RESEARCH ARTICLE OPEN ACCESS

Growth and Dispersion the Silica Particle on the Glass via Modified Stöber Method and Spray Technique

Van Thi Thanh Ho*
[1]*Hochiminh City University of Natural Resources and Environment (HCMUNRE), Vietnam*

ABSTRACT
We presented the fabrication of a novel geometric light trapping structure based on silica particle. This light trapping structure with good morphology was fabricated through modified Stöber method and spray technique. More importantly, the silica particles were dispersed onto glass by using bottom-up or chemical approach involves a common route used to synthesis silica particles from atomic or molecular scale. An optimized synthesis condition is a combination of optimal values of each reaction parameter of Stöber method that could produce homogenous and mono-dispersed silica nanoparticles with uniform size. The diameter of silica particles can be varied from 200, 430, 560, 460 nm by increasing concentrations of ammonia from 2.33 mM to 4.65 mM, 9.31 mM and 13.96 mM, respectively. We found that the silica particle size was well controlled and uniform from ~200 nm to 560 nm by controlling the ammonia concentration. These results open up the possibility to further fabricate geometric light trapping structure with high scattering level (haze) and subsequently to increase the short circuit current density (Jsc) in the solar cells.
***Keywords*:** SiO_2, Stöber method, dispersion, growth, TCO

I. INTRODUCTION

Colloidal silica spheres occupy a prominent position in scientific and technological research, because of their unique optical, biological significances, as well as their important industrial applications such as catalysis, pigments, pharmacy and thin film substrates [1-4]. The specific applications of SiO_2 nanoparticles are highly dependent on their size and size distribution. In 1968, Stöber, et al. [5] reported a method for the synthesis of spherical silica nanoparticles from aqueous alcohol solutions of silicon alkoxides in the presence of ammonium hydroxide as a catalyst, with the size of the particles depending on the type of silicon alkoxide and alcohol. They reported that the smallest particles were those prepared in methanol solutions, and that the particle size increases with the chain length of the alcohol, from 50 nm to 1 µm. Various works have being published confirming that the choice of precursor and reaction conditions govern the final form of the silica spheres [6-8].

Transparent conductive oxides (TCOs) with rough textured surface play a critical role in the silicon based thin film solar cells. As a top electrode for thin film silicon solar cells, textured surface is required to scatter the incident light for effective light trapping, and subsequently to increase the short circuit current density (Jsc) in the solar cells. Several techniques are used for obtaining an optimized surface texture such as chemical etching and natural growth using chemical vapor deposition (CVD). The first approach of chemical etching is to make the film surface bumpy by using chemicals such as crater-like (ZnO:Al film after etching in acids). This approach can make the surface structures of ZnO:Al films for effective light scattering, however, the uniformly textured surface is hard to be obtained by this way and also complicated treatment is needed to obtain textured topography, like soaking in an acid solution, lithography, etc. which increases the fabrication cost and increase the risk of introducing unexpected impurities. The second approach of natural growth is to make the grains at the film surface present pyramidal by atmosphere pressure CVD of F doped SnO_2, and low pressure CVD of B doped ZnO by controlling the growth process. [9-14] This method is highly-efficient and can avoid the poor uniformity in the corrosion method. Recently, novel types of high haze SnO_2:F TCO superstrates with double-texture (W-texture) surface morphology have been also developed. [15-19] These TCO superstrates obtained by natural growth are aiming to boost the scattering level (haze), especially at longer wavelengths (λ>600 nm), however, generally requires the vacuum and needs expensive metal organic salts. Therefore, the cost is high by this method. In order to reduce the cost and achieve large scale industrial production, we develop an effective way to fabricate a light-trapping TCO structure based on dispersed Silica particles onto glass for effective light trapping in thin film silicon solar cells. More importantly, the silica particles were dispersed onto glass by the bottom-up or chemical approach involves a common route used to synthesis silica particles from atomic or molecular scale. Some of the widely used methods to synthesize silica nanoparticles are sol-gel process, reverse microemulsion, and flame synthesis. The high temperature flame decomposition of metal-organic precursors can be produced silica nanoparticles but this process is difficult in controlling the particle size, morphology, and phase composition.

In this work, we present an effective and reliable method to growth sub-micrometer mono-disperse and multi-layer disperse silica on glass through Stöber method and spray technique as a chemical approach based on silica with uniform size. Significantly, the pure silica particles were produced through without using any surfactant and/or stabilizer.

II. EXPERIMENTAL SECTION

II.1. Materials and characterizations

Ethanol 99.5% from Shimakyu's pure chemicals, 1-butanol 99%, isopropanol 99% and tetraethoxysilane (TEOS), 98% was purchased from Acros. Ammonium hydroxide (35%) was purchased from Fisher Scientific, Spin-on-glass (SOG) 211 from Honeywell. All chemicals were used without further purification. The silica particles size were characterized using scanning electron microscopy (SEM, JEOL JSM-6500F).

II.2. Synthesis of silica sphere on glass via modified Stöber method and spray technique

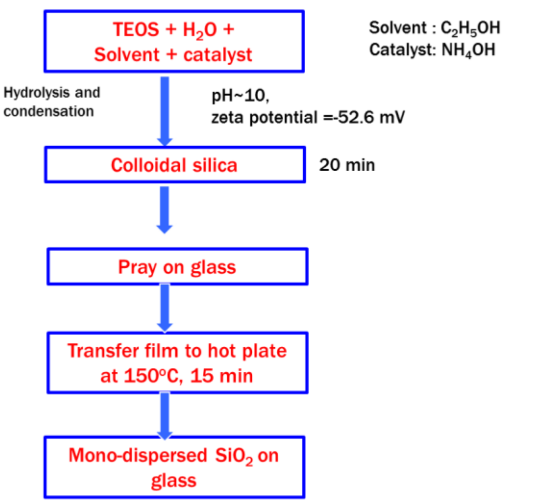

Figure1. The process for preparation SiO_2 via modified Stöber method and Spray technique

The textured surface based on silica for effective light trapping was fabricated in this work. First, the silica particles were synthesized through Stöber method as a chemical approach. For a typical synthesis procedure, 0.037 mmol was added to a solution containing 11 ml of ethanol and 5 ml of deionized water to form ammonia concentration of 2.3 mM , followed by the addition of 0.088 mmol of TEOS in 10 mL ethanol to the solution with vigorous stirring at room temperature. After 20 min reaction time, the resulting mixture was changed from transparent solution to turbid solution, suggesting silica colloidal is formed by hydrolysis and condensation of tetraethoxysilane in the presence of NH_3 play as a catalyst and in water-alcohol as a solvent. Next, the silica colloidal was dispersed onto glass substrate through spray technique. Corning EAGLE 2000 glasses was served as a substrate, had been cleaned through sonication in acetone for 5 min, cleaning in ethanol for 5 min, rinsing with DI water, and then blowing dry under N_2 before using. Substrate temperature was kept constant at room temperature during spraying. The silica colloidal was sprayed with the flow rate about ~2 mL/min. Final, textured structure based on silica dispersion was formed by heating it in the air at 200 ^oC for 20 min.

III. RESULTS AND DISCUSSION

The silica particles (SiO_2) are growth and dispersed directly on the glass via modified Stöber method and spray technique as Figure 2. The formation mono-dispersed or multi-dispersed layer of SiO_2 on glass depends on the times of spraying.

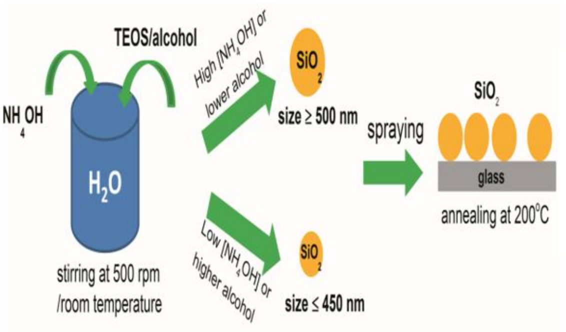

Figure2. The process to fabricate texture surface based on silica particle on glass through Stöber method and spray technique.

Figure 3 shows all the silica spheres preserve a spherical morphology and mono-dispersion onto glass without aggregation between particles or overlap to form layer-by-layer. We can see that the SiO_2 particles formation on the glass is distributed uniformly. The diameter of spherical silica is uniform at approximately 200 nm. It is clearly observed that the SiO2 dispersed mono-layer and could not observe the big aggregation for all samples with different scale bar. This result can be explained due to facile method to the prepare

SiO_2 as well as highly controlled the step of spray SiO_2 on the glass. The good distribution and highly uniformly as-synthesized SiO_2 could open the new wat to present an effective and reliable method to growth sub-micrometer mono-disperse silica onto glass through Stöber method as a chemical approach, allowing one to attain a light-strapping structure based on silica with uniform size and can be obtained the high diffuse transmittance. More important, the mono-layer or multi-layer of pure silica particles on glass substrate can produce and control by control the times of spray step.

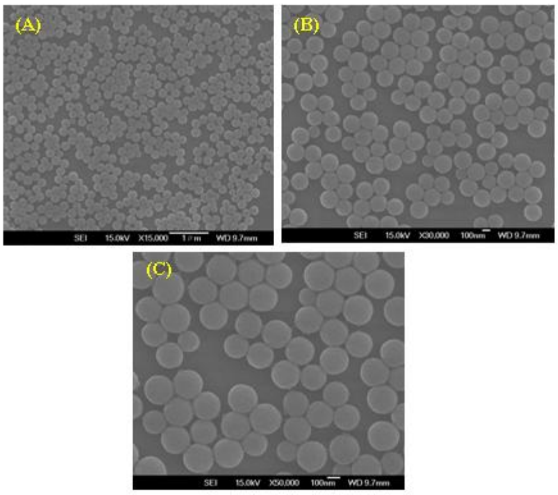

Fig.3. The SEM micrographs for the spherical silica in overview and high resolution images (showing different the scale bar) prepared at concentrations of ammonia about 2.3 mM (A-C). The diameter of spherical silica is uniform at approximately 200 nm.

Figure 4 shows the multi-layer of SiO_2 particles on the glass by controlling the times of spray step. The spraying times is increased 2 times to control the SiO_2 layers. From the Figure 4, we can observe that the SiO_2 particles formed layer by layer on the glass substrate. Interestingly, the SiO_2 particles formed multi-layer on the glass substrate however, there did not observe the aggregation between SiO_2 particles. Depend on the times of SiO_2 layers that we want; it is facile to control the spraying time which could improve the scattering level (haze) of glass substrate. This result suggests that the SiO_2 particles was grown and dispersed the silica particle on the glass via modified Stöber method and spray technique is promising way to fabricate the a novel geometric light trapping structure based on silica particle that can enhance the scattering level (haze) and subsequently to increase the short circuit current density (Jsc) in the solar cells.

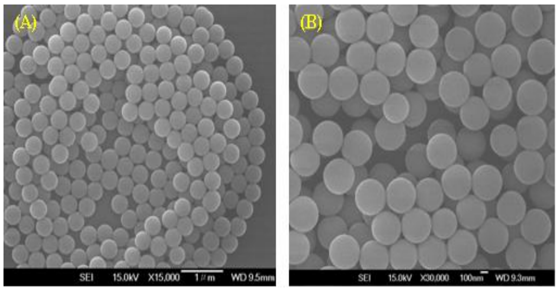

Fig.4. The SEM micrographs for the multi-layer spherical silica in overview and high resolution images (showing different the scale bar) prepared at concentrations of ammonia about 2.3 mM (A-B). The diameter of spherical silica is uniform at approximately 200 nm.

For the mechanism of SiO_2 particles formation, it is indicate that depending on the particle formation mechanism in the sol - precipitation process a large number of primary particles is first nucleated in the initial high supersaturated solution which called the induction period. Then, the primary particles are rapidly aggregated to form stable particles, which grow with the further aggregation of primary particles. After this particle induction period, any further primary particles generated under supersaturation are consumed for the growth of stable particles. In this case, the resulting particles in the product suspension were highly monodispersed in size and spherical in shape, But, if the generation of primary particles by supersaturation exceeds the consumption of primary particles for the growth of stable particles during sol -precipitation, new stable particles are spontaneously formed by the self - aggregation of the extra primary particles and a multi modal distribution of particle. The mechanism of SiO_2 particles formation using the Stöber method is shown below:

$$Si(OC_2H_5)_4 + H_2O \xrightarrow{\text{hydrolysis}} Si(OC_2H_5)_3OH + C_2H_5OH \quad (1) \text{ Hydrolysis reaction}$$

$$\equiv Si-O-H + H-O-Si\equiv \xrightarrow{\text{water condensation}} \equiv Si-O-Si\equiv + H_2O \quad (2) \text{ hydroxyl group of another hydrolysis intermediate (water condensation)}$$

$$\equiv Si-OC_2H_5 + H-O-Si\equiv \xrightarrow{\text{alcohol condensation}} \equiv Si-O-Si\equiv + C_2H_5OH. \quad (3) \text{ the ethoxy group of other TEOS (alcohol condensation)}$$

IV. CONCLUSIONS

We presented the fabrication of a novel geometric light trapping structure based on silica particle. This light trapping structure with good morphology was fabricated through Stöber method and spray technique. The silica particle size was well controlled and uniform from ~200 nm. These results open up the possibility to further fabricate geometric light trapping structure with high scattering level (haze) and subsequently to increase the short circuit current density (Jsc) in the solar cells.

ACKNOWLEGMENT: We would like to thank Professor Lu-Sheng Hong, National Taiwan University of Science and Technology Dr. Nam Giang Nguyen, Department of Engineering, Alstom PhuMy Power Service, Vietnam, Dr. Long Giang Bach, NTT Institute of Hi-Technology, Nguyen Tat Thanh University, Ho Chi Minh City, Vietnam were supported for this work.

REFERENCES

[1] R.K. Iler, The Colloid Chemistry of Silica and Silicates, Cornell University Press, Ithaca, New York, USA, 1955, pp. 730-787.
[2] G. Herberg, Synthesis of monodispersed silica powders II. Controlled growth reaction and continuous production process, J. Eur. Ceram. Soc. 14 (3) (1994) 205-214.
[3] U. Jeong, Y. Wang, M. Ibisate, Y. Xia, Some new developments in the synthesis, functionalization, and utilization of monodisperse colloidal spheres, Adv. Funct. Mater. 15 (12) (2005) 1907-1921.
[4] J.D. Wright, N.A.J.M. Sommerdijk, Sol-Gel Materials: Chemistry and Applications, Gordon and Breach Science Publishers, Amsterdam, 2001, pp. 1-20.
[5] W. Stöber, A. Fink, E. Bohn, Controlled growth of monodisperse silica spheres in the micron size range, J. Colloid Interface Sci. 26 (1) (1968) 62-69
[6] van Blaaderen, J.V. Geest, A. Vrij, Monodisperse colloidal silica spheres from tetraalkoxylanes: Particle formation and growth mechanism, J. Colloid Interface Sci. 154 (2) (1992) 481-501.
[7] Blaaderen, A.P.M. Kentgens, Particle morphology and chemical microstructure of colloidal silica spheres made from alkoxylanes, J. Non-Cryst. Solids 149 (3) (1992) 161-178.
[8] T. Yokoi, Y. Sakamoto, O. Terasaki, Y. Kubota, T. Okubo, T. Tatsumi, Periodic arrangement of silica nanospheres assisted by amino acids, J. Am. Chem. Soc. 128 (42) (2006) 13664-13665.
[9] Anna Selvan JA. IMT Progress Report. III. IMT: Neuchatel; 1995. pp. 1–7.
[10] Löffl A, Kerber H, Schock H-W, Kluth O, Houben L, Rech B, Wagner H. Solid State Phenomena.1999;67–68:277–282.
[11] Kluth O, Rech B, Houben L, Wieder S, Schöpe G, Beneking C, Wagner H, Löffl A, Schock HW. Thin Solid Films. 1999;351:247–253.
[12] Kluth O, Schöpe G, Hüpkes J, Agashe C, Müller J, Rech B. Thin Solid Films. 2003;442:80–85.
[13] Hüpkes J, Rech B, Kluth O, Repmann T, Zwaygardt B, Müller J, Drese R, Wuttig M. Sol. Energy Mater. Sol. Cells. 2006;90:3054–3060.
[14] Berginski M, Hüpkes J, Schulte M, Schöpe G, Stiebig H, Rech B, Wuttig M. J. Appl. Phys.2007;101:074903.
[15] Zhu H, Hüpkes J, Bunte E, Owen J, Huang SM. Sol. Energy Mater. Sol. Cells. 2011;95:964–968.
[16] Jost G, Bunte E, Worbs J, Siekmann H, Hüpkes J. Proceedings of the 25th European Photovoltaic Solar Energy Conference and Exhibition. WIP: Munich; 2010. pp. 3078–3082.
[17] Fernández S, de Abril O, Naranjo FB, Gandía JJ. Sol. Energy Mater. Sol. Cells. 2011;95:2281–2286.
[18] Johnson EV, Prod′homme P, Boniface C, Huet K, Emeraud T, Cabarrocas PRocai. Sol. Energy Mater. Sol. Cells. 2011;95:2823–2830.
[19] Pust SE, Becker J-P, Worbs J, Klemm SO, Mayrhofer KJJ, Hüpkes J. J. Electrochem. Soc.2011; 158:413–419. D.