JFHB Journal of Food Health and Bioenvironmental Science

Journal of Food Health and Bioenvironmental Science
Journal homepage : http://jfhb.dusit.ac.th/

Synthesis of Silica Nanoparticles by Ultrasonic Spray Pyrolysis Technique for Cream Perfume Formulation Development

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Article info

Article history:
Received : 11 June 2018
Revised : 16 July 2018
Accepted : 19 July 2018

Keywords:
Ultrasonic Spray Pyrolysis,
Silica Nanoparticle,
Cream Perfume

Abstract

This research focused on silica nanoparticles synthesis by ultrasonic spray pyrolysis (USP) technique using tetraethylorthosilicate (TEOS) as a precursor. The synthesized silica nanoparticle was used as a fixative material for cream perfume formulation. The morphology and structure of synthesized silica nanoparticles were examined by Scanning Electron Microscope (SEM) technique and X-ray diffraction technique (XRD), respectively. Adsorption ability of synthesized silica nanoparticle was also analyzed by incipient wetness impregnation technique. The results showed that the synthesis temperature of 500°C provided the smallest size of silica nanoparticle, about 106 nm. The particle size decreased from 347 nm to 106 nm when the synthesis temperature increased from 300°C to 500°C. Comparing two types of cream perfume, with and without silica, by applying cream perfume on glass slide at 37°C for 5 hours, found that odor of cream perfume with silica lasts longer than cream perfume without silica.

Introduction

In the present, nanotechnology is used for cosmetics product development. Nanoparticles are characterized by their physical and chemical properties. Nanoparticles may be suspended in gas such as aerosol (nanoaerosol), suspended in liquid known as nanocolloid or nanohydrosol or embedded in a source known as nanocomposite particle. Silica nanoparticles is an inert substance which has been widely used as a cosmetic ingredient, especially in toothpaste or scrub for cleansing. Silica has stable chemical properties at normal temperature and un-react with other chemical substance. It has high durability to resist the physical action and can be used to mix with water-soluble samples. There are several techniques that have been used for synthesis of silica particles, namely chemical vapor condensation, arc discharge, hydrogen plasma-metal reaction, and laser pyrolysis in the vapor phase, micro-emulsion, hydrothermal, sol-gel, etc. (Tavakoli et al., 2007). Silica can be divided into two types; amorphous and crystalline silica (Büchner et al., 2015). Nanosilica particle as a high porous material has good absorbance property which can be used for cream perfume development and a substitute for other expensive fixatives. Perfume product aromatherapy is a mixture of oil and alcohol (St-Gelais, 2016). It can be extracted from animals, natural flowers or chemical synthesis (Steffen, 2003). The desired features of the fragrance are a long-releasing time, low irritation, allergic reactions, high stability to light or heat

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and high durability.

This research focused on the synthesis of nanosilica using tetraethyl orthosilicate (TEOS) as a precursor by ultrasonic spray pyrolysis (USP) technique at different pyrolysis temperatures (Nocun et al., 2005). USP can also be used to directly prepare silica nanoparticles through a continuous process (Watanabe et al., 2011; Das et al., 2015). The morphology and structure of synthesized silica nanoparticle were characterized by scanning electron microscope (SEM) and X-ray diffraction technique (XRD), respectively. The synthesized nanosilica was used to replace expensive fixative which reacted as a fragrance retainer and slowly released the essential oil or fragrance. It can be used in the development of cream perfume (Sousa et al., 2014; Peña et al., 2012).

Materials and methods

Tetraethylothosilicate (99%; sigma-aldrich) was used as a precursor for the silica synthesis by the ultrasonic pyrolysis (USP) technique, which produced a mist of tertraethylorthosilicate. The ultrasonic wave passed through the reactor tube to enter the pyrolysis process at temperatures of 200, 300, 400, 500 and 600°C. The ultrasonic spray pyrolysis (USP) system consisted of (1) ultrasonic wave generator at frequency of 1.7 MHz; (2) high temperature tube furnace; (3) borosilicate reactor tube and (4) water trap for storing synthesized silica, as shown in Fig.1. The tertraethylorthosilicate solution was added to the ultrasonic generator. The mist of tertraethylorthosilicate solution was generated and then passed into the tube furnace for pyrolysis process under different temperatures. The vacuum pump was used for increasing the pressure drop in the process to improve the driving force to pull the mist solution passed to the water trap where the small solid silica particles were formed. The burning process consisted of the steps of evaporation of water followed by precipitation to obtain the nanosilica particles. The identification of synthesized silica nanoparticles was characterized by X-ray powder diffraction method (Diffractometer D8, Bruker) with Cu Kα radiation. The spectra were recorded with the range of 2θ = 5°- 50°. Surface morphology of synthesized silica nanoparticles were observed by scanning electron microscopy (JEOL, JSM-6510LV). The particles size of synthesized silica was measured by SemAfore program. The absorption property of synthesized silica nanoparticles was performed by incipient wetness impregnation method with three replication using 1 g of synthesized silica to absorb the water. Incipient wetness impregnation is a technique for the synthesis of heterogeneous catalysts. The precursor was dissolved in an aqueous or organic solution. Then the solution was added to support materials containing the same pore volume as the volume of the solution that was added.

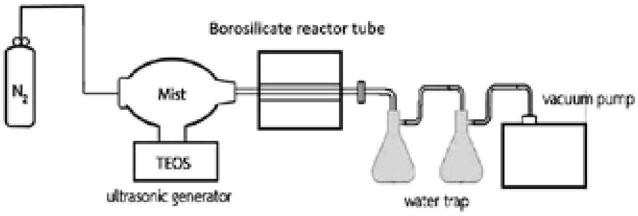

Fig. 1 Schematic diagram of USP

Results and discussion

The nanosilica was synthesized by ultrasonic spray pyrolysis (USP) technique using tetraethyl orthosilicate (TEOS) at various pyrolysis temperatures. From XRD result, all of nanosilica synthesized at temperature of 300, 400 and 500°C had broad peak centered on 23° (Fig.2), indicating that the synthesized silica structures were in a disorderly or amorphous structure (Yu et al., 2014; El-Nahhal et al., 2016).

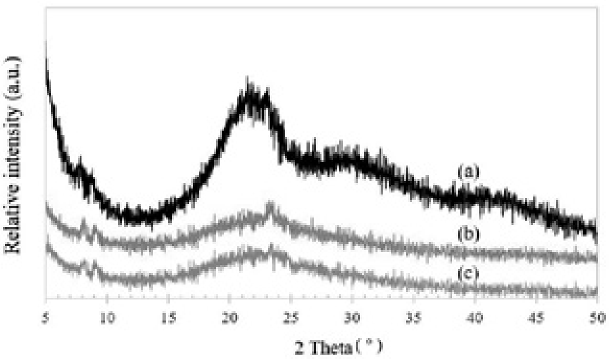

Fig. 2 XRD pattern of synthesized silica at (a) 300°C, (b) 400°C and (c) 500°C

Table 1 shows that at the synthesis temperature of 200°C, the mist of TEOS did not burn completely. Therefore, the remaining mist of TEOS solution was passed through to the flask and separated on surface water. Since the synthesis temperature of 200°C was slightly higher than the boiling point of TEOS solution at 167°C, the combustion was incomplete. The complete combustion occurred when the synthesis temperature was between 300 and 500°C, resulting in white opal silica forming inside the reactor. The synthetic temperature at 600°C could not be carried out as auto ignition occurred. At 300°C, the particles had an average particle size of 347 nm as small spherical white powder (Fig. 3(a)). In Fig. 3(b), shows silica synthesized at 400°C having an average particle size of 203 nm as a finer white particle. At 500°C, the particles have an average particle size of 106 nm, the smallest particle size as shown in Fig. 3(c). At a temperature of 500°C causes the small silica particles since a number of small nuclei was formed at high temperature (Watanabe et al., 2011; Lv et al., 2016).

Table 1 Relation between pyrolysis temperatures and silica size

Synthesis Temperature (°C)	Average particle size (nm)	Results
200	-	Incomplete combustion
300	347	Complete combustion
400	203	Complete combustion
500	106	Complete combustion
600	-	Flash point was occurred

Table 2, shows that the silica synthesized at a high temperature has a smaller particle size than the silica synthesized at low temperature, resulting in a high surface area and good adsorption capability. The highest absorption capability of silica synthesized at 500°C was 2.0547 ml/g silica. Hence, the synthesized silica nanoparticles at 500°C were selected to use as fixative for development of cream perfume because it provided the highest absorption properties. Table 3 shows the perfume creams formula containing silica and without silica.

Table 2. The adsorption of synthesized silica at various temperature by incipient wetness technique

Synthesis temperature (°C)	The volume of water absorbed per 1 g of silica (ml)
300	0.5833
400	1.3292
500	2.0547

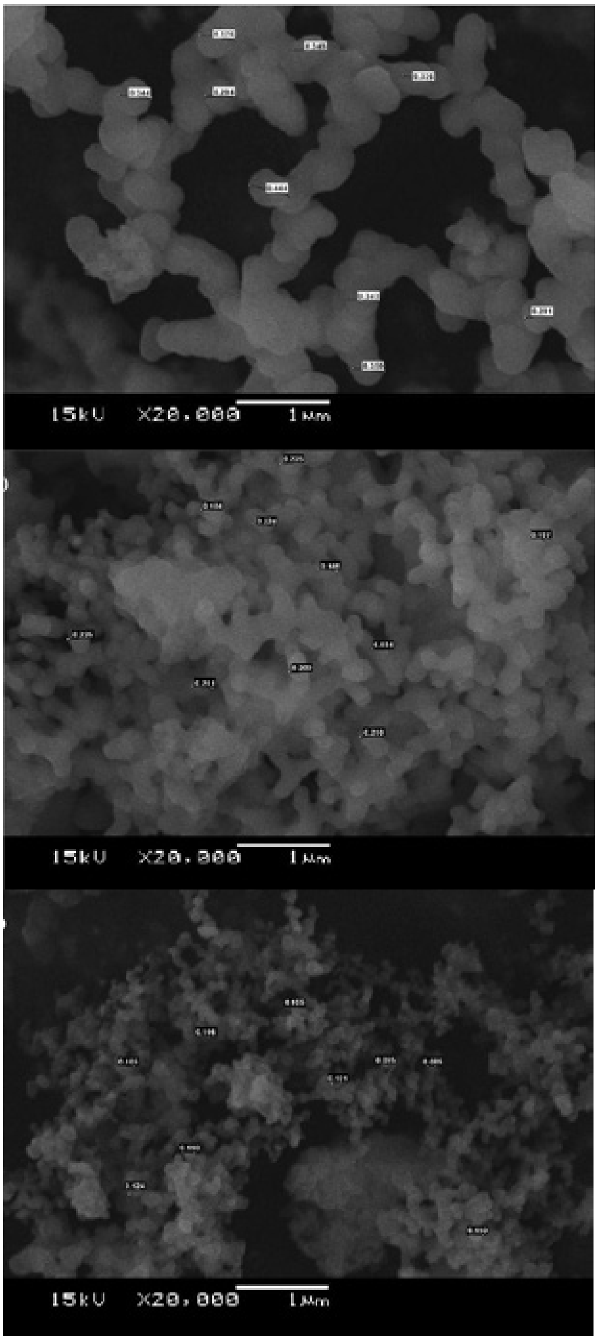

Fig. 3 SEM image of synthesized silica at (a) 300°C, (b) 400°C, and (c) 500°C

Table 3. Perfume creams containing silica and without silica (Silica-free)

Part	INCI Name	Function	% w/w	
			silica	silica free
A	DI water	Solvent	43.60	44.60
	Propylene glycol	Humectant	2.50	2.50
	Acrylates C10-C30 Alkyl Acrylate Crosspolymer	Thickener	1.00	1.00
B	Beewax	Bodying agent	20.00	20.00
	Isopropyl palmitate	Emollient	2.50	2.50
	Sodium olivate and potassium olivate	Emollient	1.50	1.50
	Butylatedhydroxytoluene	Antioxidant	0.50	0.50
	Petrolatum	Emollient	25	25
C	Perfume	Fragrance	1.70	1.70
D	Silica	Fixative	1.00	-
E	Triethanolamine	Neutralize	0.50	0.50
F	Caprylhydroxamic acid (and) Phenoxyethanol (and) Methylpropanediol	Preservative	0.50	0.50
	Total		100.00	100.00

It indicates that the synthesized silica nanoparticles unaffected to chemical ingredient with changing the pH in perfume cream. This pH value is close to the pH of human skin (pH 6). The perfume cream was therefore effective and suitable for all skin types. The appearance was white opaque, thick texture, medium sticky, and no stains on the skin. The synthesized silica was compatible with the perfume cream. There is no separation and no precipitation after leaving. (Fig. 4(a) and (b)). Silica-free perfume cream was fragrant but odorless after leaving on glass slide for 5 hours while the perfume cream with nanosilica had a pleasant aroma and was durable for more than 5 hours. pH value of the perfume creams with and without silica nanoparticles were similar at 7.04.

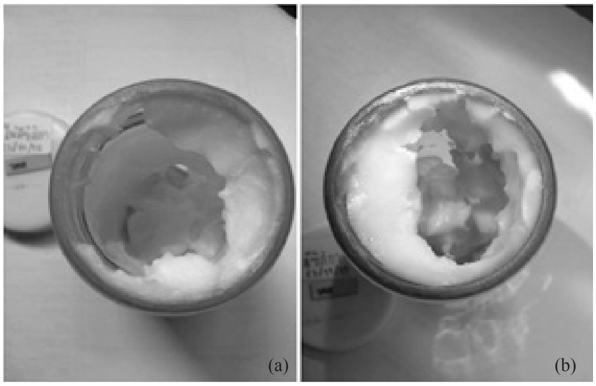

Fig. 4 Image of perfume cream (a) silica-free and (b) with silica

Conclusion

The silica nanoparticles were synthesized using tetraethyl orthosilicate (TEOS) as a precursor by ultrasonic spray pyrolysis (USP) technique at different pyrolysis temperatures. At 500°C, it provided the smallest particle with average particle size of 106 nm and highest absorption of 2.05 ml/g silica. The synthesized silica was compatible with the perfume cream. It showed long-lasting aroma more than silica-free perfume cream.

Acknowledgements

Financially supported by the Thailand Research Fund, Research and Development institute, Suan Dusit University. Department of chemical technology, Faculty of science, Chulalongkorn University also supported this work for equipment.

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