Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics

Electronic Supplementary Information

Pulsed laser ablation of a continuously-fed wire in liquid flow for high-yield production of silver nanoparticles

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1.Energy Normalization

1a. Theoretical Model

Normalization on energy of ablation efficiency values has been carried out considering the approximation of the laser beam energy profile to a two dimensional Gaussian distribution. Since the laser beam presents a diameter of 3 mm, then energy profile can be described by the equation 1.1

$$f(x,y) = \frac{1}{0.4\pi} e^{-\frac{1}{2}\left(\frac{x^2}{0.2}+\frac{y^2}{0.2}\right)} \qquad \text{eq. 1.1}$$

The integral of this equation on the boundaries of a circumference with a radius of 1.5 mm will give the total amount of energy coming out from the laser. The calculated integral, in polar coordinate system (ϑ and r), is reported on eq. 1.2

$$\int_0^{2\pi}\int_0^{1.5} \frac{1}{0.4\pi} r e^{-\frac{1}{2}\left(\frac{\cos\vartheta^2+\sin\vartheta^2}{0.2}\right)} d\vartheta dr = 0.996393 \qquad \text{eq. 1.2}$$

In the case of wire targets, integral boundaries should be fixed considering the diameter of the wire itself. In this way, wires with higher diameters will absorb a higher amount of energy respect to smaller wires. Calculated integrals, which represent the value of absorbed energies, are reported here for wires with diameter ranging from 125 μm (eq. 1.3) to 1500 μm (eq. 1.9)

$$\int_{-0.0625}^{0.0625}\int_{-1.5}^{1.5} \frac{1}{0.4\pi} e^{-\frac{1}{2}\left(\frac{x^2+y^2}{0.2}\right)} dxdy = 0.111057 \qquad \text{eq.1.3}$$

$$\int_{-0.125}^{0.125}\int_{-1.5}^{1.5} \frac{1}{0.4\pi} e^{-\frac{1}{2}\left(\frac{x^2+y^2}{0.2}\right)} dxdy = 0.21997 \qquad \text{eq.1.4}$$

$$\int_{-0.175}^{0.175}\int_{-1.5}^{1.5} \frac{1}{0.4\pi} e^{-\frac{1}{2}\left(\frac{x^2+y^2}{0.2}\right)} dxdy = 0.304191 \qquad \text{eq.1.5}$$

$$\int_{-0.25}^{0.25}\int_{-1.5}^{1.5} \frac{1}{0.4\pi} e^{-\frac{1}{2}\left(\frac{x^2+y^2}{0.2}\right)} dxdy = 0.423512 \qquad \text{eq.1.6}$$

$$\int_{-0.375}^{0.375}\int_{-1.5}^{1.5} \frac{1}{0.4\pi} e^{-\frac{1}{2}\left(\frac{x^2+y^2}{0.2}\right)} dxdy = 0.597788 \qquad \text{eq.1.7}$$

$$\int_{-0.5}^{0.5}\int_{-1.5}^{1.5}\frac{1}{0.4\pi}e^{-\frac{1}{2}\left(\frac{x^2+y^2}{0.2}\right)}dxdy = 0.735861 \quad \text{eq. 1.8}$$

$$\int_{-0.75}^{0.75}\int_{-1.5}^{1.5}\frac{1}{0.4\pi}e^{-\frac{1}{2}\left(\frac{x^2+y^2}{0.2}\right)}dxdy = 0.905746 \quad \text{eq. 1.9}$$

The ratio between values calculated for the wire and the one on the whole surface represents the ratio between energy coming inside the chamber and the energy absorbed by the wire.

1b. Experimental Comparison

Since the laser used for our experiment presents a M^2 factor of 1.8, which means that there is a not negligible divergence of the laser beam energy profile from the Gaussian shape, it seemed appropriate to compare calculations with experimental energy values in order to determine the accuracy of the model.

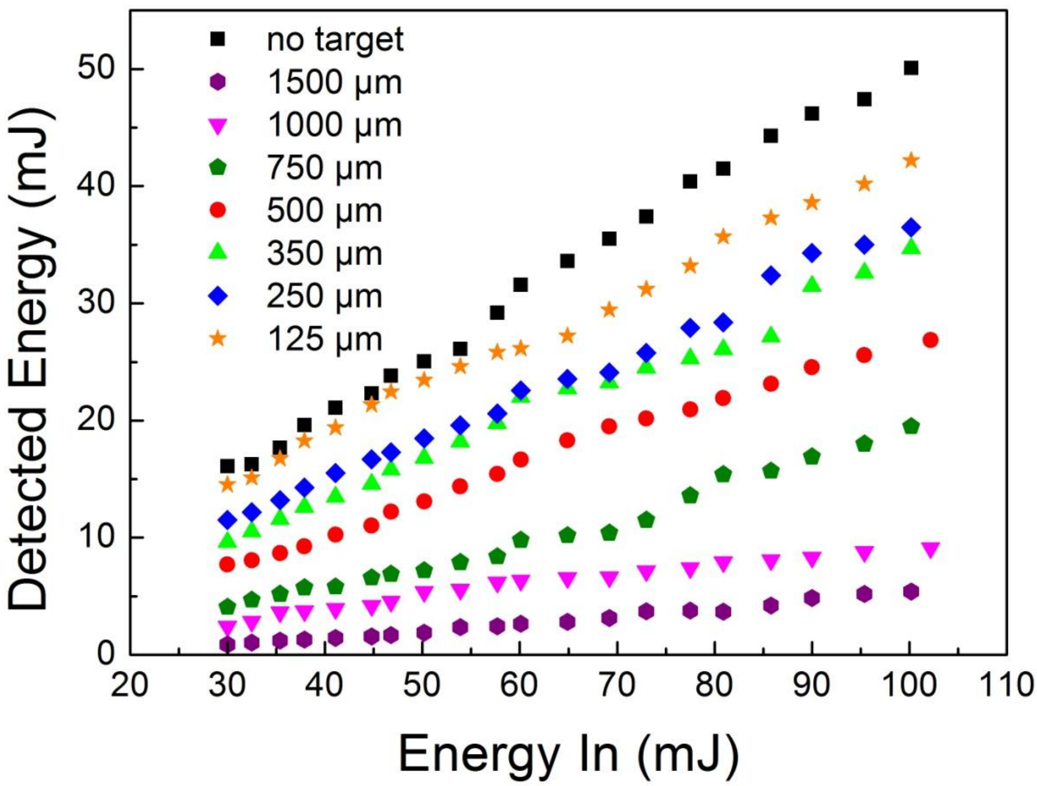

Fig. S1: Energy detected from the back window of the chamber when no target is positioned inside it (black squares) and when ablation of wire happens (colored markers).

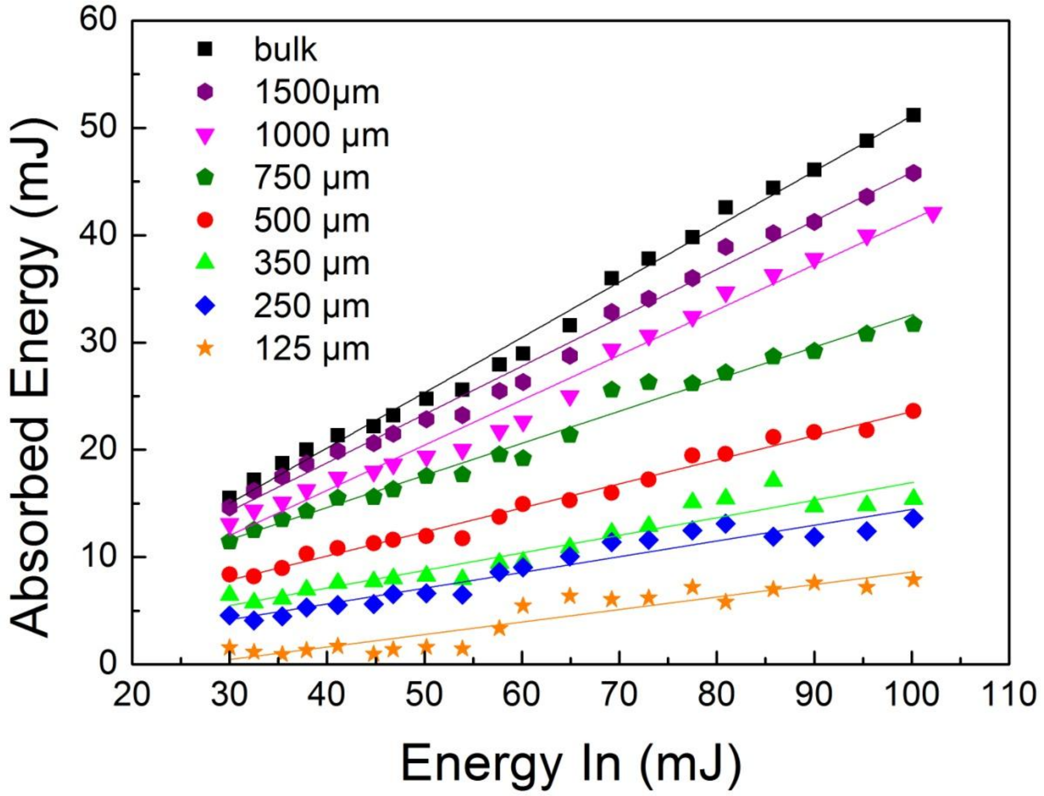

Fig. S2: Values of energy absorbed by wires of different shapes obtained by the subtraction of Detected Energy values for different wire diameters to "no target" one reported in fig. S1. The ratios between the slope of the curves obtained for the wires and the slope of the curve for the bulk have been compared to calculated values and reported in fig. 3 of the manuscript.

3. Variation of size distribution on fluence

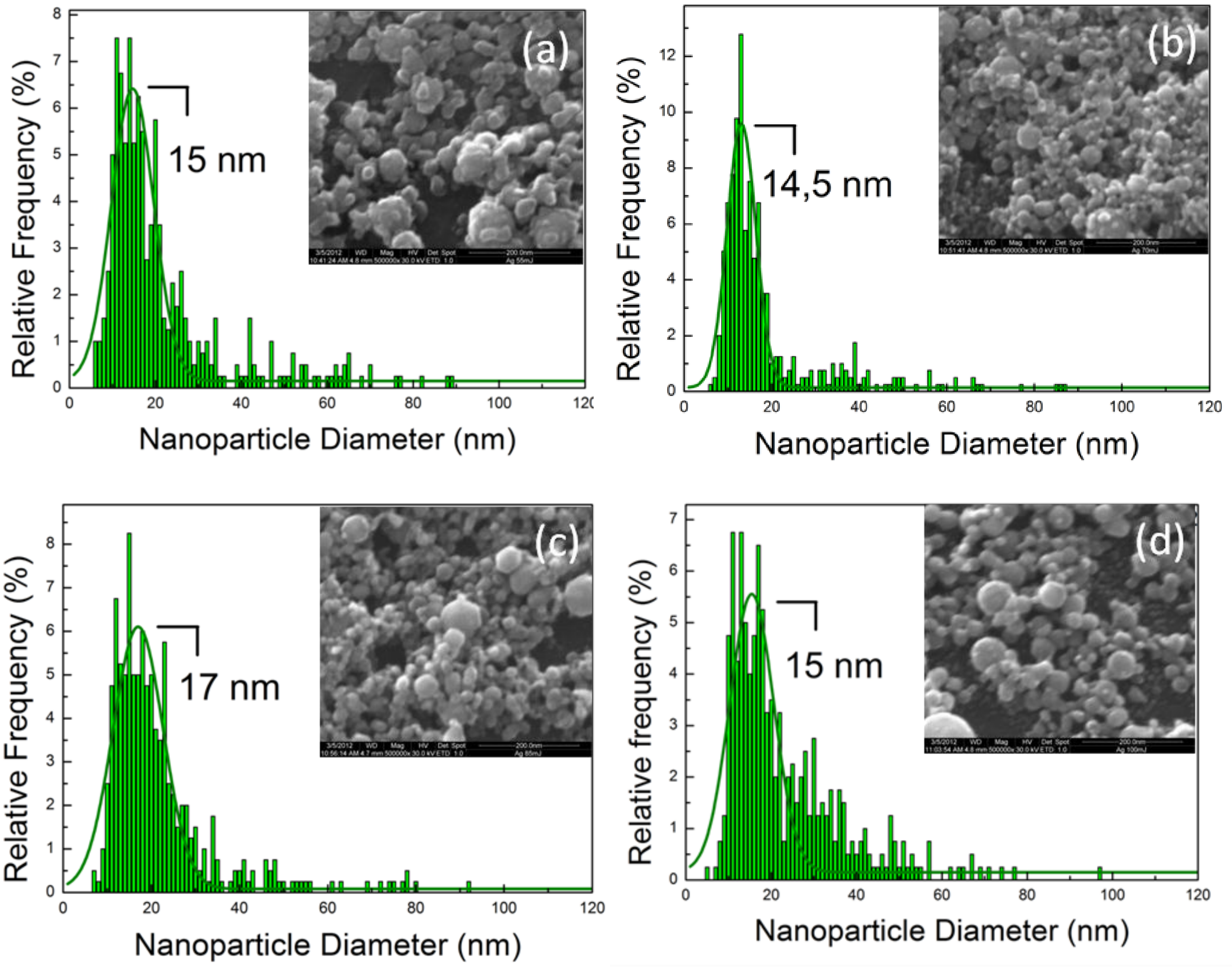

Fig. S4: Size distributions of silver nanoparticles prepared by laser ablation of a wire with a diameter of 250 µm at a repetition rate of 100 Hz and a fluence of (a) 0.8 J/cm^2, (b) 1.0 J/cm^2, (c) 1.2 J/cm^2 and (d) 1.5 J/cm^2. It is visible how, in the used range, size does not depend on fluence in a crucial way.