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Fluctuations, structure factor and polytetrahedra in random packings of sticky hard spheres

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ABSTRACT

Sequentially-built random sphere-packings have been numerically studied in the packing fraction interval $0.329 < \gamma < 0.586$. For that purpose fast running geometrical algorithms have been designed in order to build about 400 aggregates, containing 10^6 spheres each one, which allowed a careful study of the local fluctuations and an improved accuracy in the calculations of the pair distribution $P(r)$ and structure factors $S(Q)$ of the aggregates.

Among various parameters (Voronoi tessellation, contact coordination number distribution,…), fluctuations were quantitatively evaluated by the direct evaluation of the fluctuations of the local sphere number density, which appears to follow a power law. The FWHM of the Voronoi cells volume shows a regular variation over the whole packing fraction range.

Dirac peaks appear on the pair correlation function as the packing fraction of the aggregates decreases, indicating the growth of larger and larger regular polytetrahedra, which manifest in two ways on the structure factor, at low and large Q values. These low PF aggregates have a composite structure made of polytetrahedra embedded in a more disordered matrix. Incidentally, the irregularity index of the building tetrahedron appears as a better parameter than the packing fraction to describe various features of the aggregates structure.

1. Introduction

During the remote antiquity, corn trade was made by sacks, implicitly relying on the invariance of the grain volume to the external sack volume, i.e. on the packing fraction of the disordered packing of grains. However it was also known that the seller could win (or the buyer loose) about 10% if the corn were simply poured into the sack instead of being carefully shaken and densified.

Later on, the maximum value of the packing fraction of disordered packings of (sticky) hard spheres – or random close packing – has been experimentally measured between 0.636 and 0.64 [1–3]. However, this value lacks any mathematical demonstration, by contrast with the case of periodic or crystalline arrangement of spheres for which it was recently shown that the maximum packing fraction is $\pi/\sqrt{18} \approx 0.74$ (Kepler conjecture demonstrated by TC Hales [4] and still being verified by several mathematician teams).

On the other hand, the onset of electronic computers about 50 years ago allowed this problem to be numerically tackled. Schematically, two broad families of random aggregate building families exist. The most widely used nowadays – the literature is too abundant to be exhaustively mentioned here – is the family of "dynamic" methods, for which all spheres in the aggregate are included since the beginning, and the system evolves towards equilibrium either by solving equation of motion (e.g. molecular dynamics [5], Lubachevsky–Stillinger algorithm [6]) or on the basis of purely geometrical constraints (e.g. Jodrey–Tory algorithm [7]). The second family, that of static – or sequential – methods, is based on the progressive insertion of spheres in the aggregate, tangentially to three already inserted spheres. In this case, the sphere is immediately assigned its definitive position and various strategies exist to build random systems in this way [8,9]. Such approaches have been proven able to describe the structure of pure or binary liquids and amorphous metals and alloys [10] and are of interest to describe penetration [11], segregation effect [12], growth of tumour [13]…

It turns out that aggregates produced by either family of building-method differ at least in one perspective: the average contact coordination number (CCN) varies roughly between 4 and 7 (see e.g. [14–17]) from the RLP (random loose packing) to the RCP (random close packing) fraction for dynamic systems, whereas sequential methods produce aggregates with an average CCN of 6, whatever the packing fraction [18,8]. Hence, it seems that sequential methods give access to a family of random aggregates that significantly differ from the ones obtained by dynamic methods.

If various approaches have allowed a systematic study of dynamically built random aggregates by controlling some rate parameters to vary

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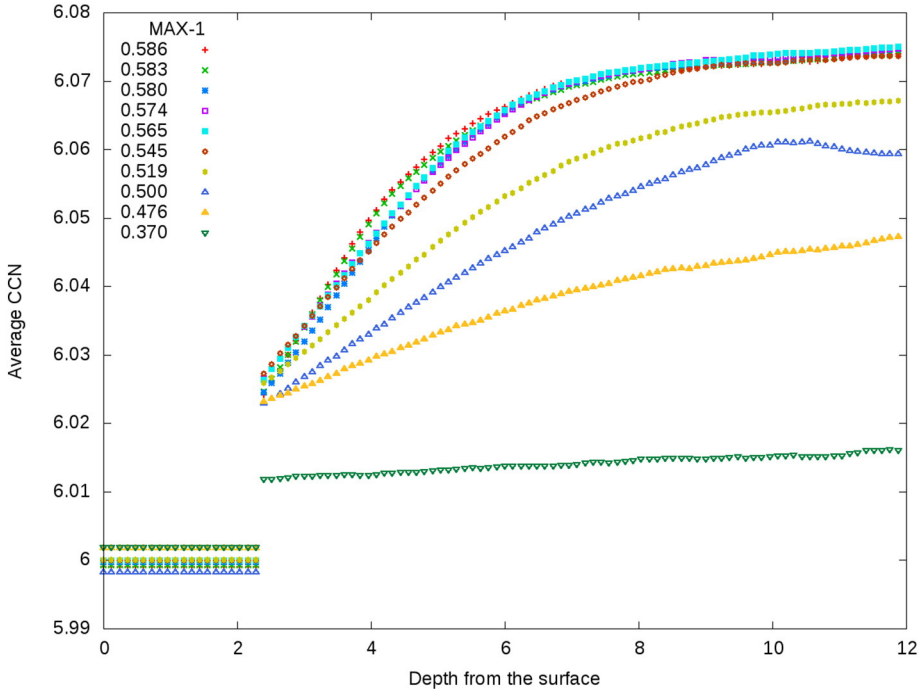

Fig. 1. Variation of $\overline{\eta}$ as a function of the thickness of the layer removed from the aggregate in r_s unit.

progressively the packing fraction of such systems (see, for example, [19]), to the best of our knowledge, such study does not exist in the case of sequentially built random aggregates. In their investigation of packings built with sequential models, Jullien and Meakin [8] were able to produce 5 types of aggregates whose packing fraction varied from 0.5447 to 0.6053 by changing the building procedure (Bennet method, ballistic, anti-Bennet, stable Eden, and Eden methods). The aim of the present study is to analyse several families of sequentially built random aggregates of large number (10^6) spheres, whose packing fraction can be controlled by varying a continuous parameter. These geometrical results will also be of interest to interpret some structural signatures in a more general perspective.

2. Methods

2.1. Building the aggregate

2.1.1. Sphere positioning algorithms

Each spherical "aggregate" or "cluster" with radius R is built by adding spheres (with diameter $d = 2$ or radius $r_s = 1$ in arbitrary unit

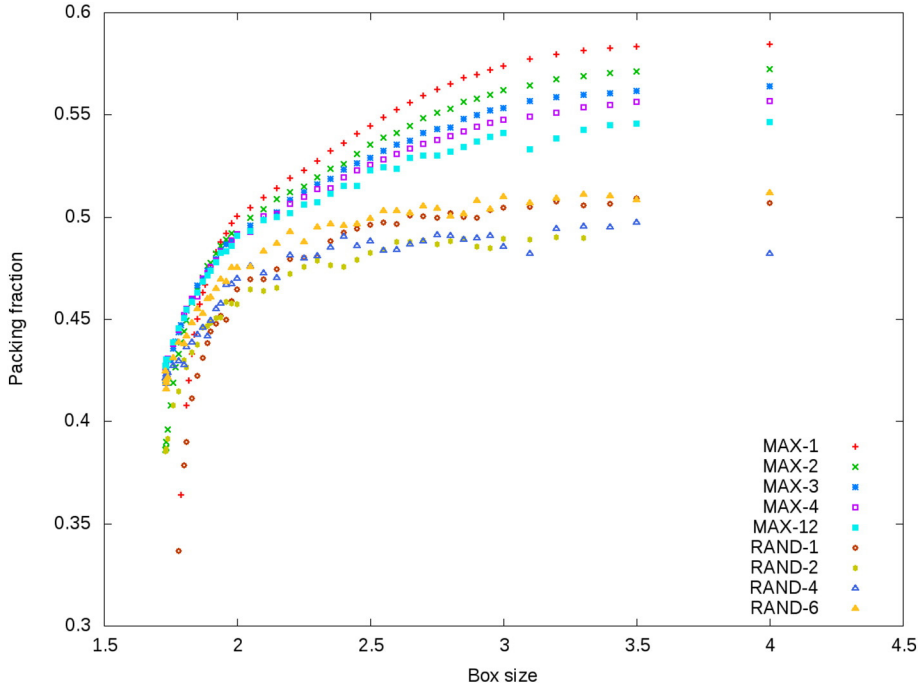

Fig. 2. Packing fraction as a function of the half box-edge length (in r_s unit) for algorithms MAX and RAND with the insertion of 1 up to 12 spheres around an origin.