Research Article

An Investigation of Optimum NLC-Sunscreen Formulation Using Taguchi Analysis

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This study used three kinds of wax and three kinds of oil, with fixed mixture ratio including UV-blocking materials of ethylhexyl methoxycinnamate, oxybenzone, and avobenzone, and applied hot high-pressure homogenization process to prepare nanolipid sunscreen formulations. The measured particle size of the sunscreen formulations was 100~300 nm around PDI of 0.2 having a moderate polydisperse system. The distribution of zeta potential was -50 mV to -35 mV, showing a stable system. The UV light-absorbing range of 9 groups of sunscreen formulations was 275 nm~380 nm ranging within UVA and UVB. The rheological analysis found that the viscosity change is shear, thinning exhibiting colloid behavior. Taguchi analysis found that the optimum combinations are the carnauba wax and the blackcurrant oil combination for crystallinity and the beeswax and CPG oil for UV absorption. In addition, UV-blocking ability shows that the SPF was 51.5 and PFA was three stars for SU9 formulation. Finally, the effect of temperature on the properties of sunscreen formulations was also explored.

1. Introduction

Nanostructured lipid carriers (NLC), which are expected to become an important carrier of cosmetics and medicine in the future, are at present used for skin care and treatment. Since their introduction to the market in 2005, they have attracted wide attention, which has led to numerous studies of their possible applications [1, 2]. With continuous development, the drug-loading capacity and storage stability of a medicine can be improved. By overcoming the defects of previous liposomes, the percutaneous absorption of a medicine or its principal composition can be improved, and any side effects can be reduced. Numerous studies have indicated the medicament forms of lipid carriers in recent decades [3], such as liposomes and O/W emulsion. Liposomes can be made into micro emulsion, multiple emulsion and solid particles, well combined with excipient; they can be made in quantity. Although the liposome protects the active compound inside the capsule from chemical degradation and regulates and controls the release effect of the compound, it is difficult to

store [1]. The substance inside the capsule leaks rapidly, and the reaction of the hydrophilic substance inside the capsule with the bilaminar membrane makes the structure unstable. The substance inside the capsule cannot be covered stably, and the substance inside the liposome is unstable, thereby limiting the application of liposome.

In order to remedy the defects of liposome, a new carrier was developed in 1990. Solid lipid nanoparticles (SLNs) are a solid colloidal particle drug delivery system with a particle size of 50~1000 nm, which uses solid-state natural or synthetic lipids as the carrier [4–12]. These lipids are biodegradable and are tolerated by the human body. The preparation process of SLN is free of organic dissolvent, the drug leakage amount is minimal, and it is convenient for mass production, thus making it a promising drug delivery system. In general, SLNs are composed of 0.1% (w/w) to 30% (w/w) solid lipid dispersed in an aqueous medium and if necessary stabilized with preferably 0.5% (w/w) to 5% (w/w) surfactant. However, SLN has some drawbacks: a single solid lipid forms lipid crystal, which limits its drug loading capacity. It changes to

perfect crystal gradually, and the drug is pushed out of the lattice during storage [13].

As the use of SLN is limited, NLC was developed in 2000. NLC is made of mixed lipids, and the liquid lipids are mixed in different physical states into the solid lipid and mixed with aqueous phase [4]. The addition of a liquid phase can disorder the regular lattice structure of solid lipid; increasing the ratio of the irregular crystallographic form in the nanoparticle structure, as well as the spatial content loading fat-soluble drug, enhances the drug loading capacity of the carrier. NLC then maintains the advantages of SLN, and its stability is enhanced. The encapsulation of actives in NLC can be found in the literature on medicine and cosmetic applications [1, 14–17].

In addition, NLC represents a breakthrough from the old emulsification technology. For example, there was only the W/O or O/W emulsion forms in the past; however, with NLC technology the solid lipid and the liquid lipid are mixed in the water solution of the surfactant. Furthermore, as the molecules of all the old emulsions had large particle sizes, when they covered the skin surface, the moisture inside the skin evaporated from the pores between the particles, thus rendering the moisture retention inefficient. NLC forms stable fine particles, which improves both the touch feeling and moisture retention. The effects of lipids, surfactants, and storage conditions on the stability of NLC have been reported in the literature [18, 19]. In addition, for cosmetic applications, the adjustment of the formulation to enhance the mechanical barrier and lubricating effect of lipid nanoparticles can protect against skin scratching in the case of skin irritation or an allergic reaction [18]. This is mainly controlled by the selection of the type of lipids and surfactants used for the production of the nanoparticles, which influences the particle size, zeta potential, occlusion, and crystallinity [2, 4, 19]. For example, tripalmitin and tristearin are high purity lipids which create high occlusions, while tricaprin and trilaurin do not show the occlusive effect [2].

The application of NLC for sunscreen formulations with a high sun protection factor (SPF) is a new development. The studies on sunscreen formulations found in the literature [13, 20, 21] report that increased crystallinity improves the UV-blocking effect and that the particulate character also acts as a UV blocker. The incorporation of molecular sunscreen into the particle matrix leads to a synergistic effect for both the molecular sunscreen and UV scattering by the particles [4]. Sunlight contains infrared rays, visible light, and ultraviolet radiation. The most harmful for the human body is ultraviolet radiation, which is divided into UVA, UVB, and UVC. UVB (wavelength 290–320 nm) has higher energy, and as it only reaches the cubicle layer of the skin, the skin turns red and burns. Sunburned skin has pigment precipitate; thus, the horny layer is thickened and the skin is darker. UVA (wavelength 320–420 nm) has a lower energy but is more penetrative; it can penetrate through glass into the dermal layer of the skin. It also stimulates the overgrowth of melanocytes, so that melanins accumulate and spots form when the accumulation is large enough. In addition, the collagen fiber and elastic fiber are destroyed, which causes atrophy. Thus, the skin lacks moisture and elasticity and

becomes flabby and wrinkly. UVC has the highest energy, but it is shielded by the ozonosphere and does not reach the ground. The harm caused by UVA and UVB to the human body is the main concern.

The common evaluation methods for the sun-protection ability of products include (1) sun protection factor (SPF) value-based evaluation and (2) UVA-protection factor (PFA) value-based evaluation. The SPF value is determined by an evaluation of the sunburned red spots and PFA by the degree of suntan. There are physical sunscreens and chemical sunscreens used in sunscreen formulations. The physical sunscreen uses powder to reflect or scatter ultraviolet. In chemical sunscreens the product is mixed with ultraviolet absorbent which can effectively filter the ultraviolet source in the sunlight and let other nonharmful light sources pass through. The physical sunscreen ingredients are mainly made of inorganic powders, such as TiO_2 , ZnO , silica, and ZrO . TiO_2 and ZnO are the ones most commonly used. The chemical sunscreen ingredients are mainly made of chemosynthetic esters, known as ultraviolet absorbents for example, para aminobenzoic acid, salicylates, cinnamates, benzophenones and other ultraviolet absorbing ingredients.

The disadvantage of physical powder sun-screening agents is that they block pores and obstruct the natural permeability of the skin, so the skin loses gloss and elasticity. The experiment herein used chemical sun-screening agent and NLC to minimize the pore blocking, and the chemical composition of the sun-screening agent improved the quality of traditional sun-screening agent as well as the drug loading capacity [20]. Most sunscreen formulations on the market are micron-sized single physical sunscreen products consisting of submicron particles (smaller than 50 nm), which are likely to block pores and have a greasy feeling. NLC can reduce the drug release so that the product is not greasy; thus, it has become the mainstay for cosmeceutical products. Therefore, this study used NLC as the carrier in the sunscreen formulations in order to add to the technology of NLC applications and to develop a new formula of sunscreen formulations at the same time. This study is important for the application of NLC and the development of a new formula. The purposes of this work were to find the most appropriate combination of three kinds of wax and three kinds of oil as reference for sunscreen formulations by using Taguchi analysis [22], to examine the stability of sunscreen formulations, to determine the SPF of the sunscreen formulations, and to find the structure of NLC obtained in this study.

2. Materials and Methods

2.1. Experimental Design. The experimental compositions shown in Table 1 include oil, wax, pentyleneglycol, decyl glucoside, lecithin, water, and the UV-protection agents of S1, S2, and S3. S1, S2, and S3 denote 4-methoxy-cinnamic acid-2-ethylhexyl ester, phenyl ketone-3, and avobenzone, ingredients which resist UVA and UVB. In addition, the oil and wax were those common in the literature, such as echium lycopsis oil, blackcurrant oil, and CPG oil and carnauba wax, Compritol 888 ATO, and beeswax. This experiment tested three kinds of oil and wax, using a thermal high-pressure

TABLE 1: NLC-sunscreen formulation conducted in this work (in wt.%).

B(oil) (mL)	A(wax) (g)	S1 (mL)	S2 (g)	S3 (g)	Pentylene glycol (mL)	Decyl glucoside (mL)	Water (mL)	Lecithin (g)
17%	10%	5.2%	3.5%	1.3%	2%	4.5%	56%	0.5%

TABLE 2: Orthogonal table showing levels of wax and oil*.

Series	A(wax)	B(oil)
SU1	Carnauba wax(1)	Echium lycopsis oil(1)
SU2	Carnauba wax(1)	Blackcurrant oil(2)
SU3	Carnauba wax(1)	CPG oil(3)**
SU4	Compritol 888 ATO(2)	Echium lycopsis oil(1)
SU5	Compritol 888 ATO(2)	Blackcurrant oil(2)
SU6	Compritol 888 ATO(2)	CPG oil(3)
SU7	Beeswax(3)	Echium lycopsis oil(1)
SU8	Beeswax(3)	Blackcurrant oil(2)
SU9	Beeswax(3)	CPG oil(3)

*1, 2, and 3 indicate the level of parameter.

** A kind of triglyceride oil which is extracted from natural plants.

homogenization method (APV, APV2000). The prepared sunscreen formulations were characterized by instrument analysis using a UV/vis analyzer (Jusco, V-670) for UV-absorbance, TGA/DSC (TA, SDT-600) for thermal data analysis, particle-size analyzer (Malvern, Zetasizer, Nano-ZS) for particle-size measurement, zeta-potential meter (Malvern, Zetasizer, Nano-ZS) for stability analysis, TEM (JEM, 2100) for structure analysis, and a rheology meter (Malvern, Gemini2 Rotonetic™ Drive2) for viscosity and dynamic analysis. The number of experiments is shown in Table 2, which is an orthogonal table from the Taguchi experimental design. The optimal set values of the parameters were determined through experiment, according to the different factor numbers and level numbers of the various factors.

2.2. Experimental Procedures and Methods. The operating conditions of mixture ratio, temperature, and pressure were fixed: 5 min preemulsification at 8000 rpm and high-pressure homogenization at 800bar were repeated 5 times to produce the sunscreen formulation, while only the oil and wax ingredients were changed. First, the oil phase ingredient and water phase ingredient were heated separately to 85°C; then, the oil phase was added to the water phase, and the temperature was maintained at 85°C. The mixture was put in the homogenizer for preemulsification at rotation speed of 8000 rpm for 5 min, the preemulsified sample was put in the high-pressure homogenizer, and the experiment was conducted at a pressure of 800bar for 5-cycle times. The sunscreen formulation sample was collected and characterized when it cooled to room temperature. In order to compare the effect of storage temperature on the property of NLC, the sample was divided into two parts, with one part kept at 5°C and the other at 25°C. The sunscreen formulation samples were used to examine the particle size, zeta potential, thermal data, UV-absorption, rheological behavior, TEM, and SPF.

3. Results and Discussion

3.1. Absorption Range of NLC-Sunscreen Formulations. Figures 1(a) and 1(b) show the absorption range of the prepared sunscreen formulation measured using UV/Vis on different days: day 1 and day 45. The peaks exhibited marked differences, depending on the formulations. On the first day, the main absorption peaks of SU1~SU9 were 275 nm to 380 nm, as shown in Figure 1(a), for the absorption ranges of UVA and UVB. After 45 days of storage, the measured UV absorption range, as shown in Figure 1(b), was also in the range of 275 nm to 380 nm, meaning that the prepared sunscreen formulation had UV absorption stability. The absorption range obtained here was comparable with the formulations reported in the literature [4]. The absorption peaks for beeswax (SU7, SU8, and SU9) were higher than those for Compritol 888 ATO or carnauba wax. This could be explained from the viewpoint of particle size and crystallinity. The particle size on the first day for beeswax (115–142.3 nm) was smaller than that for Compritol 888 ATO (140.8–160.1 nm) or carnauba wax (180.7–216.7 nm). The larger particles in the Compritol 888 ATO and carnauba wax NLC formulations were able to scatter and reflect incoming UV radiation, resulting in a decrease of outgoing UV light [20]. A similar report was found in the literature [23]. Crystallinity is another factor which affects UV-blocking ability. This phenomenon was reported by Müller et al. [4], who found that highly crystalline solid lipid nanoparticles act as particulate UV blockers by scattering the light efficiently. In the present study, the crystallinity for Compritol 888 ATO (13.57–22.30%) was smaller than that for carnauba wax (52.86–72.98%) or beeswax (47.63–82.54%), leading to a lower UV-blocking ability. The particle size and crystallinity will be discussed later.

3.2. Particle Size of NLC-Sunscreen Formulations. The particle size and its distribution are very important for the stability of the colloid system. This study used a thermal high pressure homogenization method to prepare NLC under changes in oil and wax production and storage temperature. The particle size and PDI are shown in Tables 3 and 4. As shown in Table 3 and Figure 2, the particle size was about 110–210 nm on the first day; for all systems, the particle size at 5°C was smaller than that at 25°C. In addition, for all particles, the size increased with an increase in storage time, most significantly at a 25°C storage temperature. Generally, the wax affecting the particle size in order of significance was carnauba wax > Compritol 888 ATO > beeswax. Using SU1 (carnauba waxes) as an example, as shown in Table 3 and Figure 2, the sizes were 216.7 nm, 410.2 nm, and 507.1 nm for day 1, day 30, and day 60, respectively. This indicated that the particles became aggregates with increased storage time. On the other hand, for SU7 (beeswax), the sizes were 142.3 nm (day 1), 147.5 nm

FIGURE 1: SU1~SU9 UV absorption peaks at different days, showing the stability of sunscreen formulation obtained in this work.

TABLE 3: Particle size at different storage times.

	d (nm) (day 1)	d (nm) (day 30)		d (nm) (day 60)	
		5°C	25°C	5°C	25°C
SU1	216.7 ± 20.96	235 ± 2.458	410.2 ± 10.4	253.9 ± 6.03	507.1 ± 8.10
SU2	180.7 ± 9.763	248.7 ± 3.15	250.9 ± 5.10	323.5 ± 8.61	310.3 ± 9.72
SU3	185.2 ± 3.798	266.8 ± 0.77	304.1 ± 4.91	303.1 ± 5.78	306.8 ± 4.22
SU4	140.8 ± 6.165	185.5 ± 5.08	182.8 ± 3.55	186.3 ± 3.87	195.3 ± 3.50
SU5	145.4 ± 9.296	191.6 ± 4.2	210.3 ± 4.36	182.3 ± 4.32	222.4 ± 6.10
SU6	160.1 ± 7.204	205.2 ± 4.65	244.1 ± 3.87	208.5 ± 4.45	314.8 ± 11.2
SU7	142.3 ± 4.120	142.9 ± 3.89	147.5 ± 3.10	143.2 ± 4.39	154.5 ± 4.39
SU8	127.9 ± 1.931	145.1 ± 4.47	151.8 ± 2.66	150.5 ± 5.26	156.2 ± 4.33
SU9	115.5 ± 2.951	141.7 ± 2.61	143.4 ± 2.27	153.6 ± 3.25	149.8 ± 4.36

(day 30), and 154.5 nm (day 60), indicating little change in size, with more stable formulations obtained from beeswax as compared with those from the other waxes.

The PDI value shows the distribution of particle size. Monodispersion occurs when the PDI is less than 0.05; it approaches monodispersion at a PDI of less than 0.08. A moderate dispersion system has 0.08~0.7 PDI, and a multidispersion system has PDI greater than 0.7. Table 4 shows the PDI values of NLC. As seen, the PDI value was about 0.1 on the first day, except for the carnauba wax formulations; thus, the prepared NLC was a moderate dispersion system, which remained within 0.2 for most samples after long-time storage. The samples had a consistent particle size, indicating that the prepared sunscreen formulations had particle size distribution and size stability.

3.3. Zeta Potential (ζ) of NLC-Sunscreen Formulations. The physical stability of NLC can be evaluated by measuring the zeta potential. According to the theory of DLVO, a system can be regarded as stable if the electrostatic repulsion dominates

the attractive van der Waals forces [18]. Generally, a zeta potential above 30 mV means that the colloidal system is in a stable state [15]. There is less particle aggregation because of the electrostatic repulsion between colloidal particles. According to the literature, a zeta potential of 60 mV means that the colloidal system has super high stability, a zeta potential of 15 mV means that the colloidal particles may have aggregation, and a zeta potential below 5 mV means that the colloidal particles have severe aggregation.

Data in Table 5 and Figure 3 show that the zeta potential value was about -40 mV after preparation, which is considered particle dispersion with good stability. The interface potential value did not change significantly after long-term storage, indicating that the prepared NLC had good stability. The effects of storage temperature and long-term storage on the zeta potential were compared. Although temperature affects the zeta potential, the NLC prepared in the experiment stayed above -35 mV, whether it was stored at 25°C or 5°C, and was in a stable colloid distribution state. In addition, the zeta potential for beeswax was higher than that for carnauba

TABLE 4: PDI for various materials at different storage times.

	PDI (day 1)	PDI (day 30)		PDI (day 60)	
		5°C	25°C	5°C	25°C
SU1	0.349 ± 0.035	0.264 ± 0.04	0.400 ± 0.01	0.305 ± 0.03	0.408 ± 0.01
SU2	0.186 ± 0.015	0.164 ± 0.05	0.176 ± 0.01	0.281 ± 0.01	0.164 ± 0.01
SU3	0.185 ± 0.007	0.179 ± 0.01	0.215 ± 0.02	0.143 ± 0.01	0.201 ± 0.01
SU4	0.076 ± 0.002	0.097 ± 0.03	0.098 ± 0.02	0.119 ± 0.01	0.074 ± 0.02
SU5	0.057 ± 0.015	0.125 ± 0.02	0.161 ± 0.01	0.109 ± 0.01	0.099 ± 0.03
SU6	0.090 ± 0.046	0.137 ± 0.01	0.154 ± 0.03	0.166 ± 0.01	0.138 ± 0.02
SU7	0.202 ± 0.025	0.095 ± 0.01	0.096 ± 0.02	0.080 ± 0.02	0.129 ± 0.01
SU8	0.082 ± 0.003	0.105 ± 0.02	0.124 ± 0.01	0.113 ± 0.02	0.144 ± 0.01
SU9	0.078 ± 0.008	0.083 ± 0.01	0.095 ± 0.01	0.077 ± 0.02	0.099 ± 0.01

TABLE 5: Zeta-potentials for various samples at different storage times.

	ζ (mV) (day 1)	ζ (mV) (day 30)		ζ (mV) (day 60)	
		5°C	25°C	5°C	25°C
SU1	-28.4 ± 0.987	-29.3 ± 1.03	-35.2 ± 0.99	-35.7 ± 2.00	-36.8 ± 1.65
SU2	-27.5 ± 1.160	-45.9 ± 0.69	-35.8 ± 0.38	-40.4 ± 1.60	-37.0 ± 1.45
SU3	-33.5 ± 0.240	-39.5 ± 0.50	-46.6 ± 0.46	-42.5 ± 0.55	-45.8 ± 0.61
SU4	-28.0 ± 1.41	-38.1 ± 0.99	-45.5 ± 0.9	-38.9 ± 1.45	-37.4 ± 1.75
SU5	-32.0 ± 2.76	-39.0 ± 2.70	-46.6 ± 0.45	-34.4 ± 0.97	-44.1 ± 0.49
SU6	-31.5 ± 0.70	-40.4 ± 1.31	-42.6 ± 1.65	-38.1 ± 1.28	-40.2 ± 1.06
SU7	-32.5 ± 2.65	-49.2 ± 0.61	-50.6 ± 1.37	-42.1 ± 1.63	-40.5 ± 2.07
SU8	-18.1 ± 1.03	-56.8 ± 0.30	-58.6 ± 1.03	-44.9 ± 0.96	-45.1 ± 2.45
SU9	-15.1 ± 1.84	-54.4 ± 1.31	-52.9 ± 1.12	-38.8 ± 1.25	-36.6 ± 1.25

(a) 5°C storage temperature

(b) 25°C storage temperature

FIGURE 2: Particle size variations of different materials and storage at different temperatures.

FIGURE 3: Zeta-potential variations of different materials and storage at different temperatures.

TABLE 6: Comparison of NLC crystallinity under different conditions.

Sample	MP (°C)	ΔH (J/g)	CI (%)
SU1	76.60	21.23	52.86
SU2	77.80	29.31	72.98
SU3	76.51	24.31	60.56
SU4	62.08	13.09	13.57
SU5	61.02	21.52	22.30
SU6	59.82	14.69	15.22
SU7	67.10	6.43	82.54
SU8	67.45	3.71	47.63
SU9	67.33	3.80	48.78

wax or Compritol 888 ATO after storage. In addition, the systems obtained herein were more stable as compared with SLN systems, with a zeta-potential range of (−37.9 mV)–(−18.8 mV) for the three oils [5]. This also demonstrated that NLC had more stability than SLN.

3.4. Crystallinity Index (CI) of NLC-Sunscreen Formulations. The structure of a material varies with heating. This study used DSC to measure the variation of samples under heating with a heating rate at 5°C/min for each run after preparation, as shown in Figure 4 for SU1. The variation of the enthalpy and melting point (MP) of the samples was obtained from the heat change, and then the crystallinity index of the samples was determined using enthalpy. The MP and enthalpy for SU1 determined herein were 76.6°C and 21.23 J/g, respectively. All data obtained in this work are listed in Table 6. The data on the variations were quite different, depending on the formulations. As for the three kinds of solid lipid used in this

FIGURE 4: Determination of MP and enthalpy of NLC by using DSC analysis.

study, the melting point of carnauba wax was 85.56°C and that of the enthalpy was 40.16 J/g. The melting point of Compritol 888 ATO was 74.76°C and that of the enthalpy was 96.49 J/g. The melting point of beeswax was 81.17°C and that of the enthalpy was 7.79 J/g. If the crystallinity index of solid lipid material was 100%, the crystallinity index of other samples could be deduced. The CI is defined as follows [18]:

$$CI (\%) = \left(\frac{\Delta H_{NLC}}{\Delta H_{lipid}} \right) \times 100\%, \quad (1)$$

where ΔH_{NLC} represents the enthalpy of the prepared sample and ΔH_{lipid} represents the enthalpy of pure wax. As shown in Table 6, the different compositions had different crystallinity

FIGURE 5: Viscosity change is shear thinning.

because different wax types make the crystal arrangement present an incomplete form. A larger crystallinity index improved the UV-blocking effect, as shown in Figure 1, which was similar to that reported in the literature [4, 23]. In the present study, the beeswax NLC was found to be the most efficient.

3.5. Rheological Behavior of NLC-Sunscreen Formulations. Using a rheology meter (Gemini2 Rotonetic™ Drive2) with a cone-and-plate device, the rheological behavior of the sunscreen formulation could be determined. As shown in Figure 5, the viscosity of the prepared sample decreased as the shear rate increased. This viscosity change can be called shear thinning, as with oil paint. A similar report in the literature showed the shear thinning phenomena for seven sun-blocking formulations [21]. If this sample was applied to sunscreen formulations, it could be smeared on the skin uniformly. Whether the sample is colloid or liquid can be known by measuring its viscosity and elasticity, with the elastic modulus (G') and viscous modulus (G'') obtained by using oscillation frequency to apply interference. When $G' > G''$, it is colloid if $G'' > G'$, it is liquid. The NLC prepared in this experiment was $G' > G''$, as shown in Figure 6, so it was in a colloid state. Figure 6 shows the changes in the viscous modulus and elastic modulus at different temperatures. The viscous modulus increased with the oscillation frequency at low temperature; thus, the colloid behavior at low temperature was more obvious than that at room temperature. The colloid applied to sunscreen formulations could enhance the adhesion of actives to the skin and thus enhance the uniformity of smearing. This is important

TABLE 7: Effects of the CI S/N ratio factor.

Level	A-wax	B-oil
1	35.79	31.82
2	24.42	32.60
3	35.22	31.02
Delta	11.37	1.58
Rank	1	2

for cosmetic products because these products need to create a “nice application feeling” when applied by the customer [4].

3.6. Analysis of Parameter Importance. After the sunscreen formulations were prepared, the particle size, zeta potential, crystallinity, and ultraviolet absorption intensity were tested and then analyzed by Taguchi software. The S/N ratio factors that affect particle size, zeta potential, crystallinity, and ultraviolet absorbance could be obtained from the larger-the-better and the smaller-the-better by the software. First, using CI as an example, the Taguchi analysis results are shown in Figure 7 and Table 7. The parameters affecting the CI in order of importance were carnauba wax > beeswax > Compritol 888 ATO and blackcurrant oil > echium lycopsis oil > CPG oil. Therefore, the optimum combination was SU2, with the ingredients of carnauba wax and blackcurrant oil. The effect of the oil on the recrystallization to stable lipid modifications was found in the literature [4].

Second, the parameters affecting the particle size in order of importance were beeswax > Compritol 888 ATO > carnauba wax and blackcurrant oil > CPG oil > echium lycopsis oil. Therefore, the optimum combination was SU8 for the ingredients of beeswax and blackcurrant oil. Third, the parameters affecting the zeta potential in order of importance were Compritol 888 ATO > carnauba wax > beeswax and echium lycopsis oil > blackcurrant oil > CPG oil. Therefore, the optimum combination was SU4 for the ingredients of Compritol 888 ATO and echium lycopsis oil. Fourth, the parameters affecting the UV absorption value in order of importance were beeswax > carnauba wax > Compritol 888 ATO and CPG oil > echium lycopsis oil > blackcurrant oil. Therefore, the optimum combination was SU9 for the ingredients of beeswax and CPG oil.

Table 8 shows the four dimensions obtained from this study. The optimal composition was obtained from the Taguchi analysis. The basic differences in particle size and zeta potential were small. In terms of sunscreen formulations, crystallinity is the basis of coating UV-blocking material, and UV light absorption is the effect of the sunscreen formulations. Therefore, the sunscreen formulations prepared in this subject focused on crystallinity and UV absorption.

3.7. Determinations of SPF and PFA Values. Herein, SU9 was used for the measurements of SPF for UVB and PFA for UVA, respectively. Optometrics SPF-290S Analyzer according to a US FDA measurement standard, the scan spectrum and monochromatic protection factors (MPF) are shown in Figure 8(a) for twelve time scans. The results showed that SPF

FIGURE 6: Viscoelasticity at different storage temperatures (SU3).

TABLE 8: Optimum combinations obtained from various analyses.

	Particle size	Zeta potential	Crystallinity	UV absorption
Optimum combination	SU8	SU4	SU2	SU9
Wax and oil ingredients	Beeswax Blackcurrant	Compritol 888ATO Echium lycopsis oil	Carnauba wax Blackcurrant	Beeswax CPG oil

FIGURE 7: Effects of the crystallinity S/N ratio factor.

was 51.5 and PFA was three stars, indicating high UVA and UVB protection for the SU9 formulation. In order to prepare a sun protection product for market, we take 30% of SU9 formulation mixed with cream for a modified prescription. The measured SPF and PFA were 25.22, as in the scan spectrum shown in Figure 8(b), for two stars, respectively, indicating moderate UV protection ability. However, the occlusion factors were 98% and 96% for SPFs of 51.5 and 25.22, respectively. The particulate character also acts as a UV blocker. Incorporation of molecular sunscreens into a matrix of particles leads to a synergistic effect of both molecular sunscreen and UV scattering by the particles [4, 20]. SPF investigations were reported by Nesseem [21], who studied seven sunscreen formulations by adjusting the mixture of organic and inorganic UV blockers for an SPF range of 19.97–56.17. However, the results obtained herein demonstrated that

the chemical sunscreen mixture of S1, S2, and S3 in NLC presented a new option for UV blocking in the future.

3.8. Structure of NLC-Sunscreen Formulation. Figure 9(a) shows the TEM photograph of SU9. It was found that the solid lipid matrix contained tiny liquid oil nanocompartments, as compared with Figure 9(b), the multiple type, that is, an oil-in-solid lipid- (fat-) in-water (O/F/W) dispersion system [23]. Therefore, increasing the oil concentration, the solubility of oil molecules in the solid lipid was exceeded, and the oil precipitates in the form of fine droplets were incorporated into the solid lipid matrix, resulting in phase separation and the formation of oil nanocompartments [4]. The phenomena could be found during the cooling process after production of NLC by the hot homogenization process. In this manner, the oil nanocompartments were incorporated into the solid matrix; they contained a higher amount of active compounds, but their release was still controlled by the surrounding solid lipid barrier.

4. Conclusions

This study successfully obtained sunscreen formulations from a combination of three kinds of wax and three kinds of oil by using a hot high-pressure homogenization process. The particle size of the prepared NLC sunscreen formulations was 100–300 nm; low-temperature (5°C) storage was better than room-temperature (25°C) storage. The PDI was about 0.2 of a moderate polydisperse system. Using Taguchi analysis with an S/N ratio for particle size, zeta potential, CI, and VU absorption wave, the optimum formulations were found. The obtained optimum combinations were SU8, SU4, SU2, and SU9 for particle size, zeta potential, CI, and VU absorption wave, respectively. As the particle size and zeta potential

FIGURE 8: Monochromatic protection factors (MPF) for sun protection analysis (SU9) with and without modification.

(a) TEM photograph of SU9

(b) Multiple type

FIGURE 9: TEM photograph of SU9 formulation and multiple type structure of NLC.

presented a stable system, this study took crystallinity and UV light absorption as the basis for the preparation of sunscreen formulations. According to sun protection products on the market, CI and VU-blocking are considered to be better for

sunscreen formulations. The measured SPF and PFA data for SU9 effectively exhibited UV-blocking ability. In addition, the structure of the NLC-sunscreen formulations obtained in this work was that of an O/F/W dispersion system.

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