case, the structure of coarse particles decompacts around each small particle. The "local" volume of coarse particles is calculated in the spherical cell of diameter  $d_{hyp}$  (Eq. (72)) to ensure the continuity between the two geometric interactions.
- To calculate the packing density of the particles of the dominant class in the areas disturbed by the wall effect or the loosening effect, the coordination number is determined by Eqs. (67) and (73) which reveal two remarkable results:  $N_{12}^{dense}(1) = N_{21}^{dense}(1) = 12$  and  $N_{21}^{dense}(0.225) \approx 4$ . The first one corresponds to the kissing number problem in 3D, the second one to the critical cavity size ratio  $r_0 = 0.225$  highlighted by Manegold [15].
- By analogy to molecular eutectics for which small molecules come to lodge inside the pseudo-crystalline lattice of coarse molecules [16], a reasoning at the "eutectic" point [61,62] allows to find Eqs. (76) to (78) (Table 4). The transition from the local scale of the spherical cell to the global scale of the packing is carried out using Eq. (76). The

**Table 4**  
Determination of the loosening effect coefficient  $a_{ROQ}$ .



$$\beta_1 = \frac{1}{v_1} = \frac{1}{v_1} \frac{(1+K)}{K} \quad (71)$$

$$d_{hyp} = d_2 \left(1 + \frac{2 k_{p1}}{r}\right)^{(1/2)} \quad (72)$$

$$N_{21}^{dense}(r) = \frac{\pi(1+r)}{\arcsin\left(\frac{1}{1+r}\right)} \quad (73)$$

$$(\beta_1 - 6) (1 + 2 k_{p1})^{(3/2)} + \frac{9}{2} k_{p1}^2 + 18 k_{p1} + 5 = 0 \quad (74)$$

$$\beta_1^i(r) = \frac{N_{21}^{dense}(r)}{4 \left(1 + \frac{2 k_{p1}}{r}\right)^{(3/2)}} \times \left(2 \left(1 + \frac{2 k_{p1}}{r}\right)^{(3/2)} - 3 \left(1 + \frac{2 k_{p1}}{r}\right) \left(1 + \frac{k_{p1}}{(1+r)}\right) + \left(1 + \frac{k_{p1}}{(1+r)}\right)^3 - \frac{1}{r^3} + \frac{3 k_{p1}}{r^2(1+r)} + \left(\frac{1}{r} - \frac{k_{p1}}{(1+r)}\right)^3\right) \quad (75)$$

$$\beta_1^i(r) = \frac{\beta_1^i(r)}{\beta_1^i(r_0) \left\{1 + \frac{(r-r_0)}{(1-r_0)} \left(\left(\frac{2 \beta_1^i(1)}{\beta_1^i(r_0)}\right)^{(1/3)} - 1\right)\right\}^3} \beta_1 \quad (76)$$

$$\phi_2^s(r) = \beta_2 + ((1-\beta_2)(1-b_{ROQ}(r)) - 1) \beta_1^i(r) \geq 0 \quad (77)$$

$$a_{ROQ}(r) = \frac{(\beta_1 - \beta_1^i(r))}{\phi_2^s(r)} \text{ if } r \geq r_0 \text{ and } a_{ROQ}(r) = 0 \text{ if } r \leq r_0 \quad (78)$$

**Table 5**  
Summary of K values in the TPDM.

Packing Process	Pouring	Tapping	Vibration or compaction	Vibration + compression	Optimized filling + vibration + compression	Crystalline structures	Perfect
K	3 to 4	4.75	5.6	9	15	100	$\infty$

latter makes it possible to verify that the optimal volume fraction of each granular class tends towards 0.5 when  $r \rightarrow 1$ . The reasoning is carried out by considering  $v_1 = v_2$ . In this case,  $a_{ROQ}(1) = 1$  and the *total interaction* criterion is satisfied.

- $a_{ROQ}(r \leq r_0) = b_{ROQ}(0) = 0$ . For  $K \rightarrow \infty$ , the *filling mechanism* is active when  $r \leq r_0$  and the *substitution mechanism* occurs when  $r = 0$ .
- The theory developed for spheres adapts to other types of particles thanks to  $r_0$  (Table 6).  $r_0$  evolves between 0.0001 for rough and angular particles to 0.20 for frictionless spherical particles.
- Finally, only two parameters having a physical meaning,  $K$  (Table 5) and  $r_0$  (Table 6), are necessary to estimate the specific volume of the binary mixture.

### 5.11. Wu, Li (2017)

Wu & Li [42] refer to the work of Han et al. [44]:

$$V = v_1 y_1 + v_2 y_2 - y_1 (v_1 - 1) B \quad (79)$$

However, where Han et al. used a simplified 2D packing arrangement, Wu & Li favor a 3D packing of spheres. Concerning the filling coefficient  $B$ , they consider the ratio between the number of fine particles available and the number of fine particles needed to cover the surface of coarse particles in a sediment mixture:

$$B = \min\left(\frac{P_2 R_N}{m N_c}, B_{max}\right) \quad (80)$$

where  $P_2$  is the probability for a coarse particle to contact with a fine particle,  $R_N$  the ratio of the numbers of fine and coarse particles in the sediment mixture,  $m$  the number of layers of fine particles needed to completely fill the voids of the coarse particles and  $m N_c$  the number of

fine particles needed to completely fill the void of a coarse particle:

$$m = \frac{1}{r} \left(\frac{\sqrt{6}}{4} - \frac{1}{2}\right) \approx \frac{0.1124}{r} \quad (81)$$

$$N_c = \frac{2 \pi}{\left(\arcsin\left(\frac{r}{1+r}\right)\right)^2 \sqrt{3}} \quad (82)$$

$$P_2 R_N = \left(\frac{y_1}{y_2} r^3 \left(1 + r \frac{y_1}{y_2}\right)\right)^{-1} \quad (83)$$

The authors introduced the limit  $B_{max}$  (Eq. (80)) for  $B$  because they consider that an ideal filling is difficult to achieve.  $B_{max}$  is equal to 0.85 for pebble-sand mixtures, 0.65–0.80 for sand-clay mixtures under confining hydrostatic pressures and 0.65 for sand-clay mixtures in

**Table 6**  
Critical cavity size ratio  $r_0$  for different types of particles in the TPDM.

Critical cavity size ratio	Spherical particles		Crushed particles	
	Frictionless	With friction	Rough or angular	Rough and angular
$r_0$	0.20	0.17	0.03	0.0001

**Table 7**  
Parameters  $p$  and  $s$  for different materials according to Chang & Deng [34].

	Sand-silt	Steel shots	Steel ball bearings	Glass beads	Washed sand
$p$	3.50	2.00	2.40	3.08	2.53
$s$	1.60	1.70	1.50	1.88	2.30



rivers. It can be noticed that Eqs. (5) to (7) are not satisfied.

### 5.12. Chang, Deng (2018)

Chang & Deng [34] developed the concept of effective dominant skeleton to refine the estimates near the optimum. According to them, the dominant packing skeleton is represented by an effective dominant size. The latter, instead of being a constant as  $d_1$  or  $d_2$  in a classical linear packing model, is an effective value  $\bar{d}$  between  $d_1$  and  $d_2$  and can be expressed as a continuous variable with respect to  $y_2$ . The authors model a binary mixture as a special case of  $n$ -component mixtures in which  $(n - 2)$  fictitious particles interact with others while their solid volume fraction is equal to zero. The equations are the following:

$$V(\bar{d}) = y_1 [v_1 - (v_1 - 1)f_{CHA}(\bar{d})] + v_2 y_2 (1 - g_{CHA}(\bar{d})) \quad (84)$$

The filling-mechanism coefficient  $f_{CHA}(\bar{d})$  and the embedment-mechanism coefficient  $g_{CHA}(\bar{d})$  are respectively:

$$f_{CHA}(\bar{d}) = \left(1 - \frac{\bar{d}}{d_1}\right)^p \quad (85)$$

$$g_{CHA}(\bar{d}) = \left(1 - \frac{d_2}{\bar{d}}\right)^s \quad (86)$$

According to the authors, given a mixing process, many possible packing states of the dominant packing skeleton exist. The one most likely to occur corresponds to the condition of minimum required work which corresponds to:

$$\frac{\partial V(\bar{d})}{\partial \bar{d}} = 0 \implies -\frac{p}{d_1} \left(1 - \frac{\bar{d}}{d_1}\right)^{p-1} (v_1 - 1) y_1 + s \frac{d_2}{\bar{d}^2} \left(1 - \frac{d_2}{\bar{d}}\right)^{s-1} v_2 y_2 = 0 \quad (87)$$

$\bar{d}$  is calculated thanks to Eq. (87). The specific volume of the binary mixture is then deduced from Eqs. (84) to (86). The calibration procedures carried out by Chang & Deng [34] on different materials have led to the values of  $p$  and  $s$  given in Table 7.

It can be noticed that:

- The *filling mechanism*, the *substitution mechanism* and the *total interaction* criteria are satisfied.
- Only two parameters  $p$  and  $s$  (Table 7) are necessary to estimate the specific volume of the binary mixture.
- Eq. (87) can admit several solutions of  $\bar{d}$  in the interval  $[d_1; d_2]$ .

### 5.13. Liu, Ye, Zhang, Wang, Chen, Cui (2019)

Liu et al. [36] perfected a piecewise model intended primarily for soils. Two scenarios were elaborated by the authors: an *ideal scenario* and a *general scenario*. In the first one, they distinguish the coarse dominant field and the fine dominant field where the *filling mechanism* and the *substitution mechanism* are respectively active. In the second one, the void volume of the mixture is expressed as a quadratic non-linear model with respect to  $y_1$  and  $y_2$ .

If  $r \leq r_0$ , the equations of the *ideal scenario* are Eqs. (5) and (6).

If  $r > r_0$ , the equations of the *general scenario* are the following:

**Table 8**

Parameters  $p$ ,  $s$ ,  $A$ ,  $B$  for different materials according to Liu et al. [36].

Materials	$f_{LUU}$		$g_{LUU}$	
	A	p	B	s
Glass beads	0.670	2.508	0.700	2.065
Silica sand	0.820	2.039	0.681	2.190
Crushed granite	0.622	3.198	0.552	2.832
GMZ pellets	0.718	2.384	0.654	2.233

$$V_1 = 1 + (v_1 - 1) y_1^2 - (v_2 - 1) y_2^2 + 2 (v_2 - 1) y_2 - f_{LUU}(r) [2 v_2 - 1 - (v_2 - 1) y_2] y_2 \quad (88)$$

$$V_2 = 1 + (v_2 - 1) y_2^2 - (v_1 - 1) y_1^2 + 2 (v_1 - 1) y_1 - g_{LUU}(r) (v_1 - 1) [2 - y_1] y_1 \quad (89)$$

The filling coefficient  $f_{LUU}(r)$  and the embedment coefficient  $g_{LUU}(r)$  are respectively:

$$f_{LUU}(r) = A (1 - r)^p \quad (90)$$

$$g_{LUU}(r) = B (1 - r)^s \quad (91)$$

The values of  $p$ ,  $s$ ,  $A$ ,  $B$  are provided for different materials in Table 8 [36].

It can be noticed that:

- Thanks to the piecewise model, the *filling mechanism* and the *substitution mechanism* are satisfied. However, no value of  $r_0$  (called  $R$  in [36]) is provided by the authors.
- The *total interaction* criterion is satisfied when  $v_1 = v_2$ .
- Four parameters  $A$ ,  $B$ ,  $p$  and  $s$  (Table 8) are necessary to estimate the specific volume of the binary mixture.

### 5.14. Chang, Deng (2022)

Chang & Deng solved the problem of fictitious granular classes interacting with each other [34] by appealing to concepts from thermodynamics [35]. The notions of filling-mechanism and embedment-mechanism coefficients are replaced by those of activity coefficients and the equations become the following:

$$V(\bar{d}) = y_1 [v_1 - (v_1 - 1) \alpha_1(\bar{d})] + v_2 y_2 (1 - \alpha_2(\bar{d})) \quad (92)$$

where  $\bar{d}$  is a scalar with a unit of length called the *effective particle size*. According to the authors, the bidisperse packing is then considered as a monodisperse packing with particle size  $\bar{d}$ .

The activity coefficients are respectively:

$$\alpha_1(\bar{d}) = \left(1 - \frac{\bar{d}}{d_1}\right)^\eta \quad (93)$$

$$\alpha_2(\bar{d}) = \left(1 - \frac{d_2}{\bar{d}}\right)^\eta \quad (94)$$

In Eqs. (93) et (94), the particle volume potentials of the two species, analogous to the chemical potentials between species, are governed by  $\bar{d}$  which must be constrained so that the equilibrium of the two potentials is achieved. The excess free volume potential, in analogy to the Gibbs excess free energy, must be minimized at system equilibrium which leads to:

$$\frac{\partial V(\bar{d})}{\partial \bar{d}} = 0 \implies -\frac{\eta}{d_1} \left(1 - \frac{\bar{d}}{d_1}\right)^{\eta-1} (v_1 - 1) y_1 + \eta \frac{d_2}{\bar{d}^2} \left(1 - \frac{d_2}{\bar{d}}\right)^{\eta-1} v_2 y_2 = 0 \quad (95)$$

It can be noticed that:

- The *filling mechanism*, the *substitution mechanism* and the *total interaction* criteria are satisfied.
- Only one parameter  $\eta$  (Table 9) is necessary to estimate the specific volume of the binary mixture.
- Eq. (95) can admit several solutions of  $\bar{d}$  in the interval  $[d_1; d_2]$ .

## 6. General organization of the particle packing models

Table 10 organizes the particle packing models discussed above in this paper according to:

**Table 9**  
Summary of  $\eta$  values as a function of particle shape in the model of Chang & Deng [35].

Coarse particles	Glass beads	Angular	Subangular to angular	Sharp edges, cubical	Angular to subrounded	Subrounded to rounded
Fine particles	Glass beads	Angular	Subangular to angular	Angular, subangular	Thin	Subangular to angular
$\eta$	2.4	2.4	2 to 3	4.2	4.3	2.6 to 5.5

**Table 10**  
General organization of the particle packing models (L: linear, NL: non-linear).

Ref	Authors	L or NL	Filling mechanism	Substitution mechanism	Total interaction	Parameters and leverage	Multi-sized
[47]	Mooney (1951)	L	Yes	Yes	Yes if $\nu_1 = \nu_2$		Yes
[16]	Ben Aim (1970)	L	Yes	Yes	No		No
[54]	Stovall (1986)	L	Yes	Yes	No		Yes
[7]	Westman (1936)	NL	Yes if $G \rightarrow \infty$		Yes if $G = 1$	G	No
[50]	Toufar (1976)	NL	No	No	Yes		No
[44]	Han (1981)	NL	No	No	No		No
[9,65]	Yu (1991, 1996)	NL	Yes	Yes	Yes	Piecewise model	Yes
[14]	Zheng (1995)	NL	No	No	No		No
[51]	Goltermann (1997)	NL	No	No	Yes		No
[53]	De Larrard (1999)	NL	Yes if $K \rightarrow \infty$		Yes	K	Yes
[59,60]	Kwan (2013, 2015)	NL	Yes	Yes	Yes	9 interaction functions	No
[61,62]	Roquier (2016, 2024)	NL	Yes if $K \rightarrow \infty$		Yes if $\nu_1 = \nu_2$	K, $r_0$	Yes
[42]	Wu (2017)	NL	No	No	No		No
[34]	Chang (2018)	NL	Yes	Yes	Yes	p, s	Yes
[36]	Liu (2019)	NL	Yes	Yes	No	p, s, A, B & piecewise model	Yes
[35]	Chang (2022)	NL	Yes	Yes	Yes	$\eta$	No

**Table 11**  
Material parameters.

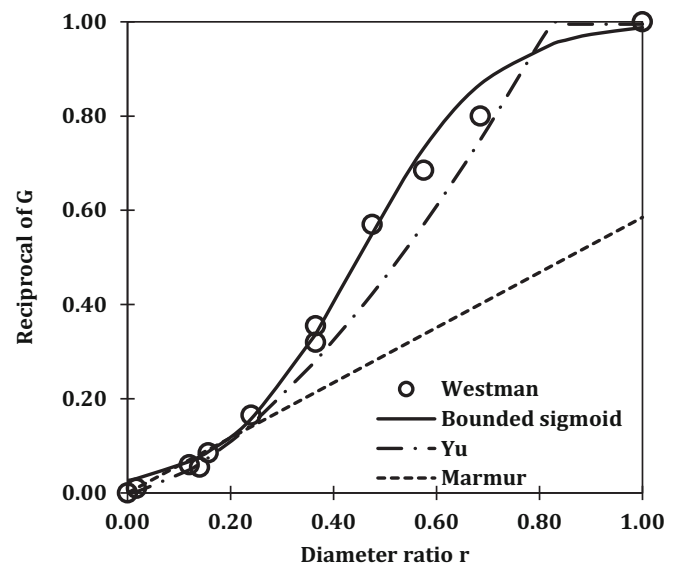
Materials and Models	Data from Jones et al. [67]			Data from Standish & Borger [68] recalled in [67]			Data from Goltermann et al. [51] recalled in [67]		
	Gravel G20	Gravel G10	Sand 1	12.7 mm	9.6 mm	6 mm	8–16 mm	2–8 mm	0–2 mm
Packing process	Loose bulk density – BS 812–2:1995			Hand mix + piston + vibration			30 fall cycles of 9 mm of fall height		
Type of particles	Natural gravel and sand			Smooth wooden spheres			Crushed	Crushed	Natural deposit
Characteristic diameter	14.6	6.76	0.339	12.7	9.6	6	12.6	5.4	0.45
Specific volume	1.686	1.689	1.695	1.724	1.786	1.724	1.559	1.700	1.470

- Their linearity or non-linearity with respect to the solid volume fractions.
- The respect of the filling mechanism (Eq. (5)), the substitution mechanism (Eq. (6)) and the total interaction (Eq. (7)).
- Their parameters and leverage.
- Their extension to the multi-sized mixtures (>3 sizes).

Analysis of Table 10 shows that relatively few particle packing models respect the equations for *filling*, *substitution* and *total interaction mechanisms*: Mooney (without providing equations for  $\lambda_{12}(r)$  and  $\lambda_{21}(r)$ ), Stovall, Westman, Yu, de Larrard, Kwan, Roquier, Chang (2018 and 2022) and Liu, some of them conditionally. Three of them, however, have not yet been extended to multi-sized mixtures: that of Westman (1936), that of Chang (2022) and that of Kwan (except in Wong's PhD Thesis [66]).

## 7. Comparison of five particle packing models for binary and ternary groups

In 2002, in order to optimize concrete mixes by minimizing voids, Jones et al. [67] selected four particle packing models to compare their predictive qualities: Goltermann (i.e. modified Toufar, 1997) [51], Dewar (1999) [52], Stovall (LPDM, 1986) [54] and de Larrard (CPM, 1999) [53]. They applied them to both an aggregate phase and a cement phase. It appeared that each model had a certain size ratio range for which it is in agreement with the test results but that the CPM, modified with a differently calibrated compaction index (modi-CPM), was slightly more relevant (Table 5 in [67]). The experimental data used in this study will now be partially reinvested to make it possible to compare the models already tested in [67] with more recent or unexploited models.



**Fig. 3.** Relation between reciprocal of parameter G and diameter ratio r for experimental data obtained by Westman [7]: comparison with Eq. (32) from Yu et al. [8], Eq. (33) from Marmur [13] and Eq. (96) (bounded sigmoid).

### 7.1. Experimental data

The materials concerned are briefly described here (Table 11) but the interested reader can find additional information in the original publication. They are: (i) a gravel G20, a gravel G10 and a sand called Sand 1 from Jones et al. [67], (ii) smooth wooden spheres of 12.7 mm, 9.6 mm

and 6 mm from Standish & Borger [68] (recalled in [67]), (iii) 8–16 mm, 2–8 mm aggregates and 0–2 mm natural deposit from Goltermann et al. [51] (recalled in [67]).

The characteristic diameters (Table 11) corresponding to each granular range are respectively  $d_{50}$  (50% residue) for the natural gravel and sand of Jones et al. (Table 1 in [67]) and  $d_{63.2}$  (36.8% residue) for the crushed aggregates of Goltermann et al. (Table 1 in [51]). With regard to the latter, the authors [51] in fact consider that many crushed particles follow particle size distribution curves of the Rosin-Rammler-Sperling-Bennett (RRSB) type, characterized by the sieve size on which the residue is equal to 36.8%.

The data concerning mixtures of cement, slag, ash and filler will not be used because the packing density gains obtained with them remain low, in particular due to an unfavorable overlap of their granular distributions.

7.2. Particle packing models tested in the present article

The particle packing models that will now be tested are the following:

- i. Chang (2022) [35] by analogy with chemical thermodynamics.
- ii. Kwan (2013,2015) [59,60] with the 3-parameter Particle Packing Model (3PPM).
- iii. Westman (1936) [7] with a new mathematical relation between  $G$  and  $r$ .
- iv. Liu (2019) [36] with the Non-Linear Particle Packing Model (NLPPM) for multi-sized granular soils.
- v. Roquier (2016,2024) [61,62] with the Theoretical Packing Density Model (TPDM).

7.3. Westman model (1936): proposal of a new relation between  $G$  and  $r$

The Westman conic equation (Eq. (29)) involves a parameter  $G$  whose relation between its reciprocal and the size ratio  $r$  has been established experimentally by its author in 1936 for mixtures of sand and lead shot (Fig. 4 in [7]). However, Westman does not offer an equation. Many years later, Yu et al. [8] and Marmur [13] each proposed a mathematical relation (respectively Eqs. (32) and (33)) shown in Fig. 3. By comparison with these experimental data, it can be seen that Marmur’s proposal is only effective in the size ratio range that he explored, i.e. for  $r$  between 0 and 0.16. Yu’s formula, for its part, underestimates the results for size ratios around 0.5. The proposal made by the author of the present article consists of a bounded sigmoid equation with an inflection point for  $r = 0.45$  but, however, without horizontal asymptotes:

$$G^{-1} = 1 + \exp(-8(r - 0.45)) \tag{96}$$

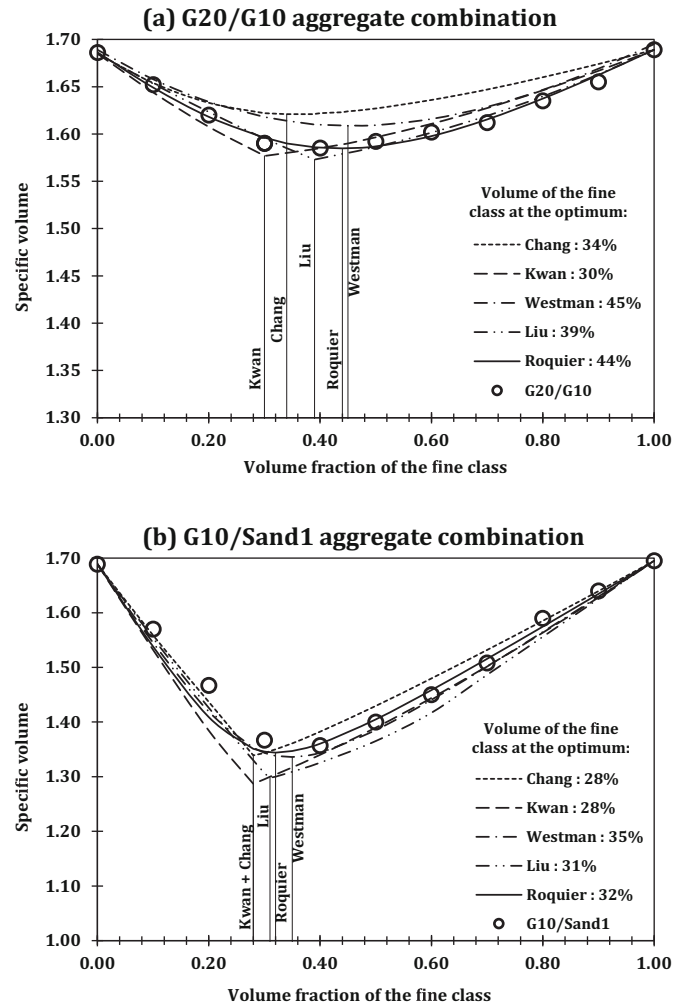
This new formula will be applied in the Westman conic equation.

7.4. Particle packing model parameters

The particle packing model parameters are provided in Table 12.

**Table 12**  
Particle packing model parameters for aggregate.

Materials and Models	Data from Jones et al. [67]			Data from Standish & Borger [68] recalled in [67]			Data from Goltermann et al. [51] recalled in [67]		
	Gravel G20	Gravel G10	Sand 1	12.7 mm	9.6 mm	6 mm	8–16 mm	2–8 mm	0–2 mm
Westman (1936)	$G^{-1} = 1 + \exp(-8(r - 0.45))$								
Kwan (2015)	3 interaction functions for spheres			3 interaction functions for spheres			3 interaction functions for compacted angular particles		
Roquier (2016)	$K = 4.75, r_0 = 0.20$			$K = 9, r_0 = 0.20$			$K = 9, r_0 = 0.03$		
Liu (2019)	2 interaction functions for glass beads with 4 parameters: $A = 0.670, p = 2.508, B = 0.700, s = 2.065$								
Chang (2022)	$\eta = 2.4$ for G20/G10, 8–16/2–8, 2–8/0–2, 8–16/2–8/0–2, 12.7/9.6/6 mixtures $\eta = 4$ for G10/Sand1 mixture								



**Fig. 4.** Comparison of different particle packing models to obtain minimum specific volumes with (a) a combination G20/G10 and (b) a combination G10/Sand1.

Westman’s model involves the new eq. (96). Kwan’s model, for its part, uses interaction functions for spheres in order to study the natural gravel and sand of [67] and the smooth wooden spheres of [68] and interaction functions for compacted angular particles intended for the crushed aggregates of [51]. Roquier’s model involves  $r_0 = 0.20$  for natural aggregates and for spheres,  $r_0 = 0.03$  for crushed aggregates,  $K = 4.75$  for the loose bulk density and  $K = 9$  for a packing process by piston + vibration or by fall cycles. Liu’s model, for its part, was subject of a calibration for four specific materials in Table 8 but preliminary work showed that the one carried out for the spheres was the most suitable in all cases. Finally, Chang’s model recommends  $\eta = 2.4$  for glass-beads/glass-beads and angular/angular particle mixtures (Table 9). This is the value that will be adopted with one exception, as a preliminary study has shown that it

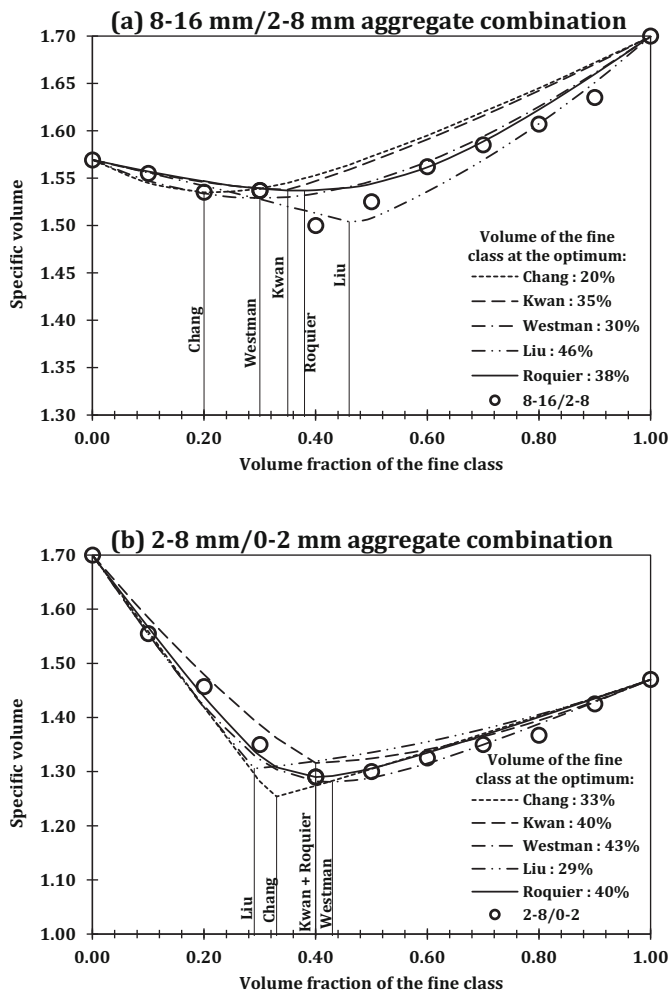


Fig. 5. Comparison of different particle packing models to obtain minimum specific volumes with (a) 8–16 mm and 2–8 mm aggregates and (b) 2–8 mm and 0–2 mm aggregates.

is preferable to select  $\eta = 4$  for the G10/Sand1 mixture.

#### 7.5. Comparison of particle packing models for binary groups of aggregates

Figs. 4 and 5 compare the specific volumes measured and estimated by the five different models for combinations of fine and coarse aggregates.

Fig. 4(a) concerns the mixture G20/G10. It can be seen that the Westman and Chang models overestimate the specific volumes while Kwan's model suggests that a higher coarse fraction is required to achieve the minimum specific volume. The other models lead to good agreement between measured and calculated values.

In Fig. 4(b), the mixture results in a combination between G10 and Sand 1. Chang's model provides good estimates, even if a "sharp" curve emerges with a fraction of sand underestimated at the optimum. These latter trends are found in Kwan's model which underestimates the specific volumes at the optimum and on the side of the dominant coarse particles. Liu's model is overall too optimistic whereas Roquier's model demonstrates a certain efficiency, albeit slightly attenuated on the side where G10 is dominant. Finally, Westman's model seems most suited to studying the specific volume of this mixture. It can be noticed that only the latter two models predict a smooth evolution around the optimum.

Fig. 5(a) concerns the combination of 8–16 mm and 2–8 mm aggregates from Goltermann et al. [51]. Liu's model is the one that best targets the value of the minimum specific volume of the mixture while

managing to approximate the fractions of the constituents at optimum relatively well. Chang and Kwan's models are reliable on the side where the 8–16 mm aggregate is dominant. Westman and Roquier's models provide very similar estimates but they sin near the optimum.

The process is repeated with the combination of 2–8 mm aggregate and 0–2 mm sand in Fig. 5(b). Chang and Liu's models provide a curve made up of two straight sections. The first model is over-optimistic in terms of estimates of specific volume in the vicinity of the optimum, whose composition is imprecisely targeted by the second. Kwan, Roquier and Westman's models are the best suited, the last two achieving an even higher degree of precision.

#### 7.6. Analysis of particle packing models for binary groups of aggregates

Fig. 6 provides an analysis of the difference between the measured and calculated minimum specific volumes for each of the different models (Fig. 6(a)) and the difference in the amount of smaller particles at the optimum (Fig. 6(b)). The particle packing models tested performed well. Chang and Kwan's models have in common that they underestimate the amount of fine elements at the optimum, the former proving slightly better than the latter at predicting the minimum specific volume of the mixture. Liu's model is effective on both the criteria tested, despite a tendency to form peaked curves, which may be slightly offset from the optimum. Roquier and Westman's models are the most reliable on the mixtures tested, with a special mention for the latter, both for its ingenuity and its simplicity.

In Fig. 7, an overall comparison of the differences between measured and calculated specific volumes and void ratios (for comparison with [67]) has been plotted. The mean deviations for void ratio vary between 2.1% and 4.2%, demonstrating reasonable accuracy (in [67], with a supplementary "cement-sized" particle packing, the variation is between 2.4% and 5.5%). According to this criterion, the performance of the Chang, Kwan and Liu models on the one hand (mean deviations for void ratio between 3.7% and 4.2%) and the Westman and Roquier models on the other hand (mean deviations for void ratio equal to 2.7% and 2.1%) are very close. The latter two benefit from less dispersion at low specific volumes, i.e. high packing densities.

#### 7.7. Comparison of particle packing models for ternary groups of aggregates

The study is now extended to three different particle groups. Westman's model has never been developed for mixtures with three particle sizes and will therefore not be considered in this paragraph.

Fig. 8 shows the packing densities obtained by Goltermann et al. [51] for ternary combinations of 8–16 mm, 2–8 mm and 0–2 mm for which the maximum packing density equal to 0.82 is obtained for volume fractions of 50%, 10% and 40% respectively. From the ternary diagrams and the supplementary table provided, it can be seen that Chang and Liu's models slightly overestimate this maximum packing density, while Kwan and Roquier's models target it well. However, none of them is able to estimate the ideal combination, three of the four models even favouring a mixture with only the two extreme grain sizes in order to achieve the most compact mixture.

Fig. 9 shows porosities measured by Standish & Borger [68] on combinations of three series of perfectly monosized smooth wooden spheres of 12.7 mm, 9.6 mm and 6 mm. As these diameter values are relatively close, experience shows that the intermediate size must be omitted to achieve the minimum porosity, which is confirmed by all the models. However, only two of them, Chang and Roquier's models, agree with the experimental volume fractions of the extreme granular classes at optimum. It can be seen that three models significantly overestimate porosities. Only Roquier's model manages to provide isoporosity curves close to reality and to approximate the minimum porosity. It should be noted that the mixing process used by Standish & Borger [68] was particularly elaborate, with hand mix, piston and vibration until the



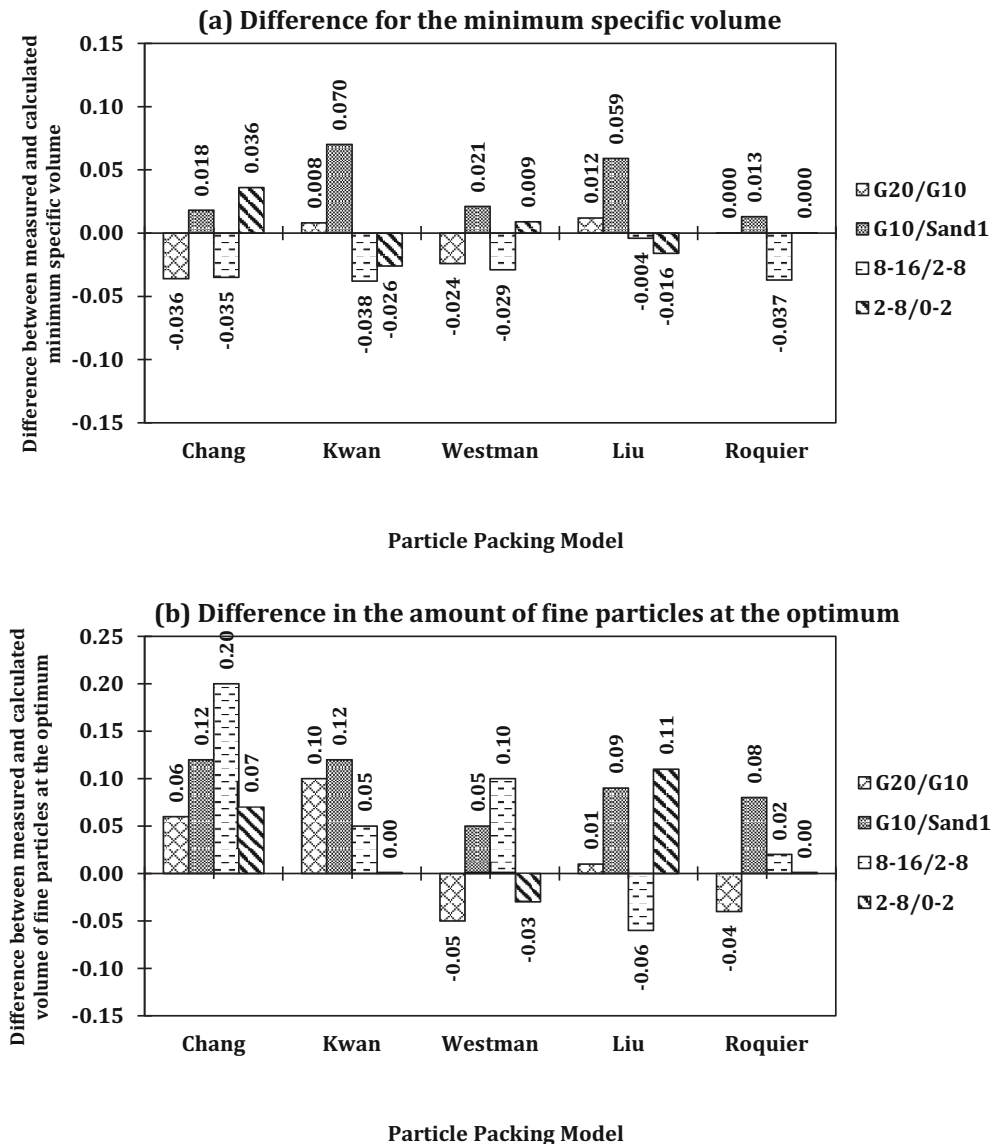


Fig. 6. Differences between (a) measured and calculated minimum specific volumes and (b) measured and calculated amounts of fine particles at the optimum.

height of the mixture was completely stabilised. In this respect, the use of a compaction index makes it possible to adapt more effectively to the packing process adopted.

## 8. Conclusion

In this paper, a review of particle packing models is presented over a 100-year period. Some estimate porosity, others the void ratio, packing density or specific volume of mixtures. The choice made in this paper was to rewrite all the models in terms of specific volume for two reasons. Firstly, the limiting cases can be expressed by straight line equations: the *filling mechanism*, the *substitution mechanism* and the *total interaction*. Secondly, the similarities and differences between models are easier to highlight.

The 100-year history of packing models shows that their design is influenced by their authors' field of research, but that cross-disciplinary approaches have developed over the years. Symbolizing this multi-disciplinarity approach, Westman, an eminent chemistry specialist whose research ranged from coal processing to phosphate glasses and ceramic clays, proposed a conical equation in 1936 that remains a benchmark almost a century later. More generally, chemists and crystallographers have focused on the regular packing of spheres, the

coordination number and the notion of insertion and filling. Very quickly, they established the link between molecular packings and thermodynamics of binary solutions. New concepts based on chemistry and thermodynamics were then developed for granular packings: eutectic, compactivity, excluded volume, statistical approach, etc. In the field of sediments, Han's (1981) precursor model involves both randomness and filling. In the field of soils, Chang (2022) introduced the notion of minimum excess free volume potential for finding an effective particle size of a mixture. Another area of chemistry, that of suspensions, has also contributed to our understanding of granular interactions. By focusing on concentrated suspensions, Mooney (1951) took into account a first-order interaction described as a crowding effect and introduced crowding factors. Expressed differently, the latter lead to Stovall's (1986) geometric interaction functions (wall effect and loosening effect), who developed one of the first efficient packing density models. In parallel with Stovall, Yu (from 1987 to 1996) designed several generations of models, finally combining two of them to create a piecewise model, one of which was inspired by the work of Westman (1936) and Marmur (1985). This idea of piecewise model was taken up by Liu (2019) in the field of soils, who proposed simpler equations. Stovall's model was improved by de Larrard (1999) who, as a concrete specialist, introduced the notion of packing process via a compaction index that

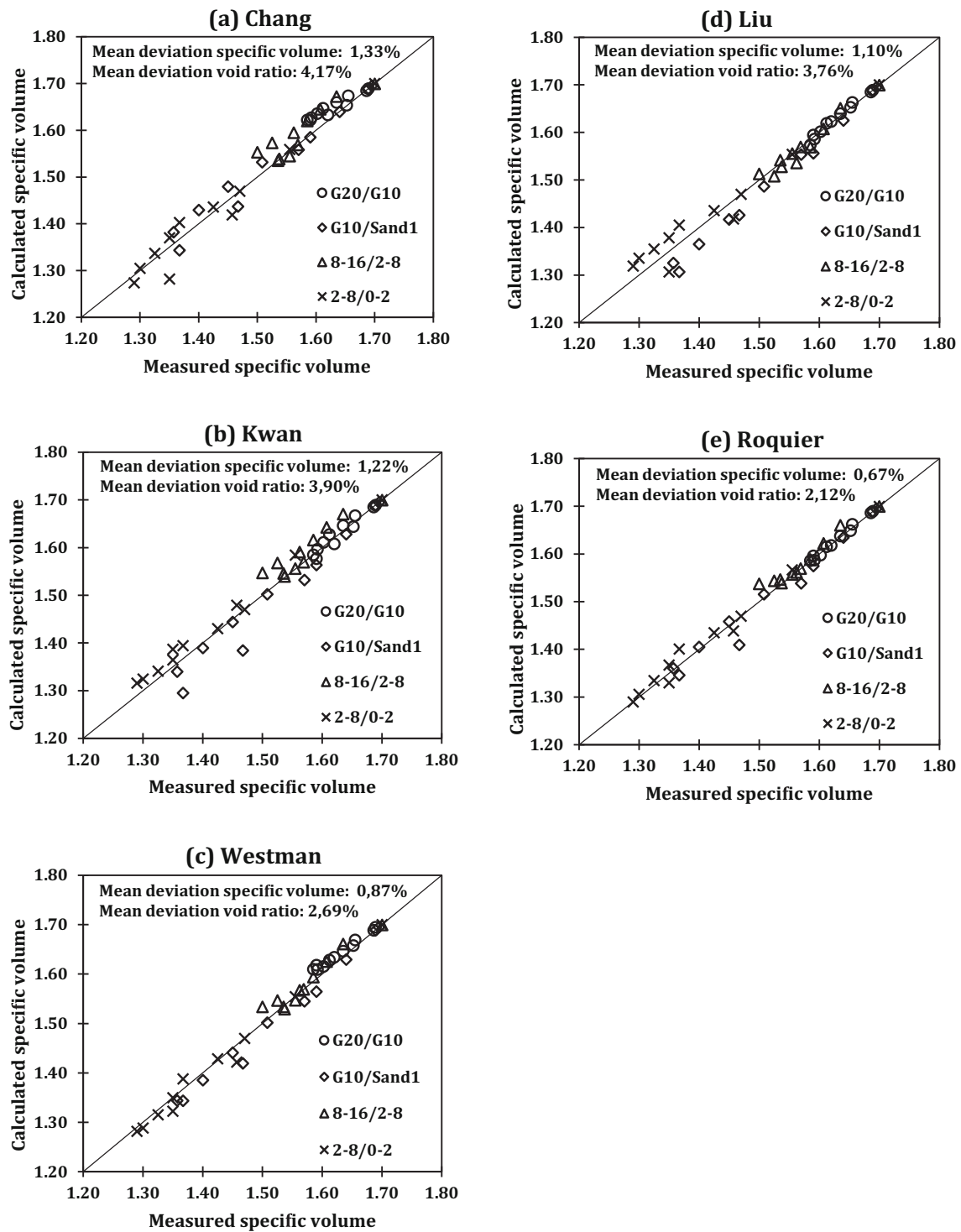


Fig. 7. Total comparison of differences between measured and calculated specific volumes and void ratios for all data for the following particle packing models: (a) Chang, (b) Kwan, (c) Westman, (d) Liu, (e) Roquier.

was much simpler to use than the concept of compactivity. But whereas de Larrard considers that the positioning of the particles in the packing is not perfect because the way in which the mixture is set up is not optimal (external action), Kwan (2013) believes that the problem is intrinsic to the mixture and introduces an additional geometric interaction (wedging effect). Finally, Roquier (2016, 2024) considers it preferable to retain the compaction index introduced by de Larrard. He advocates an individualized calculation of each granular interaction (wall effect, loosening effect) involving the size ratio, the specific volume of each

granular class, the packing process and the shape and surface roughness of the particles. The latter are taken into account via a critical cavity size ratio, which determines whether the insertion of fine particles into the coarse particle skeleton is possible without loosening it.

The particle packing models can be distinguished according to their type (simple or piecewise), their formulation (linear or non-linear with respect to the solid volume fractions), the consideration of (i) two or three (with wedging effect) geometric interactions, (ii) a packing process, (iii) a critical cavity size ratio or a critical ratio of entrance, the

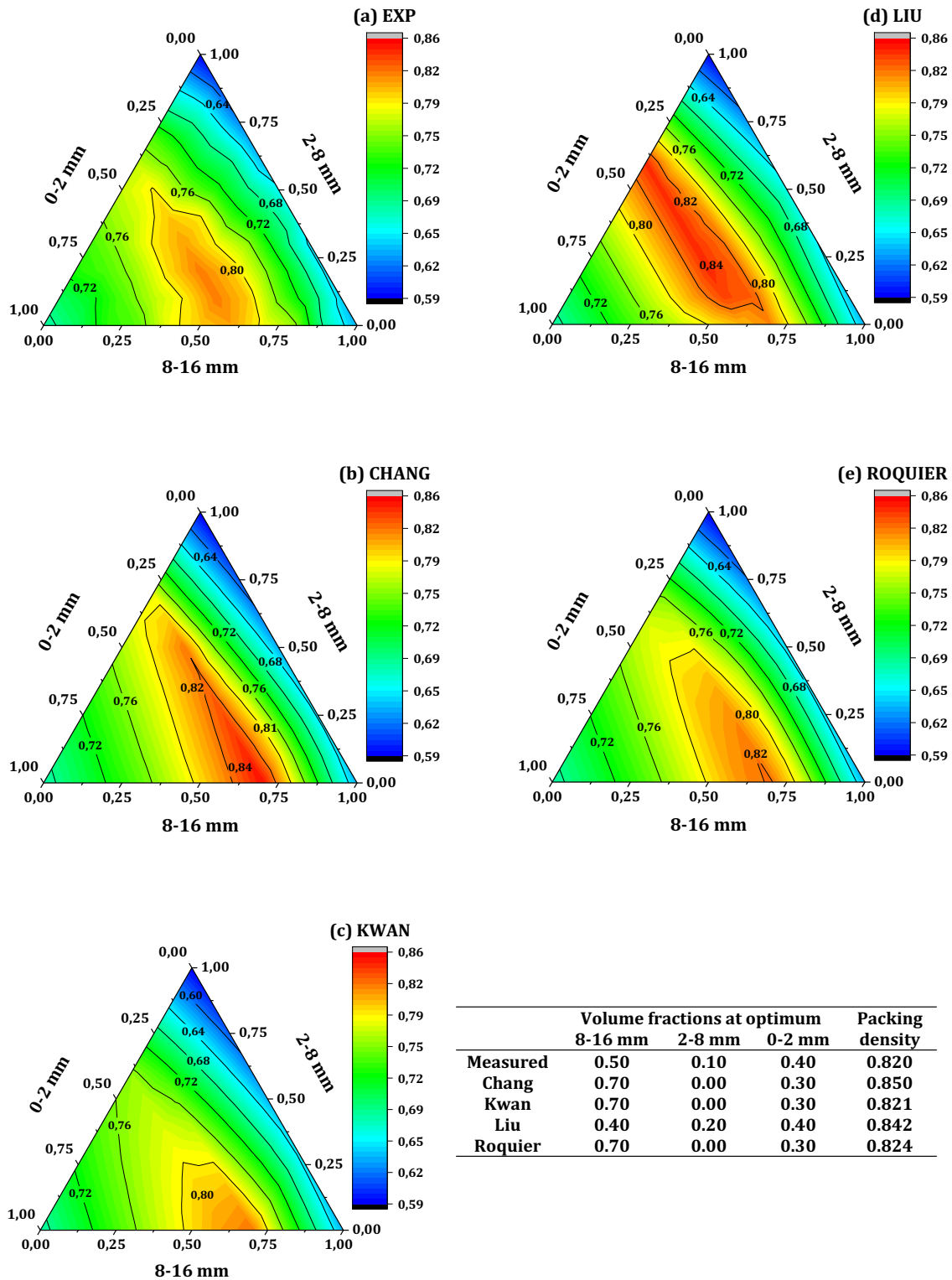


Fig. 8. Comparison of measured (a) packing densities obtained for ternary combinations of 0–2 mm, 2–8 mm and 8–16 mm with those calculated using (b) Chang, (c) Kwan, (d) Liu, (e) Roquier models.

respect of (i) the *filling mechanism*, (ii) the *substitution mechanism*, (iii) the *total interaction* criterion, the physical significance of the parameters taken into account, their generalisation to multi-sized mixtures. Table 10 organizes the main packing models discussed in this paper according to the characteristics listed above.

Five models were then analyzed on binary combinations of coarse and fine aggregates: those of Chang, Kwan, Westman, Liu, Roquier. The

Westman model is tested with a new equation (Eq. (96)) linking  $G$  and the size ratio  $r$ . Data come from the study by Jones et al. [67], which had already made it possible to compare the estimates of four other models. The analysis shows that the new particle packing models are effective in predicting both the minimum specific volumes and the material combinations leading to these optimum mixtures. Two models nevertheless stand out slightly from the crowd: Roquier's and, above all, Westman's

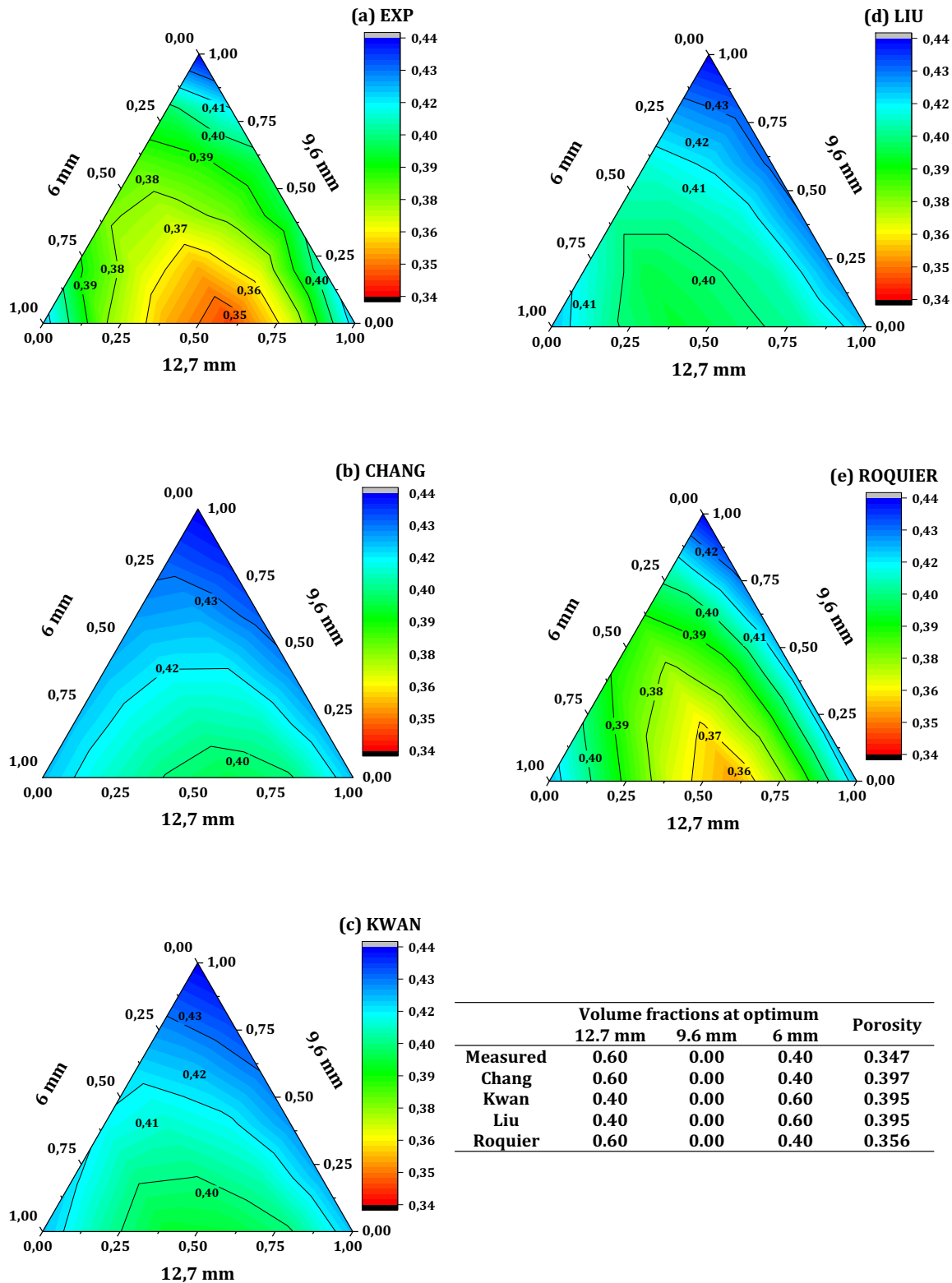


Fig. 9. Comparison of measured (a) porosities obtained for ternary combinations of 6 mm, 9.6 mm and 12.7 mm wooden spheres with those calculated using (b) Chang, (c) Kwan, (d) Liu, (e) Roquier models.

for its ingenuity and simplicity.

The comparison was then extended to ternary combinations of concrete aggregates on the one hand and spheres on the other. Westman's model has not been used in this section, as it has not been developed for ternary mixtures. With regard to the experiments carried out with aggregates by Goltermann et al. [51], the analysis shows that Kwan's and

Roquier's models perform best for estimating the maximum packing density. However, none of the four models succeeded in finding the ideal combination. With regard to the experiments carried out with spheres by Standish & Borger [68], the porosities are difficult to predict for two reasons: the three particle sizes are relatively close, and the mixtures are very carefully placed in the test cylinder. In the end, of the four models



tested, only the one using a compaction index (Roquier's model) provides reliable estimates.

Overall, most of the granular packing models presented in this article have great potential given all the factors actually involved. Nevertheless, some of them deserve to be valued because of their original design, pioneering character, performance, flexibility, simplicity or strong disciplinary impact:

- i. Westman's model for its subtlety in the study of binary mixtures. With a single parameter, he manages to respect all the boundary conditions and to cover all possible diameter ratios. The only major drawback is that this model cannot be extended to multi-sized mixtures.
- ii. Mooney's model for its pioneering character. Although integrated into a viscosity model, this was the first packing model to take granular interactions into account (without their equations, only with their boundary conditions) that could be generalized to multi-sized mixtures.
- iii. Yu's models and Chang's models because they have enabled considerable progress in two sectors of global importance: the powder industry and geosciences respectively. Yu's models have contributed to significant advances in the understanding of powder and particle stacking, a parameter of critical importance in high-value-added scientific sectors such as process engineering, metallurgy and chemical or pharmaceutical engineering. Chang's models, for their part, have made it possible to progress in the estimation of the jamming density and therefore in the identification of a crucial point in soil mechanics: the phase transition point that can lead to rock avalanches or landslide instabilities.
- iv. de Larrard's model and Kwan's model because they have contributed to a scientific formulation of concrete, until then essentially empirical, by integrating the packing process in two different ways. De Larrard's model is particularly flexible thanks to its compaction index.
- v. Liu's model that combines simplicity and precision.
- vi. Roquier's theoretical model for its versatility and for the high accuracy of its estimates.

### CRedit authorship contribution statement

**G. Roquier:** Writing – original draft, Investigation, Formal analysis, Conceptualization.

### Declaration of competing interest

None.

### Data availability

Data will be made available on request.

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