# Comment on "Effects of the medium on synthesis of nanopowders by wire explosion process" [Appl. Phys. Lett. 91, 141501 (2007)]

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In a recent letter, Cho *et al.*[1] reported an attractive process of wire explosion to synthesize silver nanoparticles. The results showed that uniform silver nanopowders were synthesized in water, while clustered and much larger-sized products were found in air. The authors ascribed the reasons to "higher energy deposition in the wire, sufficient expansion volume, and quick cooling of the particles" in water. The higher energy deposition in the wire (being regarded as a main cause in Ref. 1) is questionable although they observed a longer duration for plasma preformation in water. In this comment, we would like to address this questionable conclusion and provide a further understanding of the discharge process.

In Ref. 1, Cho *et al.* subliminally assumed that the discharge processes in water and in air were similar. They are actually quite different, although the waveforms of currents in water and in air are analogical. Based on the waveforms of current and voltage across the capacitor bank (Fig. 1: a copy of Fig. 2(a) from Ref. 1), we give a detailed consideration of the discharge process as follows. The waveforms of current and voltage across the capacitor bank are similar for both media so that Fig. 1 can represent both conditions. The letters are inserted to indicate different stages of the discharge.

For the discharge in air, (A–B) the electric field applied on the wire is pretty small at first due to the suspending wire so that the circuit is nearly broken. With increasing time, the electric field applied on the wire increases and electrons are emitted at certain electric field value. These emitted electrons will ionize the air. This period requires some time to make a considerable ionization to conduct the circuit; as a result, one can find that the voltage across the capacitor bank varied slowly initially (no obvious change during initial 10 $\mu$s). In this process, the energy dissipation is small and mostly consumed in the electron emission.

(B–C) From point B, the ionization is significant, the circuit is well closed and the wire heats up because there is a considerable current in the wire. The current increases with continued ionization and the wire continues to heat up. The wire will melt at one point between B and C. In this process, the energy is consumed in the electron emission and in the heating.

(C–D) At point C, the wire begins to vaporize and the resistance in circuit increases,[1,2] which results in a decrease in the current. Again, the energy is consumed in the electron emission and in the heating. Because of the high resistance, the heating could account for most of the energy deposition in this period.

After point D, the plasma is produced on the wire[1,2] and the inductance of the circuit becomes stable so that the $V$ and $I$ can be expressed by damping oscillatory functions.

For discharge in water, (A–B) the discharge is similar to that in air due to a nearly broken circuit. However, there is electrolysis of water taking place gradually in the wire by which hydrogen[3] or oxygen gas are created and absorbed near the wire. The electrolysis will be accompanied by the ionization of the gas (mixture of $H_2$ and $O_2$) when the electric field applied on the wire reaches certain value. The resistance of the circuit will decrease with the electrolysis and ionization, but it requires certain time to reach a critical point to conduct the circuit. Thereby, one can find that a very small current lasts 10 $\mu$s. In this process, the energy is consumed in the ionization and the electrolysis, but it is negligible due to the nearly broken circuit.

(B–C) Enough ionization in the gas sheath around the wire causes a further decrease of the circuit resistance, then the current increases and the wire is heated by Joule heat. Meantime, there must be a heat exchange between the water and the heated wire since the wire is surrounded by the water. In this process, the wire will heat up to melt at one point between B and C similar to that in air, the energy is con-

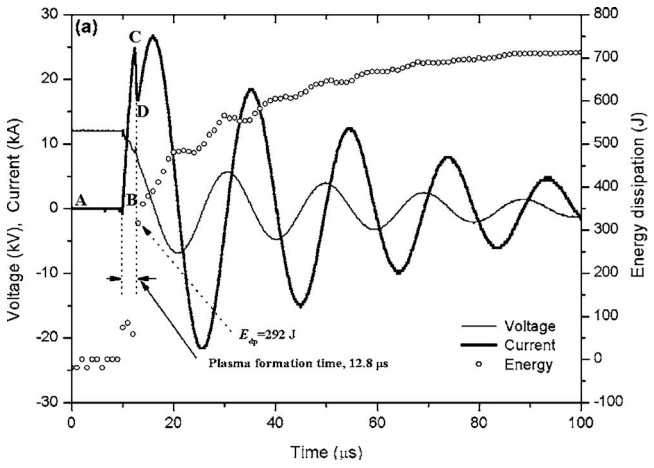


FIG. 1. Copy of Fig. 2(a) from Ref. 1. Letters are inserted to indicate the stages of the discharge.

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sumed in the ionization, the heating of the wire and the heat exchange.

(C–D) At point C, the wire begins to vaporize,[1,2] resulting in an increase of the resistance, so that the current decreases. In this period, the energy is consumed in the electron emission, the heating and the heat exchange with water. The heating will account for most of the energy deposition because of the high resistance.

After point D, the process is similar to that in air but still there is a heat exchange between the water and the discharging wire.

From the description above, one can get the energy depositions in air and in water are in different ways, therefore, one cannot compare each other. Compared to the energy consumption in air, there are additional heat exchange and electrolysis in water. Therefore, it is impossible on the basis of current and voltage measurement data to claim unambiguously how many percents of energy is dissipated on the wire heating. The higher energy deposition in the wire in water proposed by Cho *et al.* is questionable and we suggest that it might originate from the heat exchange and the electrolysis of water. Compared with the discharge in air, the duration of plasma preformation (BCD) is longer because the energy is not all dissipated in the wire heating.

In BCD range, the difference of the dissipated energy in water and in air before plasma formation is 121 J based on the data of Ref. 1. This additional energy can heat 0.413 g water 30 °C (as room temperature) to 100 °C. 0.413 g is equal to the mass of a water cylinder 1.669 mm in thickness, 3.638 mm in outsider diameter, 0.3 mm in insider diameter, and 40 mm in length estimated from Eq. (5) around the 40 mm wire. If one considers that water is heated to vapor, the water cylinder will be 0.0190 mm in thickness (0.338 mm in outsider diameter, 0.3 mm in insider diameter, and 40 mm in length) estimated from Eq. (6). One can find that these thicknesses are in a reasonable range.

Because the wire is not vaporized before point C, we propose that the energy dissipated in the electrolysis and the heat exchange ($E_a$) in BC range can be written in a more accurate form as

$$E_a = \frac{1}{2}CV_0^2 - \left[\frac{1}{2}Cv(t)^2 + \frac{1}{2}Li(t)^2\right] - \int_B^E i(t)^2 R dt, \qquad (1)$$

where $C$, $V_0$, $L$, $i(t)$, and $R$ are capacitance, initial charging voltage, instant voltage on the capacitor, inductance of the discharge circuit, instant current, and the resistance of the wire, respectively.

In order to further understand the discharge, the discharge process can be considered as a circuit with the elements consisting of a capacitor bank ($C$), an inductance ($L$), and a wire resistor ($R$) in series. If the voltage and the current of the capacitor bank are $V$ and $I$, they can be expressed as

$$V + L\frac{dI}{dt} + IR = 0,$$

$$LC\frac{d^2V}{dt^2} + CR\frac{dV}{dt} + V = 0,$$

$$I = C\frac{dV}{dt}. \qquad (2)$$

Given the initial conditions are

$$V = V_0(t=0),$$

$$I = 0(t=0). \qquad (3)$$

The solution of Eq. (2) is shown below (considering the damping solutions),

$$V = \frac{V_0}{2}\exp\left(-\frac{R}{2L}t\right)\cos\left(\frac{\sqrt{4LC - C^2R^2}}{2LC}t\right),$$

$$I = -\frac{CV_0}{2}\exp\left(-\frac{R}{2L}t\right)\left[\frac{R}{2L}\cos\left(\frac{\sqrt{4LC - C^2R^2}}{2LC}t\right) + \frac{\sqrt{4LC - C^2R^2}}{2LC}\sin\left(\frac{\sqrt{4LC - C^2R^2}}{2LC}t\right)\right]. \qquad (4)$$

From the expressions of $V$ and $I$, one can qualitatively understand the waveforms in Ref. 1. Also, one can find from Fig. 2 in Ref. 1 that the damping voltages for both media have the same periods (about 20 $\mu$s) which means that the $(4LC\text{-}C^2R^2)^{0.5}/2(LC)$ for both media are close after point D. However, the voltage in water have a faster damping speed than that in air, which implies that the value of $R/2L$ in water is larger than that in air.

In addition, the formula (1) in Ref. 1 for calculating the energy dissipated in the wire will not be applicable after point C since the nature ($L$, and $R$, etc.) of the vaporizing wire is much different from the solid wire.

In conclusion, although Cho *et al.* fabricated attractively uniform nanoparticles in water by silver wire explosion, the interpretation of high energy deposition is questionable. Based on a detailed analysis of the discharge processes, we conclude that the heat exchange and the electrolysis also account for some portion of the energy dissipation during the discharges. The validity of the formula for calculating the energy dissipation in the wire in Ref. 1 was also investigated. One alternative formula was proposed to estimate the energy dissipated more accurately before the wire starts to vaporize. The oscillatory solution of the voltage and current across the capacitor bank was given to further understand the characteristic of the discharges.

The specific heat of water is 4.187 J/g and the heat of vaporization of water is 2260 J/g. The difference of the dissipated energy in water and in air before plasma formation is 121 J based on the data of Ref. 1. The wire is 0.3 mm in diameter and 40 mm in length. $R$ and $d$ are the diameter and the thickness of the water cylinder around the wire, respectively. The density of water is 1 g/cm$^3$.

*i.* If the water cylinder around the wire is only heated from 30 to 100 °C, the thickness of heated water cylinder can be estimated as follows:

$$40 \times 10^{-1}\pi\left[\left(\frac{R}{2}\right)^2 - \left(\frac{0.3}{2}\right)^2\right] \times 10^{-2} \times (100 - 30) \times 4.187 = 121,$$

$$d = \frac{R - 0.3}{2} = 1.669 \text{ mm}. \qquad (5)$$

**ii**. If the water is heated from 30 °C to vapor, the thickness of heating water around the wire, $d$, can be estimated as follows:

$$40 \times 10^{-1}\pi\left[\left(\frac{R}{2}\right)^2 - \left(\frac{0.3}{2}\right)^2\right] \times 10^{-2} \times (100 - 30) \times (4.189 + 2260) = 121,$$

$$d = \frac{R - 0.3}{2} = 0.0190 \text{ mm}. \quad (6)$$

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