

# Uniform Spray-based Large Area Nanoparticle Coating at Nanometric Thickness

M. Rukosuyev\*, P.C. Lee\*\* and M.B.G. Jun\*\*\*

\*University of Victoria, Victoria, BC, Canada, [maxr@uvic.ca](mailto:maxr@uvic.ca)

\*\*University of Vermont, Burlington, VT, USA, [patrickc.lee@uvm.edu](mailto:patrickc.lee@uvm.edu)

\*\*\*Purdue University, West Lafayette, IN, USA, [mbgjun@purdue.edu](mailto:mbgjun@purdue.edu)

## ABSTRACT

Various types of coatings are widely used in both research and industrial applications. In spray coating, droplet velocities, average size, and dimensional uniformity are some of the factors that are crucial for the final coating parameters. Therefore, the process and the equipment used during spray generation and coating will determine its quality and suitability for a particular application. This paper presents development of a decoupled system for the spray coating and micro printing, which include an ultrasonic spray generation device and a nozzle for the spray deposition independently operated. Design and development of the system as well as testing for different applications are also presented in this paper. The system design can be potentially used for large area coating, such as windows and solar panels, as well as micro printing of electronic circuits and numerous other applications.

**Keywords:** Spray coating, nanoparticle deposition, atomization, spray control

## 1 INTRODUCTION

The range of applications requiring various types of coatings stretches from biomedical devices to microelectronics, and the number of technological solutions provided by the use of advanced coating materials is still growing. Two characteristics of coatings that are of primary importance are coating layer uniformity and thickness control. The key factors that determine the quality of a spray-coated layer are the size (order of a few microns in diameter) and dimensional stability of droplets in the spray and the droplet impact velocity [1].

Quite often, to produce a functional coating, suspensions of various nanoparticles are used. In that case, atomised droplets will contain nanoparticles within, which will be left on the coated surface once the liquid solvent is evaporated. The need for small size of the atomized droplets arises from the fact that in order for the nanoparticle coating to be uniform across the surface, one would require the particles to be finely dispersed within an

atomised droplet. In case of larger droplets, nanoparticles inside the liquid tend to agglomerate. Therefore, once deposited, nanoparticle agglomerates are left on the surface thus increasing surfaces' roughness and decreasing the coating's uniformity. Consequently, the fewer nanoparticles are in a single droplet, the more uniformly they will be distributed over the coated surface and the better the coating thickness control will be.

Traditionally, conventional air pressure atomising nozzles are used for spray coating [3-5]. Although providing adequate performance in most spray coating applications, drawbacks associated with this method make it unsuitable when tight thickness and uniformity requirements are in place. Due to the nature of the atomisation mechanism in air atomising nozzles, one should have certain air pressure to ensure atomisation. That limits the range of spray velocities that can be controlled. In addition, droplet size distribution is non-uniform, strongly dependant on the pressure applied, and generally in the region of tens to hundreds of  $\mu\text{m}$ .

One way to produce droplets of consistently small size is to use ultrasonic atomization process [6]. Atomization of the coating solution/suspension with the aid of a vibrating piezoelectric crystal provides the spray droplets of desired size and can accommodate the use of wide range of materials. Several companies, such as Sono-Tek, Sonaer, and Optomec are successfully using ultrasonic atomization process to generate aerosols used for coatings and micro printing (in case of Optomec).

Device presented in this paper is an innovative system for the spray coating and micro printing which includes an ultrasonic particle generation device and a nozzle for the spray deposition. The deposition nozzle, due to the larger diameter of the outlet channel, allows for high throughput of droplets. At the same time, it will accelerate and focus spray droplets using high-speed air flow in the center of the nozzle exit. Therefore, the proposed system attempts to solve both, droplet size and proper droplet impact velocity

problems to achieve close to optimum coating conditions. The described design could be successfully used for large area coating, such as windows and solar panels, as well as micro printing of electronic circuits and numerous other applications.

## 2 DESIGN CONCEPT AND SYSTEM DEVELOPMENT

The core of the design consists of the atomized droplet generation and the deposition nozzle decoupled in their respective operation. The low velocity carrier gas takes the droplets from the droplet generation unit to the deposition nozzle. At the nozzle exit, the particles are accelerated and focused by the high speed gas flow from the tube in the center of the nozzle. Therefore, a dual velocity regime is realized by using the lower velocity carrier gas internally, while the droplets are accelerated at the nozzle exit just before the deposition occurs. To avoid condensation, facilitate droplet size uniformity, and ensure symmetry of the particle stream, an extra intermediate flow-conditioning unit (FCU) was added to the design. FCU also doubles as a mixing chamber when two or more materials are deposited simultaneously (Figure 1).

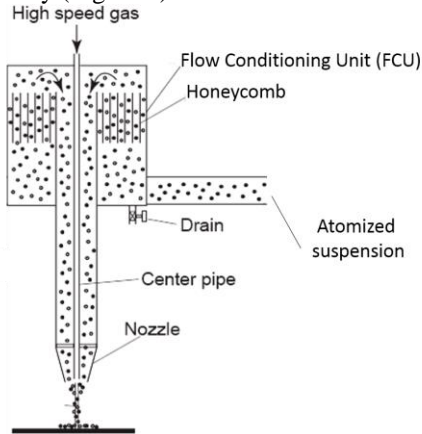


Figure 1. Nozzle with double atomizer schematics.

The FCU is placed in between the atomizer(s) and the nozzle and consists of a hollow cylinder with honeycomb structure imbedded in it. Asymmetric entrance ensures a vortex forming in the lower section to sort the particles. Due to a ‘cyclone’ effect, larger particles will collide with the wall of the chamber, condense, and will be drained out of the FCU. Smaller and lighter droplets will travel through the honeycomb and further down to the nozzle exit. The carrier gas stream is also made less turbulent while passing through the honeycomb structure. Furthermore, FCU allows the addition of the long high-speed central gas tube, which creates axisymmetric exit channel thus reducing turbulent disturbances within the nozzle and at the nozzle exit.

Figure 2 clearly shows that most of the atomized droplets are tightly focused into a high-speed jet. Consequently, the accelerated particles are deposited onto the substrate in such a way as to maintain close to optimum coating conditions.

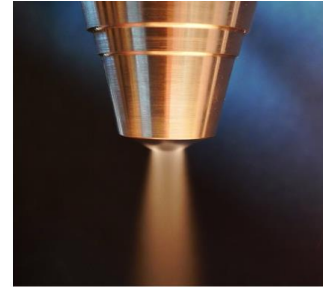


Figure 2. Exit jet.

The combination of appropriate nozzle exit distance and feed rate produced a coating layer where the solvent was evaporated within 1-2 seconds after deposition. This rapid evaporation ensured that smaller droplets would not coalesce, thus providing the opportunity for the nanoparticles to settle on the surface before they could agglomerate into large clusters.

To study the overall structure of the jet forming at the nozzle exit, a series of images using high speed recording equipment and PIV technique were analyzed. The near-field structure of a coaxial jet is considerably complex. The mixing between the jet streams is critically controlled by the dynamics and interactions of the vortical structures in shear-layers developed between the two jets, and also between the outer jet and the ambient fluid. The images shown in the Figure 3 were collected using particle image velocimetry technique (PIV).

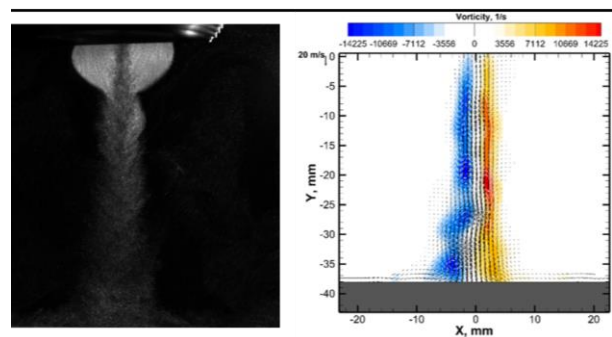


Figure 3. Flow patterns of the coaxial jet with the mean center circular jet velocity,  $U_i = 35$  m/s.

The introduction of the center jet led to the formation of the proper coaxial jet and changed the dynamics of the flow significantly. The presence of the inner jet led to a substantial decrease of the all three regions of the impinging jet and allowed to focus the coaxial jet on a

much smaller area, compared to that of the “no center jet” case.

### 3 UNIFORM COATING PERFORMANCE TESTING

In order to test the uniform coating distribution performance of the nozzle, a number of coating experiments were conducted. These experiments indicated that the nozzle design allows for the particles to be uniformly deposited onto a variety of substrates with different geometries.

Polymer coated silica nanoparticles were coated onto glass substrate with subsequent sintering at 500°C for 4 hours. Nanoparticles used in this experiment were Vive Nano Silica (+) (by Vive Nano, Canada) polymer encapsulated silica nanoparticles. Results shown in Figure 4 indicate a uniform thickness with very slight variation in the range of about 10 nm. The image was taken using Hitachi S-4800 field emission scanning electron microscope and QUARTZ PCI (version 8) software. The polymer used to encapsulate the particles is evaporated during sintering process. The remaining nanoparticles form a coherent layer sufficiently covering the substrate with no observable gaps in the coating.

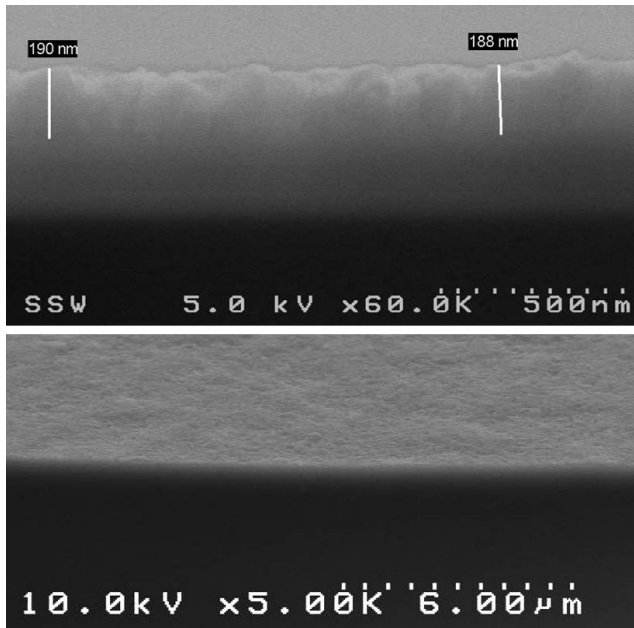


Figure 4. The edge of silica coated glass slide.

Pure silver nanoparticles were also deposited onto glass substrate. To highlight the distribution of the nanoparticles on the substrate, an EDX analysis has been performed (Hitachi S-4800 SEM). The resultant spectrum and the accompanying mapping image are shown in Figure 5. EDX results clearly show that silver nanoparticles are evenly

dispersed on the glass substrate surface. The particles were synthesized in house and average around 20-30 nm in diameter. Modified polyol process was used to synthesize the nanoparticles [7-8].

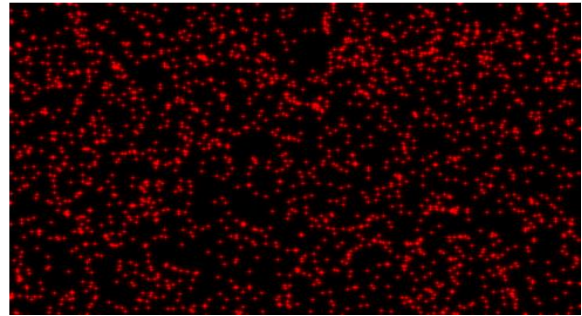


Figure 5. EDX mapping of silver particles.

Figure 6 shows the nanoparticles evenly distributed over the surface of a coated glass slide. The coating is a single layer, but can be made as thick as needed for a particular application by increasing the number of coating passes.

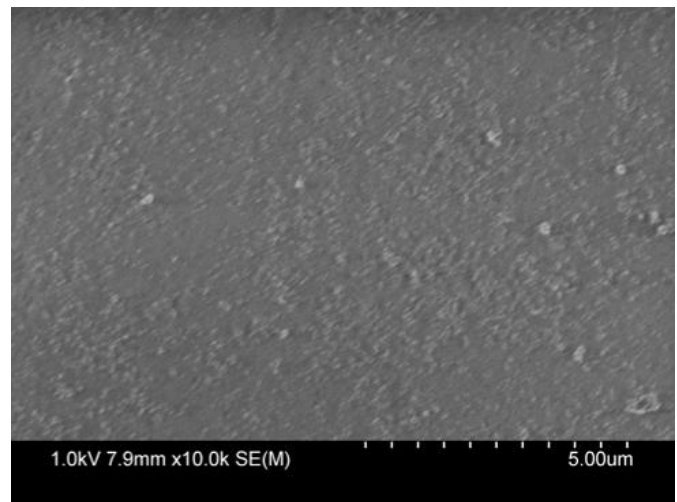


Figure 6. Silver nanoparticles on glass.

Some agglomeration and contamination can also be seen in Figure 6. However, the majority of the particles are spread evenly on the substrate surface which confirms nozzle’s ability to produce thin layer coating.

### 4 CONCLUSION

In conclusion, the coating system presented in this manuscript has been shown to be capable of producing uniform coating at sub-micron thickness. Preliminary coating results indicate that the minimum coating thickness of a nanoparticle layer is largely determined by the nanoparticle size as the system ensures even distribution of particles across the coated surface. The core principle of the system’s operation is the decoupling of the droplet

generation device and the nozzle that facilitates close to optimum coating conditions. Dual regime nozzle ensures the particles travel through the nozzle at relatively low velocity to prevent particle adhesion to the interior walls. At the same time, at the exit of the nozzle the particles are accelerated to enter the “spreading” regime. The unique tunability and adaptability of the system configuration allows nozzle design changes suitable for a number of different coating applications in both research and industrial environment.

## REFERENCES

- [1] C. Mundo, M. Sommerfeld, C. Tropea, Droplet-Wall Collisions - Experimental Studies of the Deformation and Breakup Process, *International Journal of Multiphase Flow*, 21 (1995) 151-173.
- [3] H.B. Scher, D.K. Giles, J.W. Tringe, H.W. Levie, Aerosol coating process based on volatile, non-flammable solvents, Google Patents, 2015.
- [4] I.J. Kramer, J.C. Minor, G. Moreno-Bautista, L. Rollny, P. Kanjanaboos, D. Kopilovic, S.M. Thon, G.H. Carey, K.W. Chou, D. Zhitomirsky, A. Amassian, E.H. Sargent, Efficient Spray-Coated Colloidal Quantum Dot Solar Cells, *Advanced Materials*, 27 (2015) 116-121.
- [5] D.Y. Byun, V.D. Nguyen, B.H. Seong, Coating System Using Spray Nozzle, Google Patents, 2015.
- [6] J.C. Simon, O.A. Sapozhnikov, V.A. Khokhlova, L.A. Crum, M.R. Bailey, Ultrasonic atomization of liquids in drop-chain acoustic fountains, *Journal of Fluid Mechanics*, 766 (2015).
- [7] Y. Sun, Y. Xia, Shape-Controlled Synthesis of Gold and Silver Nanoparticles, *Science*, 298 (2002) 2176-2179.
- [8] I. Donati, A. Travan, C. Pelillo, T. Scarpa, A. Coslovi, A. Bonifacio, V. Sergio, S. Paoletti, Polyol Synthesis of Silver Nanoparticles: Mechanism of Reduction by Alditol Bearing Polysaccharides, *Biomacromolecules*, 10 (2009) 210-213.