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Synthesis of aluminum nanoparticles using spark discharge for applications in ultraviolet plasmonics

M. Nouraldeen✉, N.S. Shestakov, O.V. Vershinina, V.V. Ivanov

Moscow Institute of Physics and Technology (National Research University), Dolgoprudny, Russia

✉ messannouraldeen@phystech.edu

Abstract. This work demonstrates synthesis Al metal nanoparticles with plasmon resonance in the ultraviolet region by the spark discharge method in an argon atmosphere. The resulting primary particles have an Al metal core and a natural oxide shell and size in range from 5 to 50 nm. Importantly, these nanoparticle ensembles show wide extinction peaks, with the highest point between 250 and 480 nm wavelength. The position of the peak can be varied by synthesis parameters. During our research, we employed laser radiation at a wavelength of 355 nm, with pulse energies reaching up to 350 μJ and pulse repetition rates of up to 2000 Hz. We observed that the sintering process of nanoparticles exhibited a dynamic change in size, which correlated with the energy of the laser pulses. This dependence was illustrated by an S-shaped shrinkage curve. By subjecting the initial agglomerates to a series of impacting laser pulses, we successfully achieved complete sintering, resulting in the transformation of the agglomerates into spherical nanoparticles.

Keywords: aluminum nanoparticles, nanoparticles sintering, laser sintering, plasmon resonance

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Синтез наночастиц алюминия с использованием искрового разряда для применения в ультрафиолетовой плазмонике

М. Нуралдин✉, Н.С. Шестаков, О.В. Вершинина, В.В. Иванов

Московский физико-технический институт (национальный исследовательский университет), г. Долгопрудный, Россия

✉ messannouraldeen@phystech.edu

Аннотация. В данной работе продемонстрирован синтез металлических наночастиц Al с плазмонным резонансом в ультрафиолетовой области методом искрового разряда в атмосфере аргона. Полученные первичные частицы имеют металлическое ядро из алюминия и оболочку из натурального оксида, а их размер находится в диапазоне от 5 до 50 нм. Важно отметить, что эти ансамбли наночастиц демонстрируют широкие пики экстинкции с наивысшей точкой между длиной волны 250 и 480 нм. Положение пика можно варьировать параметрами синтеза. В ходе исследований мы использовали лазерное излучение с длиной волны 355 нм, с энергией импульсов до 350 мкДж и частотой следования импульсов до 2000 Гц. Мы заметили, что процесс спекания наночастиц демонстрирует динамическое изменение размера, которое коррелирует с энергией лазерных импульсов. Эту зависимость проиллюстрировала S-образная кривая усадки. Подвергнув исходные агломераты серии воздействующих лазерных импульсов, мы успешно добились полного спекания, в результате чего агломераты превратились в сферические наночастицы.

Ключевые слова: наночастицы алюминия, спекание наночастиц, лазерное спекание, плазмонный резонанс

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Introduction

Al nanoparticles are interesting due to their light absorption properties in the UV range, which has applications in light harvesting using solar cells, as well as enhancing light absorption in thin silicon solar cells [1, 2]. The reason behind this property is that Al nanoparticles are capable of absorbing light as a result of their interaction with conduction electrons and surface plasmon resonance, making them effective light absorbers. In addition, Al nanoparticles with different sizes have different optical properties [3]. Al nanoparticles that are small in size will act as excellent UV absorbers, while for bigger particles, this would be the role of an optical scatterer. There is active research in this area, and emerging applications of plasmonics are still being identified through ongoing research and development activities.

Materials and Methods

The spark discharge method was used to obtain Al nanoparticles [4]. Synthesis was carried out in argon of purity (99.9999%) at pressure of 1,2 atm and flow of 50 mL/min with the following conditions: capacitor of 107 nF, pulse repetition rate of 500 Hz and discharge voltage of 2 kV. The high purity of the starting material and gaseous medium, together with the vacuum tightness of the gas path and the synthesis chamber, ensured the highest possible purity of the nanoparticles under study during the experiment.

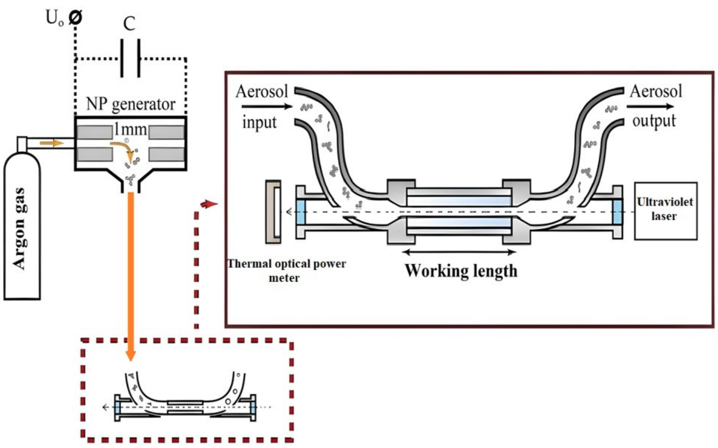

Fig. 1. Diagram of experimental setup: the inset shows a schematic representation of laser optimization cell combining aerosol flow and optical radiation

Schematically, the entire experimental setup is shown in Fig. 1, including the electrical part, the optical part for the spectroscopic tasks, and the necessary measurement options. Pairs of hollow aluminum cylinders with an outer diameter of 4 mm and an inner diameter of 2 mm were used as consumable electrodes in the setup. To conduct studies of the interaction of optical radiation with aerosol nanoparticles a specially designed laser modification cell was used, which made it possible to combine the aerosol flow with optical radiation along its length. The construction of laser modulation cell is described in [5]. The work used an aerosol flow Q = 50 ml/min, which defines the value for the speed of movement of nanoparticles in the aerosol in the working area inside a quartz capillary: υ = 117.9 mm/s.

The agglomerates size distribution in the flow was measured using a TSI SMPS 3936 Aerosol spectrometer. The impact on aerosol Al NPs was studied by nanosecond pulsed laser (CNI AO-355A) with wavelength of 355 nm, pulse repetition rate in the range of 0.2 to 10 kHz with a step of 100 Hz, pulse width of 15 ns and pulse energy in the range of up to 350 μJ. Aerosol HEPA filter were installed in the cell to collect NPs after the cell and deposited particles on it for 30 minutes, with laser frequencies 200, 1500 and 2000 Hz. Absorption spectra of nanoparticles in isopropanol solution were obtained using a JASCO V–770 spectrophotometer.

Results and Discussion

The change in mean diameter of the agglomerates with varying 355 nm laser energy due to sintering shows a size reduction from ~460 to ~290 nm (Fig. 2).

a) *b*)

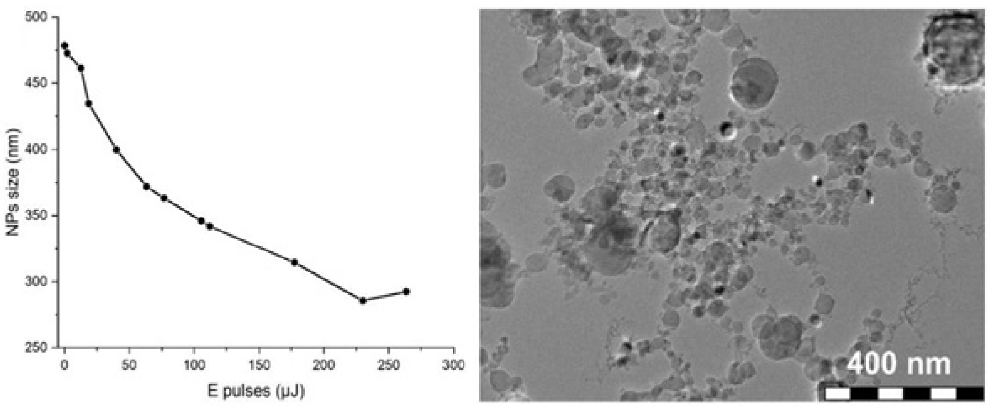

Fig. 2. Dependence of the average particle diameter of the agglomerates on the laser power (*a*), TEM image of typical particles (*b*) at laser pulse frequency of 1500 Hz and gas flow of 50 mL/min

a) *b*)

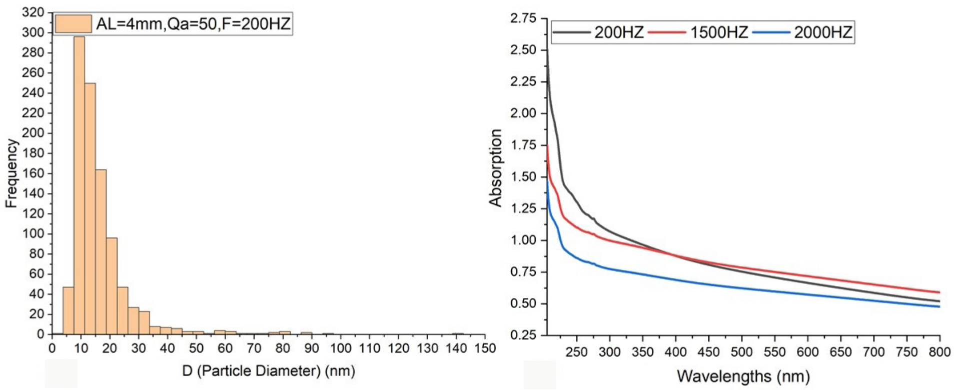

Fig. 3. Numerical distributions of nanoparticles by size at gas flow of 50 ml/min and laser frequency of 200 Hz (*a*), absorption of aluminum after laser sintering (*b*)

From the obtained TEM images, we collected statistics on the diameters of the modified spherical NPs, processing an average of 500 pieces, and rounding them as spheres. Most of the particles are characterized by sizes in the range from 5 to 50 nm. For samples interacting with the ultraviolet laser, the range increases from 5 to about 100 nm with some single particles with sizes of up to 140 nm.

According to spectrophotometry data, it has been proven that the absorption of aluminum after laser sintering is characterized by pronounced plasmonic absorption peaks in the ultraviolet region at position 218 nm.

Conclusion

We demonstrated that using the gas discharge method Al nanoparticles with plasmon resonance in the ultraviolet region can be produced. The results correspond with studies using thermal modification in which Al nanoparticles have weakly pronounced plasmonic peaks at 275 nm. The appearance of the second peak is caused by presence of large particles. Due to the spectral dependences of the absorption regions of NPs with an increase in the NP diameter, the maximum shifts to the long-wavelength region of the spectrum and expands.

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THE AUTHORS

NOURALDEEN Messan
messannouraldeen@phystech.edu
ORCID: 0000-0001-5437-0021

VERSHININA Olesya V.
seraia.ov@phystech.edu
ORCID: 0000-0001-6945-4818

SHESTAKOV Nikita S.
shestakov.nr@phystech.edu
ORCID: 0009-0001-6926-5205

IVANOV Victor V.
ivanov.vv@mipt.ru

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