Identifying Nanoparticle Geometry from Emissivity

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Abstract

A crucial step in nanoparticle synthesis is to verify whether the synthesized particles are of the desired shapes and sizes, since the morphology of nanoparticles largely determines their function. Currently, such verification depends on complex analytical measurements, such as transition electron microscopy (TEM).

With machine learning, we can infer the shapes and sizes of nanoparticles from simpler, cheaper emissivity spectra measurements. Physically, a particle's morphology determines its optical properties, which are reflected in the emissivity spectra. Based on physical simulation data, we learn inverse models to identify nanoparticle geometry from emissivity. We find that a ResNet-based convolutional neural network performs well on shape classification (85.9% accuracy), while both random forest and ResNet are comparable for size regression (0.91 correlation). Our models also discover scientific insights on which wavelengths are distinct to different nanoparticle shapes and sizes.

1 Introduction

Nanoscale materials find applications in diverse areas, from conversion of light into electricity in solar cells to the visualization of cancer cells. Nanoparticles are broadly defined as particles that have dimensions in the nanometer range. We focus on these three materials in this study: *Gold (Au), silicone dioxide (SiO_2), and silicon nitride (SiN)*, which have many applications as nanoparticles. Due to their stability and low toxicity [1], gold nanoparticles are desirable for drug delivery, biological imaging, and solar cells. SiO_2 and SiN nanoparticles also have many applications to solar technology [2, 3].

In most nanoparticle applications it is crucial to synthesize nanoparticles of the specified sizes and shapes, often to the nanometer level precision. Usually, researchers turn to more complicated and time consuming methods, such as transition electron microscopy (TEM), to attain this level of detail.

We seek a simpler approach to verify particle shapes and sizes in nanoparticle synthesis, by *leveraging machine learning to extract size and shape information from emissivity spectra*. The emissivity spectra are simpler to obtain experimentally than direct analytical TEM measurements. The spectra represent a particle's emissivity at different wavelengths, which are highly dependent on the chemical composition, size, and shape of nanoparticles [4]. This clear connection between a particle's emissivity and its geometry makes emissivity a good candidate for building models to automate the analysis of sizes and shapes in nanoparticle samples.

While particle emissivity can be simulated by numerically solving Maxwell's Equations [5], there is no simple known rule for determining the sizes, shapes, and materials of nanoparticles from a sample's emissivity spectrum. Therefore, we use numerical simulation to construct a large volume of training data, and then train machine learning models to solve the inverse problem, i.e. to identify particle shapes and sizes from spectra.

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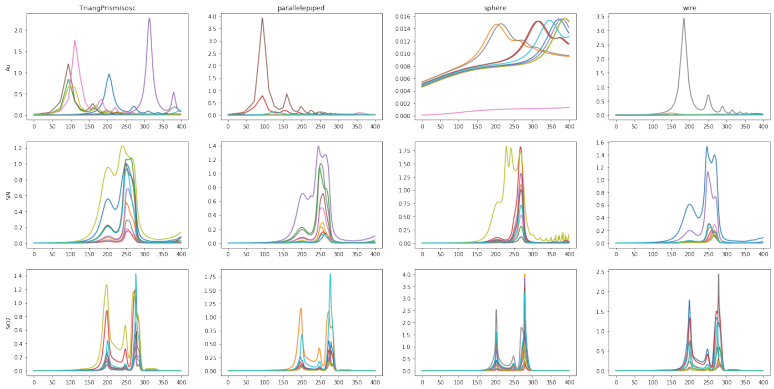

Figure 1: 10 randomly sampled emissivity spectra from each class. Columns (shapes): triangular prisms, parallelepipeds, sphere, and wire. Rows (materials): Au, SiN, and SiO_2.

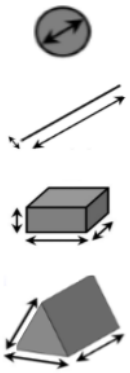

Figure 2: Four nanoparticle shapes in the dataset.

Our contribution We build an inverse model as an experimental analytics tool, where a user can input an experimental emissivity spectrum of a nanoparticle sample and determine the shape, size, and material of their sample. In addition to numerical simulation data, we also include data augmentation by a forward model [5] that predicts the numerical simulation results. We compare random forests and 5 neural network architectures including fully connected networks and convolutional neural networks. We provide the first machine learning based inverse models for automated emissivity spectrum analysis. Our models achieve 85.9% top-1 accuracy on shape classification and 0.91 correlation on size regression.

2 Related work

Most related to our work, Elzouka et al. [5] study the inverse problem in a different context, where they want to design nanoparticles to achieve certain optical properties. Due to the different motivation, their model is not optimized for automated spectrum analysis, and the model accuracy is measured in the spectrum space (by comparing target spectrum to the spectra of the predicted particles). Their approach to the inverse problem is also different: After training a decision tree model for the forward problem, they perform inverse design by finding the output leaf on the decision tree whose value corresponds closest to the target spectrum.

Additionally, He et al. [6] use 2-layer fully connected neural networks for both forward prediction of optical properties and inverse prediction of nanoparticle dimension parameters. He et al. focus on size regression of gold particles. We study a more general prediction problem on a larger dataset (their dataset contains 3118 nanoparticles and 57 points on each spectrum, compared to 160,000 nanoparticles in ours), and also compare more diverse machine learning models.

3 Methods

Dataset Construction. Our dataset was taken from [5] with permission from the authors and constructed by solving Maxwell's equations for the emissivity spectra of 15625 particles. The dataset was evenly divided between three materials: gold (Au), silicone dioxide (SiO_2), and silicon nitride (SiN); with the same shape distribution per material: 38.5% triangular prisms, 38.5% parallelepipeds, 15% wires, 8% spheres (shapes visualized in Figure 2). The size of each particle is determined by four parameters: the log of the area over volume, and the shortest, middle, and longest dimensions.

The emissivity spectrum of each nanoparticle is represented by the emissivity at 400 different angular frequencies, chosen in a logarithmic spacing from 10^{13} rad/s to 0.8×10^{14} rad/s. These angular frequencies can be converted to wavelengths, and represent a wavelength region from roughly 1900 to 19000 nm, meaning these emissivity values are located in the infrared region.

We use 50% of the numerical simulation data (7812 particles) for training, 25% (3906 particles) for validation, and the remaining 25% for test.

We use the forward design model trained on the same numerical simulation dataset in [5] as data augmentation. This model was then used to predict spectra for $20 \times 7812 \approx 160,000$ particles as additional training data.

Prediction task. Given the 400-point emissivity spectrum as input, we want to predict the shape, material, and dimensions (size) of the nanoparticle. We measure classification performance by top-1 accuracy, and we measure size regression performance by mean square error (MSE).

Random forest models. We use random forest classifiers and regressors from scikit-learn [7] to predict material and shape classification and size regression. The random forests have 50 trees, with 20 of the 400 spectral inputs available for each tree. Changing other hyper-parameters from scikit-learn defaults does not improve model performance, and therefore were left at default values.

Fully-connected networks. We include three types of fully-connected networks in our model comparison: a one-layer neural network with 512 hidden units, a two-layer neural network with 512 and 128 hidden units, and a three-layer neural network with 512, 256, and 64 hidden units. All of them use dropout and ReLU activation.

Convolutional neural networks. We adapt the popular AlexNet [8] and ResNet [9] architectures from the computer vision community by replacing 2D-convolutions with 1D-convolutions. For AlexNet, we keep the kernel size and stride fixed from the original AlexNet architecture, but reduce the number of channels. For ResNet, we replace the 3x3 2D-convolution with 1D-convolution of kernel size 3 in the basic block. and keep the overall ResNet-18 architecture.

In addition to the 400-point emissivity data, we also include log emissivity, and the finite-difference derivatives of emissivity and log emissivity in the inputs. To investigate the effects of including the additional features, we report ablation result in Table 1 for ResNet18-1D on only raw emissivity data as "ResNet18-1D (1 channel input)".

After a coarse hyperparameter search within our computing budget, we use Adam optimizer with learning rate 1e-4 and batch size 64 for 20 epochs for neural networks.

4 Results

Material classification proved very easy for random forest models as well as neural networks. The random forest model produced 100 percent classification accuracy and most neural networks attempted also achieve >99% material classification accuracy.

Shape classification and size regression results are shown in Table 1 and Table 2. The ResNet-based model achieves the highest 85.9% top-1 accuracy for shape classification, while being comparable to the random forest model in size regression. The overall MSE of the random forest model corresponds to a 0.91 correlation between the predicted values and the true values.

The shape classification and size regression accuracy largely depends on the shape. For spheres, both classification and regression are near perfect, with 99% classification accuracy and 0.004 MSE. In contrast, triangular prisms have by far the largest MSE (6.08), and are more often confused with parallel pipes in shape classification.

Why does random forest work well for size regression but not shape classification? For size regression, the emissivity at certain wavelengths already contains a lot of information for recovering log(area/volume) and shortest dimension. When examining feature importance in random forest, a particular wavelength is extremely useful with feature importance of over 0.4. This is especially striking when compared with the shape classification, where no single wavelength has a feature importance above 0.04. This gives a hint that more complex models, such as convolutional neural networks, might be necessary for identifying the full nanoparticle geometry.

Multi-task training. We experiment with multi-task training by using a multi-head model to predict shape classification, material classification, and size regression at the same time. Contrary to our expectation that multi-task training would increase performance, it degrades performance in both shape classification and size regression. For ResNet18-1D, the shape classification accuracy drops from 85.9% to 81.8%, while the size regression average MSE increases from 2.96 to 3.47.

Model	Classification Top-1 Accuracy				
	Triangle Prism	Parallelpiped	Sphere	Wire	Average
Random forest	0.72	0.63	0.99	0.78	0.78
One-layer neural network	0.549	0.177	0.938	0.578	0.561
Two-layer neural network	0.645	0.216	0.919	0.478	0.565
Three-layer neural network	0.615	0.177	0.938	0.486	0.554
AlexNet-1D	0.487	0.385	0.997	0.796	0.667
ResNet18-1D	0.803	0.768	0.997	0.867	**0.859**
ResNet18-1D (1-channel input)	0.762	0.691	0.993	0.888	0.834

Table 1: Shape classification top-1 accuracies when trained on all materials combined.

Model	Mean Square Error			
	Short Dim	Middle Dim	Long Dim	log Area/Vol
Random forest	**0.068**	0.296	16.113	**0.029**
One-layer neural network	0.269	1.517	31.334	0.182
Two-layer neural network	0.252	1.894	32.443	0.302
Three-layer neural network	0.289	2.177	31.987	0.372
AlexNet-1D	0.317	1.408	18.003	0.307
ResNet18-1D	0.098	**0.263**	**11.414**	0.055

Table 2: Size regression mean square error.

Joint training on materials. When we compare joint training on all three materials together against training separate models for each material, we find no significant difference in model performance, likely because the spectra for different materials are highly distinct as we have seen in the near perfect material classification results.

Effects of data augmentation. 20x data augmentation increases shape classification performance on ResNet18-1D (from 83% to 85.9%, but beyond that model performance does not increase with more data augmentation, even when we increase the data augmentation ratio to 300x.

5 Discussion

Machine learning for material simulation and analysis is an exciting research direction. We have built models capable of reliably predicting material, size, and shape of nanoparticles from their emissivity spectra. Our work shows that automated nanoparticle emissivity spectral analysis with machine learning has the potential to replace more time intensive analytical techniques.

We also provide a starting point to navigate the design choices in models and training procedures for the nanoparticle geometry identification problem. In particular, we show that data augmentation by a forward model can increase the performance of the inverse model.

In addition to being a practical tool in nanoparticle synthesis, the interpretable random forest models also discover scientific insights about the interactions between nanoparticles and light. The wavelengths at indices 293 and 269 contain crucial information for determining log(area/volume) and the shortest dimension of the nanoparticle, respectively. The random forest models also shed light on what emissivity wavelengths are distinct to the different shapes. Strikingly, the same wavelength at index 269 is the most important wavelength in size regression for the shortest dimension as well as in sphere classification. This knowledge helps nanoscientists build increased understanding of how emissivity at certain wavelengths is connected to the morphology of nanoparticles.

Together, our results suggest that machine learning models are not only useful predictive tools in automated spectrum analysis, but also valuable tools for analyzing experimental data and gaining new scientific insights.

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