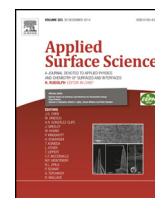




Contents lists available at ScienceDirect

Applied Surface Science

journal homepage: www.elsevier.com/locate/apsusc



Nanosecond (ns) laser transfer of silver nanoparticles from silver-exchanged soda-lime glass to transparent soda-lime glass and shock waves formation

Mohamed Chérif Sow^{a,*}, Jean-Philippe Blondeau^a, Nadine Sagot^a,
Nadège Ollier^b, Teddy Tite^b

^a Laboratoire CEMTHI UPR CNRS 3079, Université d'Orléans, IUT de Chartres, 28000 Chartres, France

^b Laboratoire Hubert Curien UMR CNRS 5516, Université de Lyon, Université Jean Monnet, 42000 Saint-Etienne, France

ARTICLE INFO

Article history:

Received 15 July 2014

Received in revised form

17 November 2014

Accepted 21 November 2014

Available online xxx

Keywords:

Nanosecond laser
Femtosecond laser
Soda-lime glass
Ion exchange
Nanoparticles
Shock waves

ABSTRACT

In this contribution, we showed for the first time in our knowledge a single-step process for silver clusters and nanoparticles growth and transfer from silver-exchanged soda-lime glass to un-exchanged soda-lime glass (transparent glass in visible and NIR domain) by nanosecond (ns) laser irradiation. The transferred silver nanoparticles in transparent glass are strongly linked to the glass surface. In addition, we point out the formation of shock waves, with selective silver clustering on the top wave. This technique provides an alternative and simple way to obtain metallic nanoparticles in different media which can be traversed by laser wavelength used. Moreover, this experiment is made at room temperature and air environment. It is worth noting that our technique requires a glass previously doped with the corresponding silver ions.

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1. Introduction

For the last decades, pulsed laser manipulation of glass has been widely studied for fundamental and applied research. Most of these studies relate to laser irradiation of glass embedded with metallic ions [1–3] or small metallic [4,5] nanoparticles (called metal–glass nanocomposite) in order to grow metallic nanoparticles with controlled size, shape and volumic fraction. In fact, metallic nanoparticles are expected to have several potential applications such as functional optical devices [6,7] or sensing [8,9]. Most applications of metal–glass nanocomposite are due to the optical properties of the surface plasmon resonance (SPRs) [6]. The spectral position and shape of the SPRs depend on the composition of the host material but also be tuned by the size, shape or volumic fraction of the nanoparticles [10,11].

Silver ions are often generated inside glass by ion exchange technique which is a widely applied technology to introduce metallic ions in glass [12]. This technique has been used for

centuries to modify the optical absorption properties of glass for purposes of decoration and coloring. After silver ions introduction in glass, several experimental techniques could be adopted to obtain nanoparticles in glass. Few results have been obtained in the case of direct pulsed laser irradiation of silver exchanged soda-lime glasses; currently, these nanoparticles are generally formed by thermal annealing [13]. This thermal annealing promotes the formation of spherical nanoparticles with diameters up to few tens of nanometers. Pulsed lasers are often used to change the size, shape and volumic fraction of the initially spherical silver nanoparticles [14]. For example, the formation of non-spherical silver nanoparticles has been shown and used to fabricate micro-polarizers, based on their dichroism [15]. Laser-induced cleaning by dissolution of the silver nanoparticles previously formed in glass matrix has also been observed in the case of prolonged pulsed laser exposure [13].

Femtosecond (fs) and nanosecond (ns) lasers are most widely used in the case of laser manipulation of doped glasses either by silver ions or by small silver nanoparticles [1–5]. Fs lasers are most used than the ns lasers. In fact, fs laser due to their very low pulse can react with molecular structure (breaking link for example) unnecessarily heating the glass, thus with little

* Corresponding author.

E-mail address: mohamed-cherif.sow@univ-orleans.fr (M.C. Sow).

damage. ns laser induces damage on surfaces of materials via heating and ablation processes, that is why ns laser are rarely used for nanoparticles growth. Comparison of surface modification by ns and fs laser irradiation has been studied by Rethfeld et al. [16].

In order to promote silver nanoparticles in glass by ns laser with small damage, we imagined the silver nanoparticles transfer from exchanged glass to transparent glass. In fact, knowing that ns laser led to simultaneous silver nanoparticles formation and ejection from silver exchanged glass, we expected that these silver ions or nanoparticles could diffuse in a transparent glass glued together with exchanged glass during ns laser irradiation.

Laser-induced nanoparticles transfer has been already reported in the literature.

For example, Kusnetsov et al. showed femtosecond laser-induced transfer of nanostructure of gold from donor (glass) onto a receiver substrate [17]. Previous to their transfer, the nanoparticles of gold were fabricated by nanosphere lithography on one side of a transparent glass. For the transferring process, the laser pass through the transparent donor substrate which is placed upside down with the receiver substrate directly attached underneath. The transferred particles have spherical shape (40–200 nm in size) and they are arranged in a hexagonal array (for the detail of the method, see [17]). It is worth noting that this method requires a good contact between the donor and the receiver substrates, which is realized using an elastic polymer polydimethylsiloxane (PDMS) substrate as a receiver. Nanoparticles transfer is assigned to photothermal effects. Both interparticle distance and particle size can be independently controlled by this method. It also leads to the formation of partially (about 70%) embedded nanoparticles, which make them very stable and resistant against cleaning and mechanical treatments.

Nanosecond laser-induced metallic (gold) nanoparticle transfer has been very recently shown by Hanmo Gong et al. [18]. This method is based on laser-induced forward transfer. The light beam is normally irradiated on the front side of the absorber sample (placed upside down) through the receiver. A glass coated with an ITO film is used as the receiver substrate. The transferring process is simply conducted in air and requires relatively low light fluence (35 mJ/cm²) compared to existing fabrication methods. The authors explain the nanoparticles transfer by the photothermal effect induced by the laser irradiation. In fact, a metamaterial is used as the donor in order to utilize its height absorption to enhance the photothermal effect. This leads to high temperature rise above the melting point of gold nano-discs utilized as a donor. Therefore, the gold nano-discs in the irradiated area are melted and then coalesce into sphere nano-droplets. Because of the inertia and force of gravity, the droplets are detached out of the absorber sample and fly onto the receiver substrate. The transferred nano-droplets cool down and solidify again into spherical dome shape.

Even if these two methods are cheap and simple, the fabrication of donor and/or receiver substrate is a “tricky” task. Furthermore, the donor is in micro-or-nanostructure form. In this contribution, we reported the growth of silver nanoparticles in silver-exchanged soda-lime glass by ns laser irradiation and their transfer on transparent glass glued together. Thus, in our study, the donor and the receiver are simply made only by silver (ions)-doped glass and undoped glass substrates, respectively. We also found nano- and microstructures arranged like rings on contiguous faces, which are attributed to shock wave generation. We demonstrated that these nano- and microstructures located in the top wave are made of silver. Shock wave formation in glass through pulse laser irradiation has been reported and explained by Kasaai et al. [19]. We tried to explain the mechanism of nanoparticle transfer in our case.

2. Fabrication process

Let first consider the different experimental techniques used in this work. Commercial soda-lime glass substrates of composition in wt%: 71.67 SiO₂, 14.93 Na₂O, 5.40 CaO, 4.28 K₂O, 3.72 BaO with amounts of Fe₂O₃ in trace were used as substrate. Glasses were delivered already polished and cut into 76 mm × 25 mm × 1 mm. The experimental technique for nanoparticles growth and transfer used in this study is a two-step process.

First, a widely used thermal ion exchange process consisting of immersing 1-mm-thick commercial soda-lime glass slides into a mixture of molten salt bath of AgNO₃ in NaNO₃ in suitable proportions, or only in molten salt bath of AgNO₃ was used to introduce silver ions in glass matrix. In this study, ion exchange was performed only in a molten silver salt bath of 99% of molecular concentration. The duration was varied from 1 min to 2 h and the temperature was fixed to 400 °C. It is worth noting that the interesting results in our case were obtained from 10 min to 1 h ion exchange duration. After ion exchange, the sodium ions from the surface and the subsurface regions of the glass are substituted by the silver ions. This leads to introducing silver ions molar concentration of about 12% at the glass surface by replacing sodium ions. The concentration of silver ion decreases in the depth with an almost error function profile according to Fick's law. The penetration depth of the exchanged zone can reach several micrometers depending on the exchange duration. In our case, the maximum depth is estimated at around 10 μm for 1 h exchange duration. The sum of the concentration of silver and sodium ions across the exchanged layer becomes constant, which proves that one silver ion is exchanged by one sodium ion.

The second step consists of irradiating two attached glasses (transparent and exchanged glass) by ns laser with appropriate parameters in order to grow the nanoparticles in exchanged glass within the layer containing silver ions and silver transfer in transparent glass. Before irradiation, silver-exchanged soda-lime glass and transparent soda-lime glass of 1 mm in thickness was first washed in an ultrasonic ethanol bath to remove any organic contamination.

The laser used is a collimated monochromatic light source, Brilliant Eazy Nd:YAG ns-laser type of Quantel. The laser emission may be obtained in ultraviolet (355 nm), visible (532 nm) or infrared (1064 nm). Only the results obtained with λ = 355 nm (ultraviolet) are presented in this paper. The pulse duration is 4 ns. The laser pulse energy for laser emitting at 355 nm is 60 mJ. Depending on the repetition rate (frequency) at 10 Hz and the energy per pulse, the average power is 0.6 W at 355 nm. In this study, we have only varied the shots number (thus also the duration) and irradiated the glasses in two conditions, which are obtained, with or without focusing lens. According to the frequency and the number of shots chosen, the irradiation time was varied from 10 s to 1 h in the case of un-focused laser and up to a few hundred seconds for focused laser. Note that the glass samples are not moved during the irradiation time.

The experimental set-up (system design) schematically shown in Fig. 1 is fairly simple. The two attached glasses were screwed and subjected to ns laser irradiation. The direction of the incident laser beam was perpendicular to the surface of the glass samples and the laser beam passes through transparent glass.

We inscribed three series with the following conditions:

- the first one, namely S₁, was inscribed in an exchanged glass without the transparent glass in the front;
- the second one, S₂, is inscribed in the two attached glass (exchanged and transparent), through the transparent glass. For the series S₁ and S₂, Gaussian laser beam had a diameter of 5 mm

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