

A RESEARCH ON THE PRODUCTION OF SILK SERICIN POWDERS BY USING SPRAY DRYING METHOD

PÜSKÜRTMELİ KURUTMA YÖNTEMİ KULLANILARAK İPEK SERİSİN TOZUNUN ÜRETİMİ ÜZERİNE BİR ARAŞTIRMA

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ABSTRACT

Sericin protein is usually discarded as a waste in silk industry. Production of silk sericin powders, which can be incorporated into fibers used in textile industry, by spray drying method will enable value-added utilization of waste sericin. In this study aqueous sericin solutions were used as raw material for the production of dry powders using a lab-scale spray dryer. A linear regression analyses were employed, in addition to experimental design at two levels with three factors for the analysis of three responses: moisture content, particle type and agglomeration degree. The process factors were the drying air temperature (120°C and 160°C), the feed rate (1.25×10^{-4} and $2.5 \times 10^{-4} \text{ dm}^3 \text{ s}^{-1}$), and the concentration of sericin solutions of 10% and 30% (w/w) fed to the spray dryer. The three responses were analyzed statistically to determine the effective parameters and it was concluded that moisture content depended on three factors--drying air temperature being the dominant parameter. Particle size and shape depended mainly on feed rate and agglomeration depended on the moisture content of the product.

Key Words: Functional textiles, Silk protein, Sericin, Spray drying, Factorial design.

ÖZET

Sericin proteini genellikle ipek endüstrisinde atık olarak uzaklaştırılmaktadır. Tekstilde kullanılan çeşitli liflerle de etkileşimli olarak kullanılabilen ipek serisin tozlarının püskürtmeli kurutma yöntemiyle eldesi atık serisinin katma değerli bir ürüne dönüştürülmesini mümkün kılacaktır. Bu çalışmada sulu serisin çözeltileri laboratuvar ölçekli püskürtmeli kurutucu kullanılarak kuru toz haline getirilmiştir. Bu işlem sırasında proses parametrelerinin elde edilen toz serisinin nem içeriği, parça tipi ve aglomerasyon derecesine olan etkileri iki seviyeli üç değişkenli bir deney tasarımı ile incelenmiş ve doğrusal regresyon analizleri yapılmıştır. Proses parametrelerinden kurutma havasının sıcaklığı 120°C ve 160°C, besleme hızı 1.25×10^{-4} ve $2.5 \times 10^{-4} \text{ dm}^3 \text{ s}^{-1}$ ve püskürtmeli kurutucuya beslenen çözeltideki serisin konsantrasyonu ağırlıkça % 10 ve % 30 olarak seçilmiştir. Etkin parametreleri bulmak için deney çıktıları istatistiksel olarak analiz edilmiştir. Nem içeriğinin, başta kurutma havası olmak üzere üç proses parametresinden de etkilendiği ortaya çıkarılmıştır. Parça büyüklüğü ve şeklinin başlıca besleme hızına, aglomerasyon derecesinin ise nem içeriğine bağlı olduğu bulunmuştur.

Anahtar Kelimeler: Fonksiyonel tekstiller, İpek proteini, Serisin, Püskürtmeli kurutma, Faktoriyel dizayn.

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1. INTRODUCTION

Silk thread produced by the domesticated silkworm, *Bombyx mori*; is composed of two kinds of protein: a fibrous protein (fibroin) which forms the thread core, and a gum-like protein (sericin) that surrounds the fibroin fibers to join them together. To obtain shiny aspect, soft handle, and elegant drape associated with silk fibers sericin is removed by boiling aqueous solutions containing soap, alkali, synthetic detergents, or organic acids, or recently by proteolytic enzymes (1) and discarded in the silk processing wastewater. Lately, there is a growing interest in the recovery of sericin from silk industry waste water not only to reduce the environmental impact of silk manufacturing (2) but also to utilize it

as a valuable resource for many industries including cosmetics, pharmaceutical, biomedical, and food (3-6).

For decades, much biochemical research has focused on the use of fibroin in biotechnological materials and biomedical applications (7, 8). Sericin, which has excellent moisture absorption and release properties, is antimicrobial (9), UV resistant and has pharmacological functions such as anticoagulation (10), anti-cancer and antioxidant activities (11, 12) with inhibitory action of tyrosinase (13), can find applications in functional biomaterials and textiles as well. It can impart useful and unusual properties to polymer gels, membranes, foams,

fibers, and other composite materials (2). It can be used to produce cryopreservatives, anticoagulants, and biocompatible materials. With its unique properties, sericin can be used in the surface modifications of fibers and fabrics. In fact, it was used as a coating material for cellulose fibers and the treated textiles exhibited a decrease in free formaldehyde content, resistance to electricity, skin irritation and allergic reactions with increased water retention and only a negligible decrease in the textile tensile strength (14). Sericin was also impregnated into polyester fabric to overcome polyester hydrophobicity and to improve UV absorption properties of the sericin treated fabrics (15).

Sericin, proposed as a candidate material for antimicrobial finishing of the textile by incorporating its powder form into synthetic fibers during process, may offer some additional advantages (16). It is a water-soluble natural protein constituting 25%-30% of a versatile material like silk. The protein can be cross-linked, copolymerized, and blended with other macromolecular materials to produce materials with improved properties (2, 17). Considerable scientific research has demonstrated that improved functional property is achievable for carrier-based textile finishing applications by manipulation of the carrier particle size and morphology (18). Manufacturing of carrier-based dry powder formulations can be achieved by various methods including milling, freeze-drying and spray drying (19). The advantages of the spray drying process include the negligible possibility of degradation of heat-sensitive molecules and rapid production of dry powders from the solution in a one-step process with controlled particle characteristics (20-24); the disadvantages were reported to be the difficulty of obtaining a narrow size distribution and the tendency to agglomerate (19). Optimization and control of the powder dispersion and deposition properties could be an important phenomenon in the development of dry powder sericin microspheres, which may be attached to fabrics to impart unique properties.

The aim of this study was to examine the physical characteristics—including particle size distribution, shape and moisture content as well as agglomeration degree of the sericin powders produced by a lab-scale spray dryer under various conditions. Since these characteristics are governed by formulation and process variables, the significance of these variables, as well as interactions between them, were examined using a factorial experimental design and linear regression analysis. The sericin feed concentration was chosen as formulation variable, and the solution feed rate and drying temperature chosen as process variables. This information will be useful for the optimization of the spray drying manufacturing of sericin powders with different dispersion and deposition properties.

2. MATERIALS AND METHODS

Sericin was purchased in powder form from Silk Biochemical Co., Ltd.

(Hangzhou, China). Sericin solutions were prepared by dissolving 10 g sericin powder in deionized water and stirring for 15 minutes to form solutions of 10% and 30% (weight (w)/weight (w)) concentration, respectively. The feed solutions were spray dried with a laboratory-scale spray dryer (Büchi B-290 Mini Spray-Dryer). The solutions were pumped into the drying chamber at a rate of 1.25×10^{-4} and 2.5×10^{-4} $\text{dm}^3 \text{s}^{-1}$. Feed solutions were pneumatically atomized through a nozzle using compressed air at constant pressure. Spray dry conditions, consisting of feed rate and inlet temperature, were set to values of each experiment. The air flowrate was fixed to $10 \text{ dm}^3 \text{ s}^{-1}$. The powders were collected and moisture analyses were performed immediately. The particle size, shape and agglomeration analyses were performed within one week of production. During this time, the powder samples were stored in a molecular sieve 4A desiccator at ambient temperature in sealed sample cups.

The moisture content measurements of the spray dried sericin powders were carried out using a Sartorius Electronic Moisture Analyzer (Model M-100). Each analysis was achieved by using samples of 1.5 g for the duplicates. The powders were heated to the maximum temperature of 105°C and the percent weight losses were measured thermogravimetrically. In order to examine particle morphology, Scanning Electron Microscope (SEM) images were obtained for all spray-dried powders. The analyses were performed using a Philips XL 30S FEG in the Material Research Center, Izmir Institute of Technology. The agglomeration degree of spray dried powders was analyzed by a stereomicroscope (Meiji EMZ-TR) in the Department of Mechanical Engineering, Izmir Institute of Technology.

The effects of major inputs on the responses—including both process and formulation variables—were investigated. A three-factor, two-level full factorial design with two replicates was completed on the sixteen randomized experiments in this study. The process inputs chosen were the solution feed rate and drying air temperature. The formulation variable was aqueous sericin concentration. The response variables were moisture content, particle morphology and agglomeration degree of spray-dried particles. The levels of the factors were coded as -1

and +1, corresponding to the low and high levels respectively. Actual values of the levels are 120°C and 160°C for drying air temperature, 1.25×10^{-4} and 2.5×10^{-4} $\text{dm}^3 \text{ s}^{-1}$ for the solution feed rate and 10% and 30% (w/w) for the concentration of the sericin solutions. All statistical analyses of full factorial design and regression were performed by MINITAB Statistical Software, Release 14 for Windows, State College, Pennsylvania.

3. RESULTS AND DISCUSSION

3.1. Characterization of spray-dried powders

Spray drying of 10% and 30% (w/w) aqueous sericin solutions yielded light yellow powders and sticky agglomerates, depending on the formulation and drying process parameters. Morphology of sericin particles were investigated by scanning electron microscopy and it was observed that a variety of particles from hollow porous spheres to smooth or wrinkled nonporous particles exhibiting either uniform or non-uniform size distributions with particle size ranges of $1-5 \mu\text{m}$ and $5-20 \mu\text{m}$.

Scanning electron micrographs of the representative spray-dried particles are shown in Figure 1. The micrographs depicted different particle shapes, sizes, and size distribution of sericin powder, obtained at each run. Based on visual examination of the micrographs, the particles were graded in six categories (Table 1) according to their shapes (wrinkled, smooth, donut-shaped, straight or a mixture of all), sizes ($1-5 \mu\text{m}$, $5-20 \mu\text{m}$) and particle size distributions (monodisperse or polydisperse). The particles of diameter between $1-5 \mu\text{m}$ were termed "small" while those from $5 \mu\text{m}$ to $20 \mu\text{m}$ in diameter were called "large". The "type" categories indicate the adequacy of the particles for the specific use. The particles manufactured under $5 \mu\text{m}$ are expected to have good aerodynamic properties. Some studies support the superiority of the spherical particles; others claim that wrinkling imparts better aerodynamic properties (20-22). In either case, the greatest problem in application is the tendency of small particles to agglomerate. Hence, in addition to surface properties, particle size and shape are vital for the practicability of the particles.

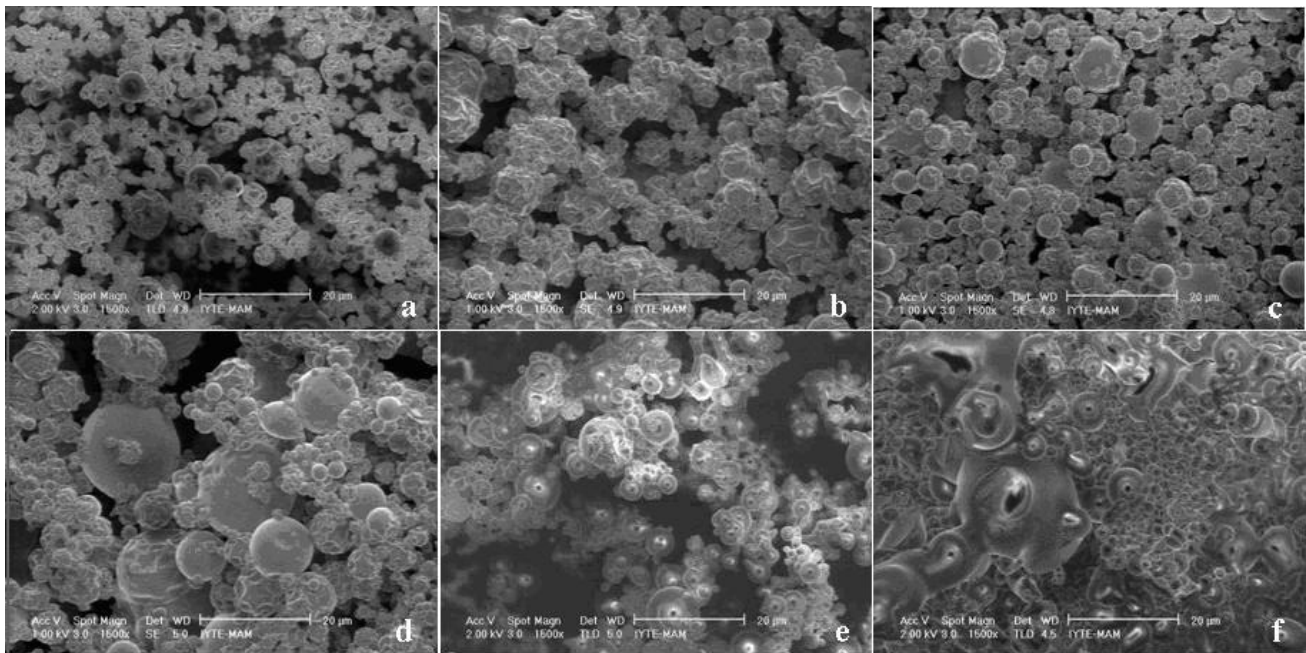


Figure 1. SEM micrographs for the spray-dried particles with different morphologies. (a) Type 1, (b) Type 2, (c) Type 3, (d) Type 4, (e) Type 5 and (f) Type 6.

Table 1. Categorization of the spray-dried particles

| Category | Particle shape | Particle size | Particle size distribution |
|---------------|-----------------------------------|---------------|----------------------------|
| Type 1 | wrinkled and smooth | 1-5 µm | polydisperse |
| Type 2 | wrinkled | 1-5 µm | monodisperse |
| Type 3 | wrinkled and smooth | 1-5 µm | monodisperse |
| Type 4 | wrinkled and smooth | 5-20 µm | polydisperse |
| Type 5 | mixed smooth and donut-shaped | 1-5 µm | polydisperse |
| Type 6 | mixed granules, smooth and sticky | 5-20 µm | polydisperse |

Table 2. Definition and results of randomized experiments in coded factors

| Run Order | Standard Order | Air Drying Temperature | Feed Rate | Sericin Concentration | Moisture Content | | Particle Type | Agglomeration Degree |
|-----------|----------------|------------------------|-----------|-----------------------|------------------|-------|---------------|----------------------|
| | | | | | D_1 | D_2 | | |
| 1 | 2 | +1 | -1 | -1 | 4.06 | 3.60 | 1 | 1 |
| 2 | 4 | +1 | +1 | -1 | 5.89 | 5.65 | 5 | 3 |
| 3 | 14 | +1 | -1 | +1 | 4.49 | 4.12 | 5 | 2 |
| 4 | 1 | -1 | -1 | -1 | 7.26 | 6.38 | 1 | 4 |
| 5 | 13 | -1 | -1 | +1 | 5.96 | 5.76 | 5 | 4 |
| 6 | 7 | -1 | +1 | +1 | 6.29 | 6.00 | 5 | 4 |
| 7 | 10 | +1 | -1 | -1 | 4.46 | 3.96 | 1 | 2 |
| 8 | 9 | -1 | -1 | -1 | 6.62 | 6.09 | 1 | 3 |
| 9 | 12 | +1 | +1 | -1 | 8.29 | 6.41 | 6 | 6 |
| 10 | 6 | +1 | -1 | +1 | 5.15 | 4.79 | 5 | 2 |
| 11 | 16 | +1 | +1 | +1 | 4.55 | 3.91 | 4 | 2 |
| 12 | 11 | -1 | +1 | -1 | 6.03 | 5.61 | 3 | 1 |
| 13 | 5 | -1 | -1 | +1 | 5.73 | 5.51 | 2 | 1 |
| 14 | 15 | -1 | +1 | +1 | 6.50 | 5.83 | 4 | 4 |
| 15 | 8 | +1 | +1 | +1 | 3.02 | 3.08 | 4 | 1 |
| 16 | 3 | -1 | +1 | -1 | 8.34 | 6.95 | 6 | 6 |

Moisture content, types of particle morphology and agglomeration degree are given in Table 2, in the run order of the experiments. Moisture content of the particles is believed to be the most

important variable for surface properties such as surface charge and stickiness. Moisture content was analyzed twice (D_1 and D_2), immediately after drying to minimize

experimental errors. The results of the two moisture analyses of the same sample were recorded as “duplicates” in Table 2.

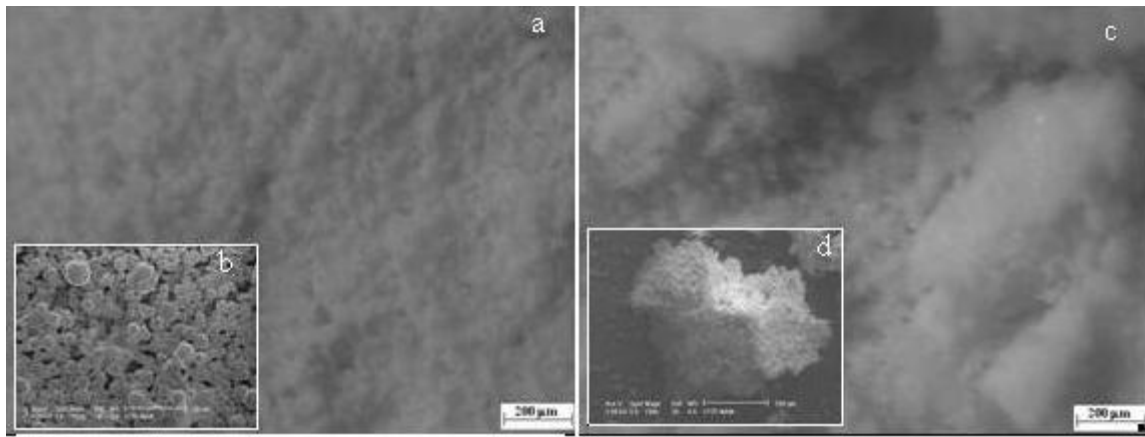


Figure 2. Stereo-microscope images of each category presenting the agglomeration degree; (a) no-agglomeration with agglomeration degree of '1', (b) SEM image of non-agglomerated sample with x1500 magnification (c) drastic agglomeration with degree of '6' (d) the SEM image of a granule with x150 magnification.

Table 3. ANOVA and the factor coefficients for moisture content (coded units)

| Source | df | Sum of Squares | P value |
|---|-------------|----------------|---------|
| Main Effects | 3 | 14.683 | 0.05 * |
| 2-way interactions | 3 | 3.103 | 0.186 |
| 3-way interactions | 1 | 3.624 | 0.028 * |
| Pure Error | 8 | 4.04 | |
| Total | 15 | 25.451 | |
| Term | Coefficient | P value | |
| Constant | 5.5091 | 0.000 * | |
| Temperature (x_1) | -0.7947 | 0.002 * | |
| Feed Rate (x_2) | 0.2628 | 0.177 | |
| Concentration (x_3) | -0.4659 | 0.031 * | |
| Interaction (x_1) (x_2) | 0.1228 | 0.509 | |
| Interaction (x_1) (x_3) | -0.1097 | 0.554 | |
| Interaction (x_2) (x_3) | -0.4084 | 0.051 * | |
| Interaction (x_1) (x_2) (x_3) | -0.4759 | 0.028 * | |

* The terms and interactions with P value less than or equal to 0.05 have been considered as significant.

The spray drying process also led to different agglomeration characteristics in the powder products. Agglomeration occurred for two reasons: Van der Waals forces between ultrafine particles (25) and granulation due to high moisture content. It has been predicted that interparticle attractions of submicron particles may cause the powder product to agglomerate, while insufficient drying caused a drastic agglomeration with formations so called granules as shown in Figure 2. The agglomeration degree of spray-dried particles was examined in six categories, graded between the two cases which correspond to the observations of "no agglomeration" (1) and "drastic agglomeration", leading to formation of large granules (6). The representative SEM micrographs and stereomicroscope images of the two extreme cases are shown in Figure 2. The results were investigated by two

level full factorial designs to observe the effects of process variables, and by regression analysis to observe the effect of response variables on each of the three responses; the results were then analyzed in detail.

3.2. Moisture content

The ANOVA for moisture content in the drying air temperature, feed rate and feed concentration, as factors of a two level full factorial design, was performed and summarized in Table 3. Coefficient estimates and the p-values verified the significance of the main factors of temperature, concentration, second order interaction of feed rate and concentration, and the third order interaction of temperature, feed rate and concentration. Taking into consideration the terms with less than 0.05 p-value, the most effective parameters on moisture content were

obviously temperature and concentration of the feed solution. Feed rate alone did not prove to be an effective parameter, but the interactions with concentration and temperature indicated significance.

The model assumptions were checked according to the normal probability plot of residuals, the predicted versus residual plot, and run order versus residuals plots. None of these plots implied a fault in the analysis, and the model assumption was confirmed. The computer output gives model coefficient estimates and a final prediction equation for moisture content of the spray-dried particles in coded factors. The regression coefficients were calculated from the averages of corresponding effect estimates. The regression equation (Equation 1) of the full model was as follows, with a regression coefficient R^2 of 84.2 %:

$$\hat{y}_1 = 5.5091 - 0.795x_1 + 0.263x_2 - 0.466x_3 + 0.123x_1x_2 - 0.110x_1x_3 - 0.408x_2x_3 - 0.476x_1x_2x_3 \quad (1)$$

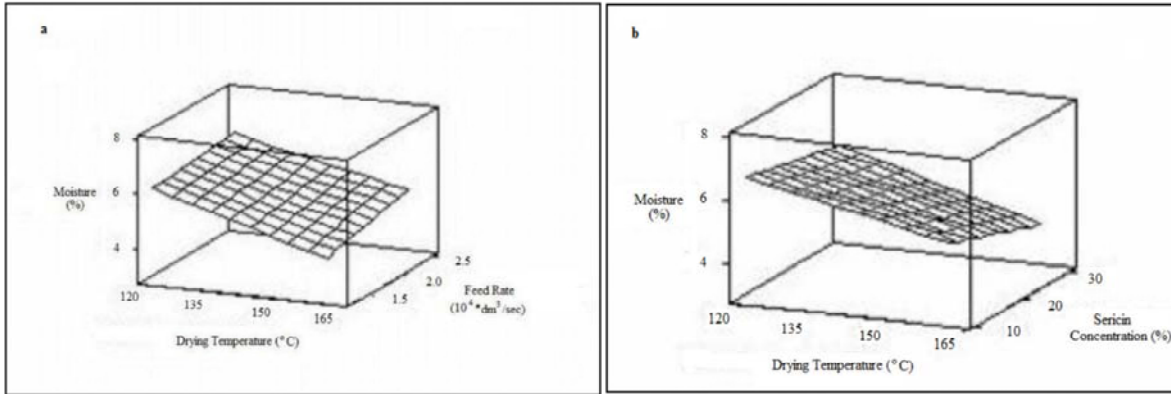


Figure 3. Response surface plots for the moisture content (\hat{y}_1) versus a) drying temperature and feed rate, b) drying temperature and sericin concentration

Where \hat{y}_1 represented estimated moisture percentages and x_1, x_2, x_3 the factors, which were temperature, feed rate and the concentration respectively. Response surface plots for each significant term in the model were analyzed. The response surface plots (Figure 3), shows the interaction of temperature-feed rate and temperature-concentration, respectively. Figure 3a shows that increasing the feed rate increases moisture content at every drying temperature value, but the variation is more significant at higher temperatures. However, Figure 3b shows that increasing the concentration solution decreases moisture content at close levels at varying temperatures. The interaction of concentration and feed rate was negligibly small when compared to the effect of temperature. In both figures (Figure 3a and 3b), the dominant effect of temperature is obvious.

In Figure 3a, it was apparent that temperature and feed rate have almost no interaction. The linear contour plots show the direct affect of temperature, which reaches a constant minimum value at 140°C at the minimum level of feed rate. The response surface becomes flat for this high temperature region and moisture does not decrease further with increasing temperature within the range. Figure 3b shows the curved contour plots indicating the interaction between the factors. That means at a low level of sericin concentration (10% w/w), moisture content increases with increasing feed rate, while at its high level (30% w/w), moisture content decreased slightly with increasing feed rate. The low moisture content was obtained at the maximum sericin concentration 30% w/w, and the minimum feed rate

$1.25 \times 10^{-4} \text{ dm}^3 \text{ s}^{-1}$, when the spray drier was operated at 140°C.

3.3. Particle morphology

Particle morphology categories were ordered according to particle size, particle shape and distributions. Type 1 represented the smallest particle size and a homogeneous size distribution; as the categorical number increased, particle size and size distribution range also increased. Generally two types of particle shape were observed--smooth spheres and wrinkled particles. Wrinkled particles were the result of collapsed hollow spherical structures due to fast evaporation. Lower concentrations of the feed solutions were expected to be the most significant parameter in particle size, and morphology for the solid content of the droplets seemed to be the determining parameter in particle formation. According to experimental analysis, it seems that the most important term was evaporation rate, which depended more on feed solution flow rate, interaction of feed rate and feed concentration. Also, a normal percent probability plot proved that temperature had no effect on particle size and morphology. The software output for the statistical analysis of the reduced model for the response, particle morphology, is given in Table 4. The model is significant, with a p-value less than 0.0005 and R^2 value of 75.7%.

Like the normal percent probability plot of the effects, analysis of variances indicated that x_2, x_3 and $x_2 x_3$ significantly affect the variability of the particle morphology. Regarding the p-values given in the ANOVA output (Table 4), the feed rate and interaction of feed rate and concentration were

equally significant with the highest effects and coefficient estimates. The resultant regression model is represented by Equation (2). The response surface of particle type, with respect to two of the most effective parameters of feed rate and sericin concentration--given as Figure 4--can be examined to observe the serious interaction of these terms.

$$\hat{y}_2 = 3.625 - 0.25x_1 + x_2 + 0.625x_3 - 0.125x_1x_2 - x_2x_3 - 0.375x_1x_2x_3$$

Table 4. ANOVA and the factors coefficients for particle type (coded units)

| Source | df | Sum of Squares | P value |
|---------------------------------|-------------|----------------|---------|
| Main Effects | 3 | 23.25 | 0.018* |
| 2-way interactions | 3 | 16.25 | 0.043* |
| 3-way interactions | 1 | 2.25 | 0.217 |
| Pure Error | 8 | 10.00 | |
| Total | 15 | 51.75 | |
| Term | Coefficient | | P value |
| Constant | 3.625 | | 0.000* |
| Temperature (x_1) | 0.250 | | 0.397 |
| Feed Rate (x_2) | 1.000 | | 0.007* |
| Concentration (x_3) | 0.625 | | 0.056 |
| Interaction ($x_1(x_2)$) | -0.125 | | 0.667 |
| Interaction ($x_1(x_3)$) | 0.000 | | 1.000 |
| Interaction ($x_2(x_3)$) | -1.000 | | 0.007* |
| Interaction ($x_1(x_2)(x_3)$) | -0.375 | | 0.217 |

* The terms and interactions with P value less than or equal to 0.05 have been considered as significant.

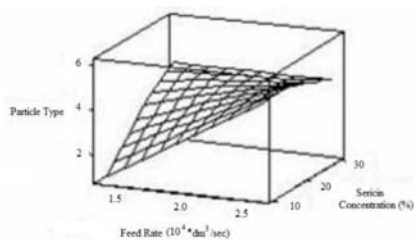


Figure 4. Response surface plot for the particle type (\hat{y}_2) versus sericin concentration and feed rate.

The plot indicates that particle type does not change much at high feed concentrations. But the effect of feedrate becomes extremely important as the sericin concentration fed to the spray dryer decreases. And small particles (less than, or equal to $5 \mu\text{m}$), can only be produced at dilute feed solutions not exceeding 15%, and the feed rate should also be adjusted.

3.4. Agglomeration degree

The half normal and normal probability plots of effects for the response of agglomeration degree designated that none of the factors were significant. As a consequence, the agglomeration response data could not be analyzed by a factorial design procedure. But a stepwise linear regression analysis, including all possible factors of temperature, feed rate, feed concentration, moisture content and particle type, suggested that moisture content and particle type were the effective parameters on agglomeration.

The output for the linear regression analysis of the agglomeration data is given in Table 5. According to the results of the analysis the regression model is given as Equation (3):

$$\hat{y}_3 = -3.33 + 0.944\hat{y}_1 - 0.278\hat{y}_2$$

Table 5. Regression Analysis: Agglomeration versus moisture content (%) and particle type

| Source | df | Sum of Squares | P value |
|-------------------------|-------------|----------------|---------|
| Regression | 2 | 31.612 | 0.000* |
| Residual Error | 13 | 10.138 | |
| Total | 15 | 41.750 | |
| Term | Coefficient | P value | |
| Constant | -3.334 | 0.006* | |
| Moisture (y_1) | 0.944 | 0.000* | |
| Particle Type (y_2) | 0.278 | 0.048* | |

* The terms and interactions with P value less than or equal to 0.05 have been considered as significant

The model explained 75.7 % of the data with respect to R^2 value. The plot of predicted versus observed agglomeration values approved the validity of the model and the plot of the residuals versus predicted values confirmed this result. The dependency of agglomeration degree, as a function of moisture and particle morphology, was given by the scatter plot of the agglomeration data with respect to the both factors of moisture content and particle morphology (Figure 5).

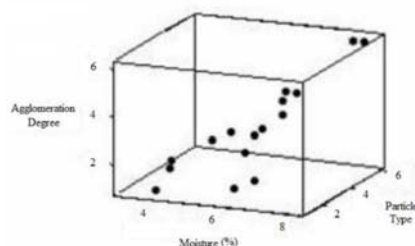


Figure 5. Scatter plot of agglomeration (\hat{y}_3) versus moisture content and particle type

The optimum conditions for the desired product must be decided by evaluating all the responses simultaneously. To be able to define the optimum conditions, the desired particle morphology, moisture and agglomeration degree must be described. Agglomeration is the most important response for application purposes and must be prevented; because the analysis proved that it depends mainly on moisture, moisture must be minimized. Particle morphology requires a more sophisticated decision making process. The term "morphology" covers the entire particle size, particle shape and distribution characteristics; hence it is not an easy task to determine an ideal morphology. Different proposals were reported contrasting spherical with wrinkled particles. But taking the main constraint into account, we can state that particle size must be less than, or equal to, $5 \mu\text{m}$. Checking the characterization results table (Table 1), the smallest particles of the Type 1 agglomerated at even the lowest moisture contents. This behavior can be explained by Van der Waals forces, which have been the main cause of agglomeration of fine particle sizes as the magnitudes get closer to submicron scales (25). Figure 6 shows the main effect plot of particle morphology on mean agglomeration values. The plot proves that there is a minimum degree of agglomeration range at intermediate values of particle types, which gets higher in number with increasing particle size. From the plot, it can be

stated that Type 2 and Type 3 (referring to Figure 1b and 1c), cause the minimum agglomeration with reasonable moisture levels. Hence an optimization process which considers all moisture content, particle type and agglomeration degree is necessary.

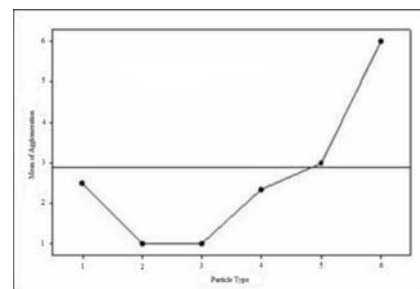


Figure 6. Main effect plot of particle type for agglomeration degree (\hat{y}_3)

4. CONCLUSIONS

As a result of the growing interest in the recovery of sericin from silk industry waste water to utilize it as a valuable resource for many industries, the crucial physical characteristics of the spray-dried sericin influencing the dispersion and deposition behavior--including particle size, morphology, moisture content and agglomeration degree--were examined for formulation and spray drying variables. The production of sericin powders of different characteristics was achieved using a lab-scale spray dryer and controlling the feed solution and operational parameters. Spray drying of the aqueous sericin solutions yielded light yellow powders and sticky agglomerates, depending on the formulation and drying process parameters. The spray drying process led to a particle size below $20 \mu\text{m}$, either in the $1\text{-}5 \mu\text{m}$ or in the $5\text{-}20 \mu\text{m}$ range. The morphology of sericin particles changed from hollow porous spheres to smooth or wrinkled nonporous particles, exhibiting either monodisperse or polydisperse characteristics. The relative importance of the feed solution and spray dryer operational variables and their interactions were analyzed statistically using a full factorial two-level experimental design with three factors. The statistical analysis for the moisture content concluded that the temperature and feed solution concentration were significant factors, while the solution feed rate contributed the interaction terms. The most effective parameters on particle size and morphology were found to be the feed solution concentration and feed rate, while the temperature was an

insignificant variable. The three variables examined had no significant effect on the agglomeration degree. A stepwise regression analysis then concluded that agglomeration strongly depended on particle size and type, in addition to moisture content.

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