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# Data-driven inverse design and optimisation of silica aerogel model networks

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Silica aerogels are highly porous ultralight materials with extremely low density and thermal conductivity. These exceptional properties of silica aerogels are often accounted to microstructure morphology, thus making them of keen research interest for analysing their structure-property relationships. The classical approach for this involved the microstructure modelling of the silica aerogels with aggregation-based modelling algorithm viz., diffusion-limited cluster-cluster aggregation (DLCA) and then performing finite element method (FEM) on the generated representative volume element (RVEs). However, the process often requires large computation time and resources.

The objective of this work was thus to introduce an artificial intelligence approach based on neural networks and reinforcement learning to eliminate the necessity of generating and simulating 3D silica aerogel models for predicting their structural and mechanical properties. To this end for the forward prediction of the elastic modulus and fractal dimension of the silica aerogels from DLCA parameters, an artificial neural network was developed. Furthermore, to reverse engineer the material and perform inverse material design, a reinforcement learning framework was developed, that is shown to have learned to determine appropriate DLCA model parameters as actions for a desired fractal dimension and elastic modulus.



## 1 Introduction

Due to their exceptional material properties, the computational modelling of silica aerogels has been of a keen interest over the years. It has been demonstrated in literature that the diffusion limited cluster-cluster aggregation (DLCA) algorithm effectively models their structural morphology [1] and mechanical properties [2]. The DLCA algorithm is based on the Brownian motion of the particles and clusters, thus mimicking the sol-gel based synthesis of silica aerogels. These generated silica aerogel model networks are then analysed for their mechanical behaviour in FEM tools. However, this approach often requires high computational time and leads to convergence problems during the FEM simulations due to the randomness involved in the creation of backbone within the generated RVEs.

The alternative to the classic modelling approach is by performing data-driven simulations, thus eliminating the need for modelling of the microstructure for each use case and providing the digital twin to perform rapid material design. Data-driven and machine learning (ML)-based material optimisation and inverse design has been implemented over diverse categories of materials. These methods have often involved implemented classic artificial neural networks (ANNs) to predict the material properties [3] or implement complex ML algorithms like generative adversarial networks (GANs) to reconstruct the microstructure based on certain target properties [4]. Zheng et al. [5] recently implemented a data-driven optimisation for the design of macroscopic bodies with an optimised elastic response. This was a remarkable progress in the multi scale modelling of materials and highlights the potential of ML for rapid material design. Initial work on reverse engineering silica aerogels incorporated ANNs and their inversion by back-propagation in input space to retrieve the DLCA model parameters [6]. However, the approach requires knowledge of the initial step of all parameters for the back-propagation, which is not feasible from synthesis perspective. Moreover, the unconstrained inversion approach experiences the classical inverse mapping problem of non-uniqueness.

Reinforcement learning (RL) is a machine learning method where an intelligent agent performs actions in an environment to achieve a maximum reward. This happens in an iterative approach, where the agent optimizes his actions to achieve this goal. RL has been applied for digital material design to inverse design composites [7], automating materials development without prior user-knowledge. RL thus provides a possible solution to the mentioned problems in inverse design of silica aerogels model networks, accelerating the development of silica aerogel with desired mechanical and chemical properties. The AI framework consists of an environment wherein the ANN performs the forward mapping to predict the fractal dimension and elastic modulus from the DLCA model parameters, while an RL agent in combination with the ANN learns to optimise the DLCA parameters for any given fractal dimension and elastic modulus. The proposed workflow of the framework is depicted in Figure 1.

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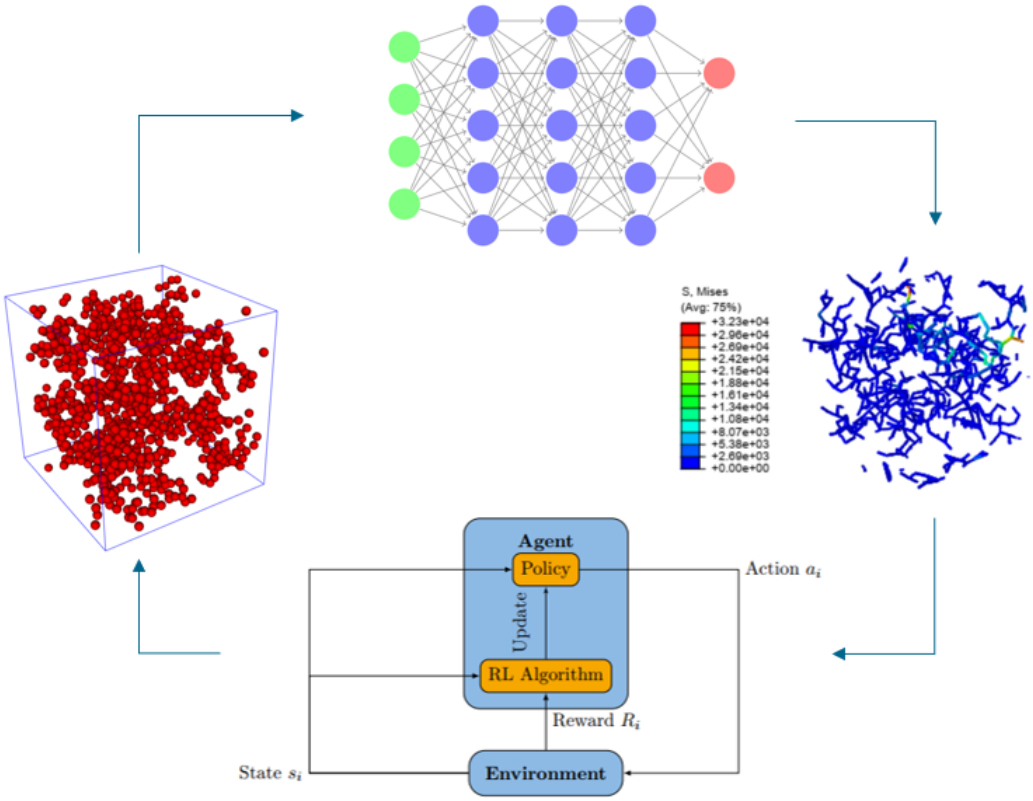


**Fig. 1:** Schematic workflow of the framework: The ANN performs the forward mapping to predict the silica aerogel model network's fractal dimension and elastic modulus, based on the data-set created by DLCA and its subsequent FEM analysis. The RL agent then in combination with the trained ANN performs the inverse design to obtain the DLCA parameters for the desired fractal dimension and elastic modulus.

## 2 Methodology

The overall development procedure of the AI framework can be categorised majorly into three parts:

- Data generation: Generating DLCA-based silica aerogel networks and executing their structural and morphological analysis by determining their fractal dimension and elastic modulus
- Training and validation of the ANN for the forward prediction of the material properties based on the DLCA model input properties
- Training and testing of the RL framework for performing inverse design for specified target material properties

### 2.1 Data generation by DLCA and FEM

A total of 1430 silica aerogel clusters were generated through the DLCA algorithm written in MATLAB. In the DLCA model the 4 variables: radius ($r$), concentration ($c$), step-size of seeds ($s_s$) and the step-size of walkers ($s_w$) were varied in the ranges $r \in [2, 4]$, $c \in [0.04, 0.1]$, $s_s \in [0.1, 0.5]$ and $s_w \in [0.1, 0.5]$ respectively. The fractal dimension of the generated clusters were calculated using the mass $m(r)$ of the particles inside a sphere initiated in the model network. The radius of a sphere is first varied, and then the logarithmic increase of $m(r)$ is recorded. The relationship between $m(r)$ and $r$ is given by a power law $m(r) \propto f_d$, where the slope of the curve (plotted on the log-log scale) corresponds to the fractal dimension ($f_d$). Representatively, 4 cluster structures having a domain size of 100 with different input parameters are shown in Figure 2.
The generated clusters were then exported to ABAQUS as RVEs to perform a uniaxial compression simulation. Each particle connection was modelled as a beam element since beam elements account for the three modes of deformation that bonds undergo: bending, torsion and tension/compression. Therefore, periodic boundary conditions were applied and a displacement boundary condition for the compression was specified. Additionally, frictionless contact between all beam elements was defined. The elastic modulus was extracted by taking the slope of the stress-strain curve as obtained from uniaxial compression of the RVE in the linear elastic region. The calculated fractal dimension and elastic modulus of the networks constitutes the desired properties and also the output of the ANN forward prediction. The detailed methodology behind the FEM and morphological analysis is reported in [2]. In order to eliminate the noise in the output features, the generated data-set was

averaged over same features and pre-processed to remove non converged FEM results, resulting in a final data-set of 470 samples. The final data-set was then split into 70-20-10 % for training, validation and testing of the ANN for the forward predictions.

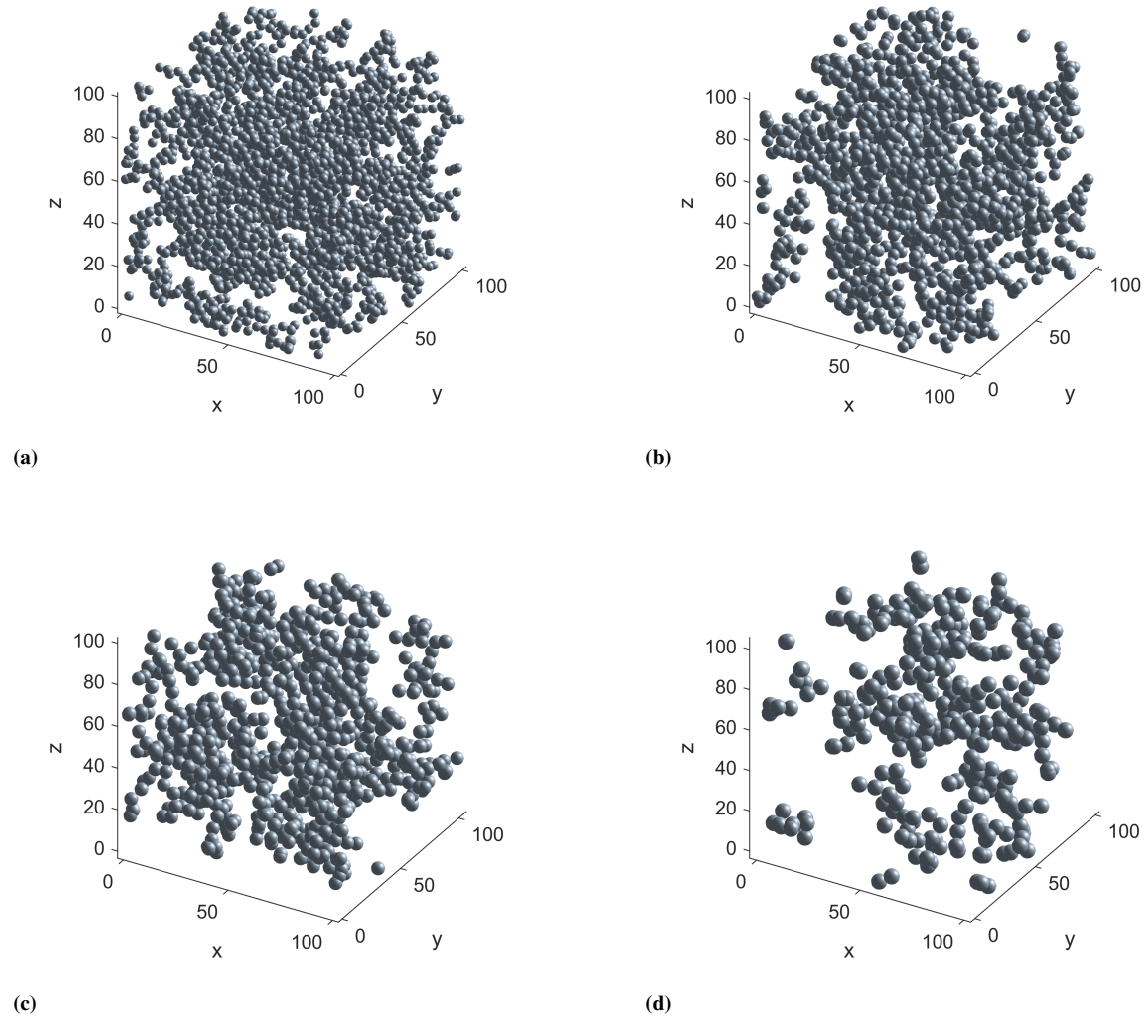


**Fig. 2:** Visualisation of generated cluster with (2a) $r = 2\,\mathrm{nm}$, $\rho = 8.5\,\%$, $s_\mathrm{S} = 0.40\,\mathrm{nm}$, $s_\mathrm{W} = 0.1\,\mathrm{nm}$, (2b) $r = 2.5\,\mathrm{nm}$, $\rho = 8.5\,\%$, $s_\mathrm{S} = 0.40\,\mathrm{nm}$, $s_\mathrm{W} = 0.3\,\mathrm{nm}$ , (2c) $r = 3\,\mathrm{nm}$, $\rho = 8.5\,\%$, $s_\mathrm{S} = 0.40\,\mathrm{nm}$, $s_\mathrm{W} = 0.5\,\mathrm{nm}$ and (2d) $r = 3.5\,\mathrm{nm}$, $\rho = 4\,\%$, $s_\mathrm{S} = 0.10\,\mathrm{nm}$ and $s_\mathrm{W} = 0.4\,\mathrm{nm}$.

### 2.2 Development of the ANN and hyper-parameter optimisation

For performing the forward mapping of the DLCA parameters and the material properties of the silica aerogel networks, a feed forward ANN was developed in PYTHON. The developed ANN had the following architecture: The input layer had 4 units, one for each variable of the DLCA model viz. radius, concentration, step-size of seeds and step-size of walkers. The hidden layer constituted 3 layers with 120, 180 and 160 nodes respectively. The outputs layer had 2 nodes one each for fractal dimension and the elastic modulus. The inner hidden layers had an activating function $f(x) = \frac{1}{1+e^{-x}}$, and it was linear for the input and the output layers. The architecture and the activation functions were selected through the grid search hyper-parameter optimisation to achieve the best metrics. Due to the complex network architecture and less data samples, the ANN was implemented with early stopping so as to prevent over-fit while training. Other hyper-parameters of the ANN are listed in the Table 1. The ANN network architecture is visualized in Figure 3.

### 2.3 Implementation of the RL framework

For implementation of the inverse design of the silica aerogel model networks by optimising the DLCA parameters for the desired fractal dimension and elastic modulus a custom RL environment was setup, as visualised in Figure 4. The environment

**Table 1:** Tuned hyper-parameter of the ANN architecture.

| Hyper-parameter | Value |
|---|---|
| Learning rate | 0.001 |
| Optimiser | ADAMS |
| Metrics | $R^2$ score, mean absolute error (MAE) |
| Loss function | MAE |
| Early stopping patience | 50 epochs |
| Training epochs | 2000 |
| Batch size | 24 |

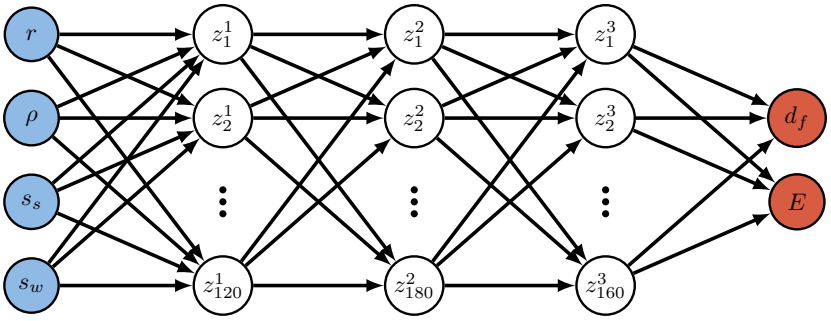


**Fig. 3:** Visualization of ANN network architecture with 4 inputs and 2 outputs.

had 4 actions viz. $r, c, s_w, s_s$ (the DLCA variables) and two states fractal dimension ($f_d$) and elastic modulus ($E$). The RL agent was selected to be the deep deterministic policy gradient agent (DDPG) [8]. The DDPG is an extension of the Deep Q network algorithm: it is an off-policy algorithm based on continuous action spaces, having an actor and a critic network. The RL agent works in tandem with the forward mapping trained ANN: for the current state ($S$) the agent takes an action ($A$), the ANN then predicts the states based on the selected actions and gives a reward as a feedback to the RL agent. Thus, the ANN works to effectively replace the FEM solver environment, thus requiring less computational time and resources. The reward function is designed to assign positive reward values based on the percentile of the relative error between the predicted states and the target state. It is critical that the reward function is continuous and not discrete so as to ensure the RL agent learns to select appropriate actions for the maximum award. The overall training algorithm of the DDPG is reported in [8].

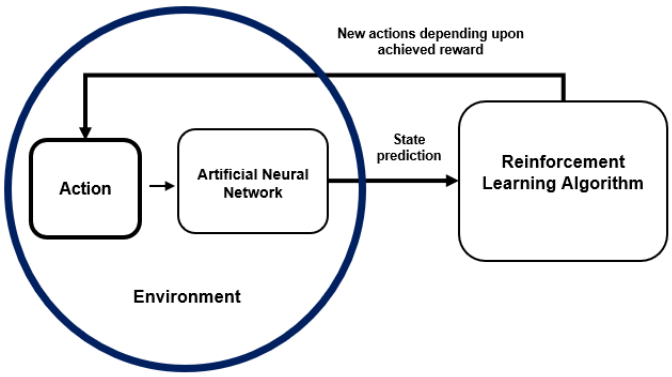


**Fig. 4:** The RL framework for the inverse design of silica aerogel model networks.

## 3 Results and Discussions

Due to the implemented early stopping in the ANN to avoid over-fit, the training stopped at around 850 epochs. The training history and the MAE over the test set is depicted in Figure 5. The trained ANN had a $R^2$ score of 0.9351 and MAE of 3.93 over a test set of 47 samples calculated over both the outputs. It can be visualised through the error histograms in Figure 6 and Figure 7 that the ANN predicts the fractal dimension to a higher degree of accuracy then the elastic modulus. This is

accounted to the high degree of skewness experienced in the elastic modulus training data, which in turn negatively effects the ANN due to the existence of outliers.

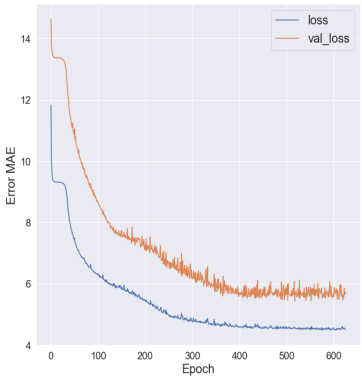


**Fig. 5:** Training and validation curves for the ANN training.

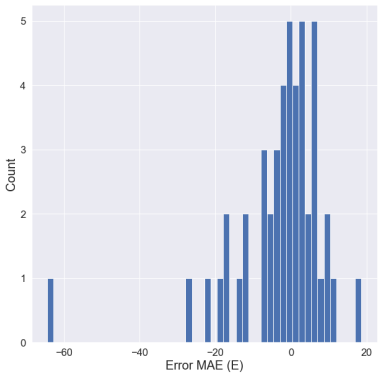


**Fig. 6:** MAE histogram for the predicted elastic modulus.

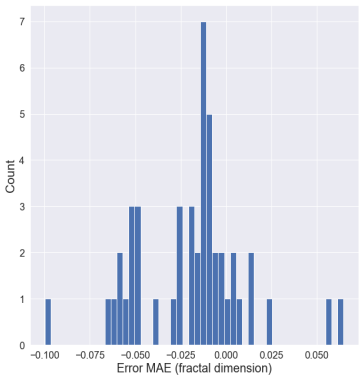


**Fig. 7:** MAE histogram for the predicted fractal dimension.

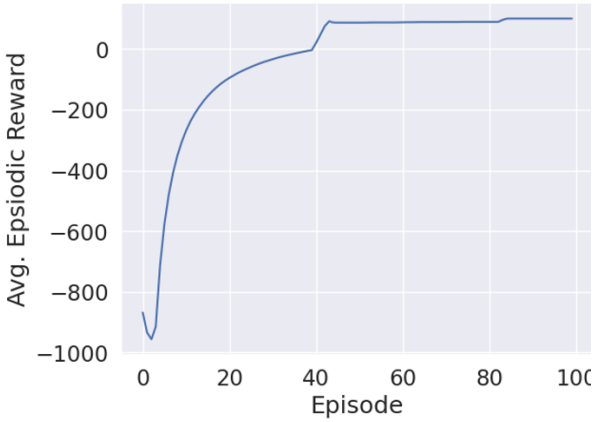


**Fig. 8:** The training history of the DDPG agent.

The RL framework was then trained for inverse design of the model silica aerogel networks with a target fractal dimension and elastic modulus. The target states were selected to be 2.36 and 20.65 MPa for fractal dimension and elastic modulus respectively. The agent trained for 100 epochs, initially achieving negative reward due to the inaccuracy of selecting appropriate actions for the desired states. Over the course of training, the agent then learns to select the actions with maximum reward, thus maximising the average reward return. This training behaviour of the DDPG agent can be visualised in the Figure 8. The final selected actions when validated with ANN yielded the fractal dimension of 2.3571 and elastic modulus of 6.74 MPa. The high absolute error corresponding to the elastic modulus can be accounted to the higher MAE encountered in training and testing the feed forward ANN, which in turn influences the inverse optimisation results as-well. Further comparison analysis with specified target inputs for the DLCA code is still necessary.

## 4 Outlook

The self-learning AI framework based on RL agent learned to optimise the DLCA model variables to achieve the target material properties. However, this initial framework struggles with the accurate prediction of the model network's elastic modulus both in the forward and the inverse directions. This is accounted due to the existence of outliers in the training data-set. The AI model thus struggles with the classical machine learning problem of the quality of the training data-set. Possible solution involves generation of DLCA clusters with bigger simulation box sizes in-order to minimize randomness involved in the generation of the clusters, which in turn directly influence their mechanical stiffness. Additionally, a larger data set would be better suited for further analysis.

Furthermore, the current work is restricted to training the RL agent over one target feature. For a more application-end deployment of the framework it is critical to train the agent over a generalised target function, which can in turn be used for microstructural and topology optimisation.

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