Antonio Espuña, Moisès Graells and Luis Puigjaner (Editors), Proceedings of the 27th European Symposium on Computer Aided Process Engineering – ESCAPE 27
October 1st - 5th, 2017, Barcelona, Spain 

# Modelling of Aerogels Structures Using Intelligent System «AeroGen Structure»

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## Abstract

In this paper, the development of the framework for modelling of hybrid aerogels structures as well as structure characteristics, implemented in an intelligent system "Aerogen Structure" is presented. The framework integrates five models for generating structures of different types for organic, inorganic and hybrid aerogels (including aerogels based on cellulose, chitosan, alginates), modules for calculating specific surface area and pore size distribution. Thus, comprehensive set of models integrated into systematic framework within intelligent system "Aerogen Structure" provides an interface to perform modelling and validation of aerogel structures.

**Keywords**: aerogels, aerogel structures, structure modelling

## 1. Introduction

Aerogels are unique materials that have potential to be used in different areas. Most important aerogel properties are high porosity (usually more than 90%), low density (0.003-0.5 g/cm$^3$) and high specific surface area (200-1500 m$^2$/g). In recent years, new techniques have been used in order to improve functional characteristics of aerogels that led to production of different types of organic, inorganic and hybrid aerogels (including aerogels based on cellulose, chitosan, alginates, polyurea). Therefore, the research of heat and mass transfer (adsorption, chromatography, extraction, drying, etc.) in aerogels becomes a crucial task for the optimization and scale up of these processes as well as for predicting the properties of materials based on them. Investigating the process at the nanoscale level (aerogel structures) allows us to study heat and mass transfer processes occurring in the pores of aerogels. Different types of models for structure generation have been developed previously as well as models for obtaining certain characteristics of aerogel structures, but no systematic framework exists yet that would incorporate examination of generated structures against each other in terms of visual 3D representations and such crucial characteristics of structures as specific surface area and pores size distribution.

In this study, the systematic framework employs the following structure generation models: overlapping spheres method (OSM), reaction limited cluster aggregation with multiple centers (multiRLA), diffusion limited cluster aggregation (DLCA), ballistic particle-cluster aggregation (BPCA) and Random Walker (RW). Such variety in models makes it possible to identify the most suitable model for different types of aerogels based on their structures that may contain globules (i.e. silica aerogels, polyurea aerogels) or may be represented as combination of fibers (i.e. cellulose aerogels, chitosan aerogels, alginate aerogels). After the structure is generated, its characteristics are obtained using dedicated models for calculating specific surface area and pore size distribution. The combination and integration of these models are implemented in a form of a systematic framework within “Aerogen Structure”. It provides an interface to generation of the aerogel structures at nanoscale level using a set of models and manages all model-based interactions using systematic, robust and efficient approach, which allows examining generated structures from the different angles. The current version of “Aerogen Structure” software tool has modules for structure visualization (for globules and fibers containing structures), report generation and project management of computational experiments.

## 2. Framework

The framework consists of two levels: algorithmic and computational management (Fig. 1). Algorithmic level includes all models used within framework and their implementations in a set of computational tools that eventually produce 3D structures and calculate their characteristics. Computation management level introduces a set of interfaces that provide options to manage computational experiments, build reports and allow visual inspection of generated models.

### *2.1. Structure generation*

Since different approaches and materials are used to produce aerogels, their structure formation can follow several paths and therefore different types of models have to be used. “Aerogen Structure” employs such methods as particle-cluster aggregation (BPCA model), particle-particle aggregation (OSM model), cluster-cluster aggregation (DLCA), particle-cluster aggregation with multiple centres (multiRLA) and Random Walker (RW).

Ballistic particle-cluster aggregation model (Rinewalt et al., 1991) operates with the particles that follow straight line trajectories between starting point and the position of the seed. The cluster that is being formed in this model is filled out very evenly, being nearly disk-shaped.

Overlapping spheres model (Gurikov et al., 2009) can be described in the following way: the volume is being filled with slightly overlapping spheres, after which spheres have to be removed until desired porosity is obtained and links between all spheres exists.

Diffusion limited cluster aggregation model (Written et al., 1981) is well-known and widely used model. The particles following Brownian motion are aggregated into a set of clusters that eventually can encounter each other forming resulting clusters.

Reaction limited aggregation with multiple centres is rather new model that is based on reaction limited aggregation (Meakin and Family, 1987) but introduces multiple seeds or aggregation centers. This model provides an opportunity to take into account different processes occurring between particles but it does that at the price of high computational complexity. Thus, this model is very flexible meaning that it can be adopted according to the needs.

Random Walker is a general purpose algorithm describing the random movement of a single particles. When multiple particles are added to the volume, their paths would form a structure full of fibers-like objects.

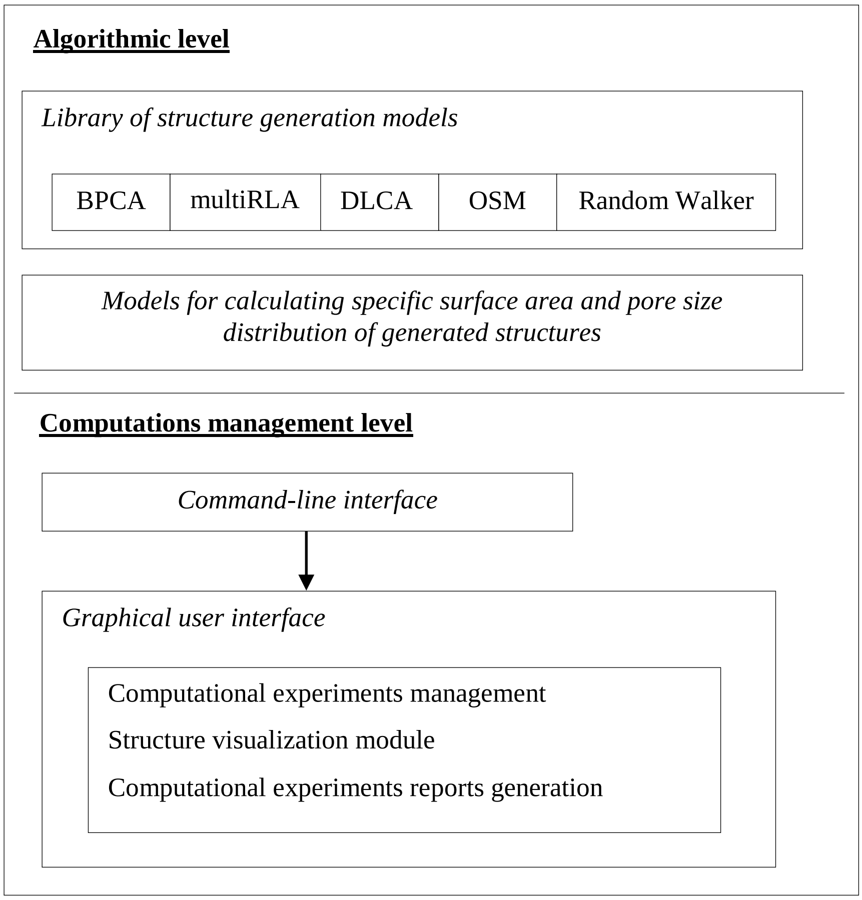



Figure 1. The systematic framework within “Aerogen Strucure”

*2.2. Structure characteristics*

Pore size distribution and specific surface area have been used as main characteristics of generated structures. These parameters usually available during the process of aerogel production and can be used as benchmarks. Models described by Menshutina et al. (2014) have been implemented with slight modifications in order to integrate them into the current framework.

*2.3. Input parameters*

The following input parameters are needed for modelling of the aerogel structure: material type (inorganic, alginate, cellulose, polyurea) that define the structure (whether it is globule containing or fiber containing), desired porosity, radius of a single globule or a fiber (according to the type of aerogel), modelling volume size and models to be used for the structure generation. Depending on the model additional properties are available, such as number of seeds for multiRLA and custom mean length of fibers for RW.

*2.4. Software features*

Besides the structure generation models, such features as report generation (including structure visualizations as well as structure characteristics) and a computational experiments management tool have been implemented in “Aerogen Structure”. These features cover wide range of tasks to be executed in order to conduct a comprehensive computational experiment for the generation of the aerogel structures of different types.

## 3. Case studies

*3.1. Polyurea aerogel*

As a benchmark, polyruea aerogel (Desmodur N3300 sample) reported by Leventis et al. (2010) has been used. The aerogel structures with the following characteristics have been modelled: porosity – 91 %, globule radius – 5 nm, modelled volume – 300x300x300 nm.

Table 1. The structure characteristics obtained using different models

| | **Experimental** | **DLCA** | **OSM** | **multiRLA** | **BPCA** |
|---|---|---|---|---|---|
| **Specific surface area, $m^2/g$** | 302 | 334.4 | 317.8 | 284.7 | 302.7 |
| **Mean pore diameter, nm** | 13 | 15.2 | 14.96 | 148 | 180 |

The results of structure modelling using DLCA and OSM models are in good agreement with experimental data when taken into account the structure charateristics (specific surface area and mean pore diameter) as shown in Table 1.

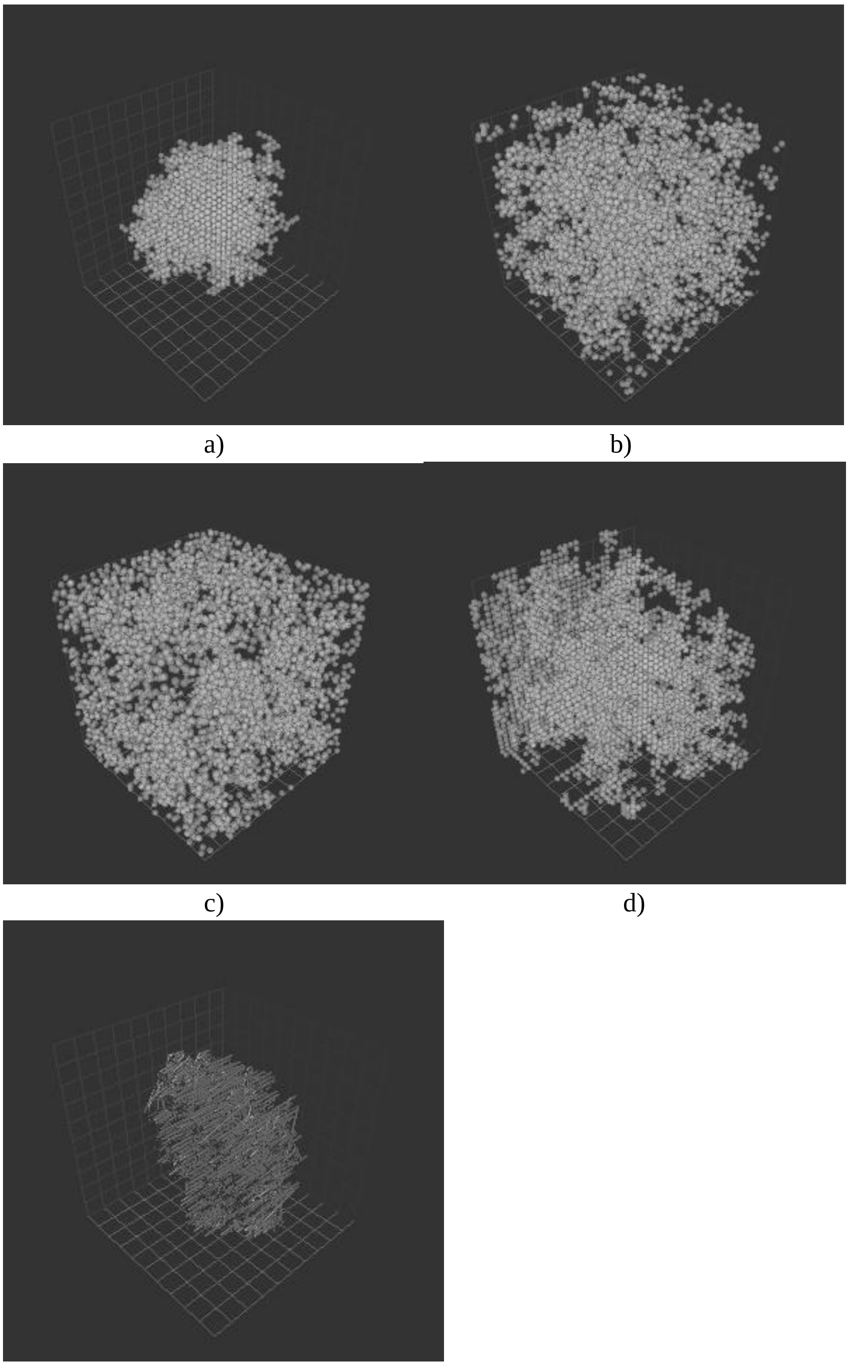
“Aerogen Structure” produces 3D visualization of the generated structures (Figure 2). In the software tool, it is possible to rotate or zoom obtained structures as well as automatically generate a report containing results of modelling.

*3.2. Cellulose aerogel*

Based on cellulose aerogel reported by Sescousse et al. (2011), the structure of aerogel has been modelled using RW model (Fig. 2e). The following input parameters have been used: porosity – 91 %, fiber diameter – 10 nm, modelled volume – 300x300x300 nm. Specific surface area has been calculated and compared against experimental value in Table 2.

Table 2. Specific surface area comparison for cellulose aerogel

| | **Experimental** | **Random Walker** |
|---|---|---|
| **Specific surface area, $m^2/g$** | 300 | 303.4 |




e)

Figure 2. 3D visualization of aerogel structures obtained using BPCA model (a), multiRLA model (b), DLCA model (c), OSM model (d), RW model (e)

## 4. Conclusions

The systematic computer-aided framework for modelling the aerogel structures has been developed, implemented into intelligent system "Aerogen Structures" and validated against experimental data. "Aerogen Structure" employs a set of interfaces to run calculations from command line or from within an application with graphical interface and provides a comprehensive set of features for aerogel structure modelling, computation management and report generation. Future work is to be done on incorporating more material-specific properties of aerogels into models in order to allow more tuning of the generated structures.

**Acknowledgement**. The research is supported by the Ministry of Higher Education and Science of The Russian Federation within the framework of state contract №14.586.21.0028 (unique identifier RFMEFI58616X0028) and by European Commission grant No 685648.

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