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Optimization of an improved aqueous method for the production of high quality white sesame oil and de-oiled meal

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SUMMARY: Research into the production of white sesame oil by aqueous extraction has been promoted because of concerns about the environment, health, and cost. The advanced aqueous method using a 1.95:10 liquid-to-raw material ratio, which was finally developed in this study, recovered 96.06% white sesame oil and produced de-oiled meal with high quality (3.98% residual oil content). The acid value and peroxide value of the oil produced were quite low at 0.19 mg KOH/kg and < 3.25 mmol/kg, respectively, which were better than the values required by the Chinese national standard for first class edible sesame oils and oils produced by hexane extraction. No wastewater was discharged during the extraction of white sesame oils by an aqueous salt solution. The protein rich de-oiled meal may be a good material for making protein isolate with high purity (e.g. > 90%). It can also be a nutritious ingredient or raw material for producing many food products.

KEYWORDS*: Aqueous solution; Extraction of oil; Good quality; High recovery rate of oil; Protein-rich de-oiled meal*

RESUMEN: ***Optimización de un método acuoso para la producción de aceite de sésamo blanco de alta calidad y harina sin aceite***. La investigación sobre la producción de aceite de sésamo blanco por extracción acuosa se ha promovido debido a las preocupaciones sobre el medio ambiente, la salud y el costo. El método acuoso avanzado que utiliza una relación líquido-materia prima de 1,95:10, desarrollado en este estudio, recuperó un 96,06% de aceite de sésamo blanco y produjo harina sin aceite con alta calidad (3,98% de contenido de aceite residual). El valor de acidez y el índice de peróxido del aceite producido fueron bastante bajos, 0.19 mgKOH/kg y <3,25 mmol/kg, respectivamente, que fueron mejores que los requeridos por la norma nacional china para los aceites de sésamo comestibles de primera clase y los aceites producidos por extracción con hexano. No se descargaron aguas residuales durante la extracción de los aceites de sésamo blancos mediante una solución salina acuosa. La harina sin materia grasa, rica en proteínas puede ser un buen material para hacer proteínas aisladas con alta pureza (p. ej., > 90%). También puede ser un ingrediente nutritivo o materia prima para producir muchos productos alimenticios.

PALABRAS CLAVE: *Alta tasa de recuperación de aceite; Buena calidad; Extracción de aceite; Harina deshidratada rica en proteínas; Solución acuosa*

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1. INTRODUCTION

White sesame (*Sesamum indicum* L.) is one of the most important oil seed crops in the world (Latif and Anwar, 2011). Its seeds normally contain as high as > 50% oil and ca. 24% protein. The health effects of white sesame seeds have been reported in the literature (Lin *et al*., 2017; Lee *et al*., 2015; Lee *et al*., 2010).

Recently, scientific evidence has indicated that white sesame seed oil (WSSO) has beneficial effects on health such as the improvement of blood lipid profile and the activities of anti-inflammation and antimutagenesis (Lee *et al.*, 2010; Aslam *et al.*, 2018; Uzun *et al.*, 2008) as well as endothelial function in hypertensive men (Kalliopi *et al.*, 2013). Experiments on animals indicated that sesame oil improved cardiac health, glucose control or biomarkers for hepatic stress (Aslam *et al.*, 2017) and increased NO generation (Cebova *et al.*, 2018). Fat-soluble functional compounds (such as sesamoline, sesamine, squalene and phytosterols) and vitamins (e.g. tocopherols and carotenoids) are present in WSSO and contribute to their health-beneficial effects. Sesame oil usually has a characteristic flavor and high stability to oxidation because it contains compounds which have a marked antioxidant activity (Crews *et al.*, 2006; Latta, 1991).

The neuroprotective potential and antioxidant activity of de-oiled white sesame seed meal (DBSSM) has been reported in the literature (Ben *et al.*, 2016). Water-soluble and functional compounds (such as phospholipids, B-vitamins and ferulic or vanillic acid) are present in DBSSM and contribute to its health-beneficial effects. Furthermore, the benefit of vegetable proteins to human health has been well reviewed in the literature (Asgar *et al.*, 2010; Craig, 2010).

Therefore, a method able to obtain both oil and DBSSM with high quality is preferred for processing white sesame seeds. Hot pressing is not good for extracting oil from white sesame seeds because it produces dark DBSSM with denatured proteins. Although direct cold pressing can produce oil and DBSSM with high quality, its extraction efficiency is low. Although solvent (e.g. hexane) extraction is able to efficiently produce oil and DBSSM with high quality, it has many disadvantages, as reported in the literature (Tu *et al.*, 2017; Tu and Wu, 2019; Zhang *et al.*, 2006; Stalker, and Wilson, 2015; Latif and Anwar, 2009). The traditional aqueous method using large quantities of water assisted by enzyme can produce both oil and hydrolyzed proteins (Latif and Anwar, 2011; Xu *et al.*, 2017; Ribeiro *et al.*, 2016; Hou *et al.*, 2013), but has disadvantages (e.g. forming serious emulsion, low free oil yield, large quantities of waste water, high cost of drying DBSSM) which have also been reported in the literature (Lv and Wu, 2018; Li *et al.*, 2016; Ravber *et al.*, 2015; Zhang *et al.*, 2011; Jiang, 2010).

The objective of this study was to develop an advanced aqueous method of efficiently producing white sesame oil and de-oiled meal with high quality based on a new theory of separation of oil and relying on the aggregation of hydrophilic groups of compounds (with or without hydrophobic groups) through hydrogen bonds with the prevention of their solubilization or dispersion by adding a small quantity of water as a pre-treatment. The experiments involved the establishment of critical conditions affecting the recovery rate and quality of the oil and DBSSM produced and a comparison of the aqueous method based on the new theory to cold pressing, the traditional aqueous method assisted by enzymes and hexane extraction.

2. MATERIALS AND METHODS

2.1. Materials

White sesame seeds were obtained from Yonghui Supermarket, Beibei District, Chongqing, China. All chemicals and reagents used were of analytical grade.

2.2. General aqueous extraction procedure for all experiments conducted in this study

The flow chart for the general process to be studied is shown in Figure 1. White sesame seeds with peel were baked at a certain temperature (1st variable factor) for a certain time. The baked

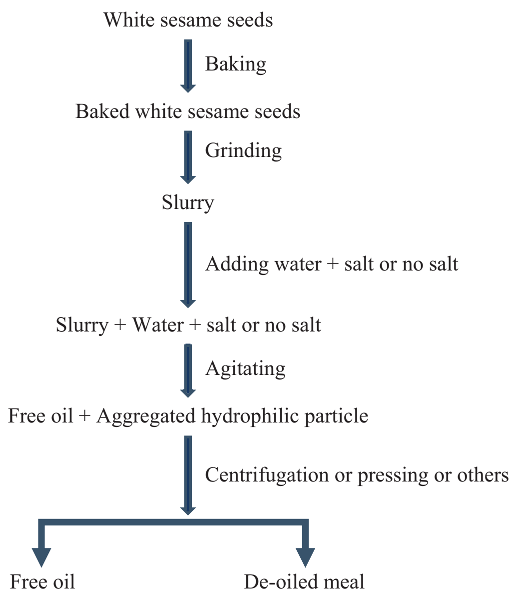

Figure 1. Flow chart for producing white sesame oil and de-oiled meal using an aqueous solution or water.

seeds were ground into continuous slurry passed through a sieve having a certain pore size (2nd variable factor). A certain amount (3rd variable factor) of aqueous salt (NaCl) solution with a certain concentration (4th variable factor) was added to 10.00 g seed slurry in a 20 ml centrifuge tube. Then, the mixture of the salt solution and the seed slurry was agitated for a certain time (5th variable factor) at a certain temperature (6th variable factor). Finally, free oils were collected by centrifugation three times at 4000 r/min for 30 min followed by cold screw expressing once (the presser made by Ai Bang Agricultural and Horticultural Machinery Plant, China) and the DBSSM was quantitatively collected and dried at 60 °C.

2.3. Optimization of experimental conditions by single factor investigation

An investigation into the effect of several operating parameters on the final yield (FY) of WSSO was conducted by following the procedure mentioned in the above section. The operating parameters investigated are summarized in Table 1.

The content of residual oil in the DBSSM was measured by the Soxhlet method. Three replicates were carried out. The FY of WSSO was calculated by using the following equation:

$$FY(\%) = (X_1\text{-}X_2) \div X_1 \times 100\%$$

Table 1. Summary of single factor experiments on operating parameters[a]

Baking time (min)[b]	Water added (ml/10 g)[c]	Baking temperature (°C)[d]	Agitation time (min)[e]	Particle size of seed slurry (μm)[f]	Aqueous salt solution added (ml/10 g)[g]	Concentration of aqueous salt solution (%; w/w)[h]	Agitation temperature (°C)[i]
1.00	0.00	90	15	180	1.40	3.00	25
2.00	0.50	95	20	154	1.50	4.00	30
3.00	1.20	100	23	125	1.60	5.00	35
4.00	1.30	105	24	100	1.70	6.00	40
5.00	1.40	110	25	74	1.80	7.00	45
6.00	1.45	115	26	61	1.90	8.00	50
7.00	1.50	120	27	54	2.00	9.00	55
8.00	1.55	-	30	38	2.10	10.00	60
9.00	1.60	-	35	-	2.20	-	63
10.00	1.70	-	-	-	-	-	64
-	1.80	-	-	-	-	-	65
-	-	-	-	-	-	-	66
-	-	-	-	-	-	-	67
-	-	-	-	-	-	-	68
-	-	-	-	-	-	-	69
-	-	-	-	-	-	-	70

[a]For all experiments except for investigation into effect of baking temperature, the seeds were baked at 110 °C.
[b]Other parameters: the seed slurry was passed through a sieve with a 154 μm pore size (100 meshes), the addition of 1.55 ml H_2O and agitation at 65 °C for 25 min.
[c]Other parameters: the seeds were baked for 1 min, the seed slurry was passed through a sieve with 154 μm pore size (100 meshes), agitation at 65 °C for 25 min.
[d]Other parameters: the seeds were baked for 1 min, the seed slurry was passed through a sieve with154 μm pore size (100 meshes), the addition of 1.55 ml H_2O and agitation at 65 °C for 25 min.
[e]Other parameters: the seeds were baked for 1 min, the seed slurry was passed through a sieve having 154 μm pore size (100 meshes), the addition of 1.55 ml H_2O and agitation at room temperature (25 °C) until the aggregation of all the hydrophilic compounds and liberation of free oil was observed.
[f]Other parameters: the seeds were baked for 1 min, the addition of 1.55 ml H_2O as well as agitation for 25 min.
[g]Other parameters: the seed slurry was passed through a sieve with 154 μm pore size, the concentration of aqueous salt solution was 6.00% (w/w) as well as agitation at 65 °C for 25 min.
[h]Other parameters: the seeds were baked for 1 min, the seed slurry was passed through a sieve with 154 μm pore size, the addition of 2.00 ml aqueous salt solution (6.00%, w/w) as well as agitation at 65 °C.
[i]Other parameters: the seeds were baked for 1 min, the seed slurry was passed through a sieve with 154 μm pore size, the addition of 2.0 ml aqueous salt solution (6.00%, w/w) as well as agitation for 25 min.

In the equation, X_1 (g) represents the amount of crude oil in the white sesame seed slurry (WSSS) [10 g × crude oil fraction in WSSS (water free)], while X_2 (g) represents the amount of crude oil in DBSSM (amount of DBSSM (g, water free) × its residual oil fraction).

2.4. Response surface method to further optimize extraction conditions

A response surface experiment was conducted to check whether the optimum aqueous extraction conditions established by single factor experimentation was the best and to study the cross-effect of these conditions on the FY of WSSO. By centering on the optimum extraction conditions established by single factor experimentation, Box-Benhnken's central combined experimental design was implemented. Four extraction conditions including concentration of salt solution (A), amount of salt solution added (B), agitation temperature (C), and agitation time (D) were selected and assigned to three different levels for performing response surface experiments.

2.5. Extraction of oil by hexane

Sesame seeds were baked to constant weight and crushed to pass a sieve with 154 µm pore size. The oil in the crushed seed was extracted for 6 h by n-hexane using a Soxhlet extraction apparatus. After n-hexane was completely removed by rotary evaporation, and the oil was vacuum-dried to constant weight at 50 °C.

2.6. Production of oil by cold spiral pressing

The extraction of WSSO by cold spiral pressing was also conducted for comparison. The residual oil content in DBSSM obtained by this method was determined.

2.7. Analytical methods

Crude oil content, acid value, peroxide value, smell and taste, hexane content, and transparency were analyzed according to Chinese National Standards GB 5009.6-2016, GB 5009.229-2016, GB 5009.227-2016, GB/T 5525-2008, GB/T 5009.37, respectively. Color was analyzed using a colorimeter (HanterLab UltraScan Pro. USA) (Gerde *et al.*, 2007). Crude protein content was measured according to the Chinese National Standard GB 5009.5-2016.

2.8. Statistics analysis

Experimental data were analyzed using one-way analysis of variance. Significant difference between pair data was estimated by the Student's t-test. The P-value was calculated using Microsoft Office Excel. The response surface analysis was conducted by using Design Expert 8.0.6 software. A multifactorial regression equation was established using SAS V8.0.

3. RESULTS AND DISCUSSION

3.1. Contents of major nutrients in the raw material studied

The crude fat content of the white sesame seeds studied was 51.25% (dry weight basis) while their protein content was 23.90%. All data in terms of FY of WSSO were calculated on the basis of these measurements. This sample was used for developing the advanced aqueous method of extracting WSSO and producing DBSSM with high quality.

3.2. Optimization of experimental conditions by single factor investigation

Division 1 Effect of water added on the final oil yield. The effect of added water on the FY of WSSO from 10.00 g WSSS is shown in Figure 2. FY was only ca. 6% without the addition of water. Therefore, water is essential for oil recovery. The FY increased significantly to ca. 92% when the amount of water added increased from 0.00 to 1.55 ml. The formation of hydrogen bonds may be involved in the mechanism for separating WSSO by adding an appropriate amount of water. Free oil is released by grinding white sesame seeds into slurry. In this state, small solid particles consisting of proteins, carbohydrates, etc. are dispersed in a continuous oil phase. The hydrophobic groups on their surface are surrounded by oil with its hydrophilic groups facing the particle core because of hydrophobic interaction. The work of cohesion of oil or hydrophilic compounds into small solid particles is smaller than the work of their adhesion. Therefore, oil bound by the hydrophobic surface of the dispersed small solid particles cannot be separated by centrifugal force. When water is added to WSSS, it comes into contact with and binds to the hydrophilic groups of dispersed small solid particles facilitated by agitation. Water and all hydrophilic groups aggregate together so that a large, viscous and rigid particle is formed. At the same time, oil escapes from the surface of small solid particles and aggregates into the free oil phase. This is due to the fact that the addition of water increases the inter surface tension of oil and small solid particles. In this state, the work of cohesion of oils or hydrophilic compounds into small solid particles is greater than the work of their adhesion.

However, a further increase in the amount of water to higher than 1.55 ml gradually reduced the FY of WSSO. The reason for this may be that some

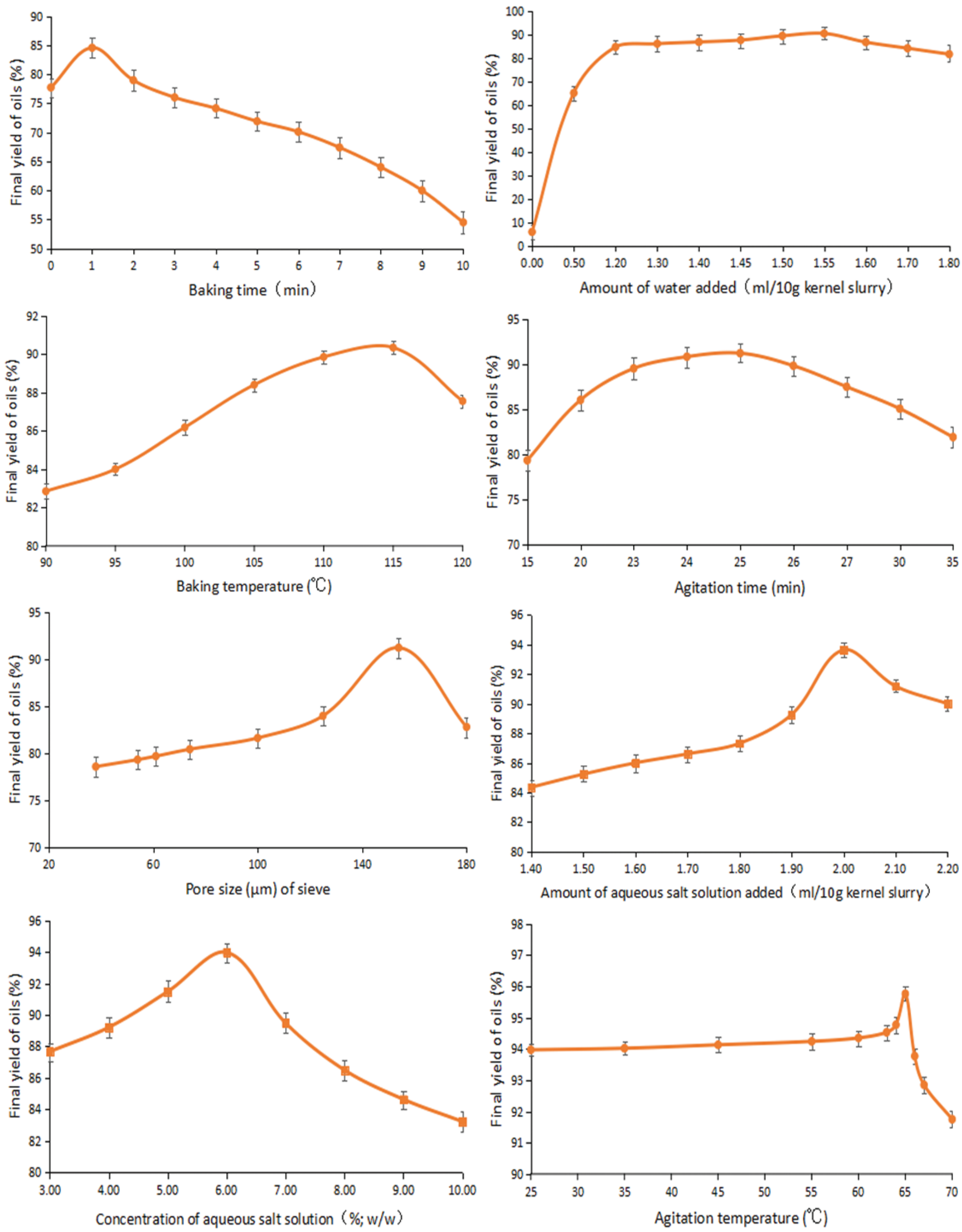

FIGURE 2. Effect of single factors on the final yield of white sesame oil.

hydrophilic compounds such as free fatty acids, phospholipids and phenolic compounds can dissolve in water so that the emulsification is enhanced and FY is reduced. Of all the added amounts of water, 1.55 ml gave the maximum FY of WSSO. This study demonstrates that adding the right amount of water is necessary for the effective extraction of WSSO.

Division 2 Effect of amount of aqueous salt solution on the final oil yield. Figure 2 shows the effect of the amount of aqueous salt solution added on the FY of WSSO. The FY of WSSO gradually increased when the amount of aqueous salt solution increased from 1.40 to 2.00 ml. However, the FY of WSSO dramatically decreased when the amount of aqueous salt solution further increased to be greater than 2.00 ml. Therefore, the trend of the effect of the aqueous salt solution added on the FY of WSSO is obviously different as compared to that of the pure water added. The amount (2.00 ml) of aqueous salt solution required to obtain the highest FY of oil was much higher compared to pure water (1.55 ml). Particularly, the highest FY (ca. 94%) of oil obtained by adding an aqueous salt solution was much higher compared to the one (ca. 92%) obtained by adding pure water.

Division 3 Effect of concentration of aqueous salt solution on the final oil yield. The effect of the concentration of aqueous salt solution on the FY of WSSO is shown in Figure 2. The FY of WSSO gradually increased as the concentration of aqueous salt solution increased from 3.00 to 6.00% (w/w). The mechanism of increase in the FY of WSSO by adding salt may be as follows: The addition of salt may alter the surface tension and surface charge of such compounds as proteins or carbohydrates so that the polarity of their hydrophilic groups is increased. Therefore, the non-covalent interaction of water with polymers by the action of ionic bonds is increased, and oil is subsequently extruded from the aggregated polymer. Also, water density may be increased by adding salt so that the density difference between supernatant oil and aqueous sediment is increased. However, further increasing the concentration of aqueous salt solution to > 6.00% (w/w) dramatically decreased the FY of WSSO. This may be due to decrease in available water for separating oil caused by adding too much salt.

Division 4 Effect of baking time on the final oil yield. The effect of baking time at 110 °C on the FY of WSSO is shown in Figure 2. An increase in baking time significantly increased the FY of WSSO. However, the baking time ≥ 2 min significantly reduced the FY of WSSOs. This shows that baking white sesame seeds for a certain time (1 min) is essential for obtaining a high oil FY. The reason for this result may be that lipase can be deactivated by proper baking so that the loss of neutral oil during the aqueous extraction of WSSO can be reduced. Furthermore, water can be removed by proper baking, which facilitates the destruction of cell wall or oil body and the release of oil by grinding. However, baking for too long (≥ 2 min) may denature proteins. The denaturation of proteins may increase their ability to bind to oil and thus reduce the FY of WSSO. Similar behaviors of rice protein have also been reported in the literature (Tabara *et al.*, 2015).

Division 5 Effect of baking temperature on the final oil yield. The effect of baking for 1 min at different temperatures on the FY of WSSO is shown in Figure 2. The FY of WSSO increased significantly as the baking temperature increased. However, when baking temperature increased to ≥ 115 °C, the FY of WSSO gradually decreased. The reason for this result may be similar to the reason described in the above section.

Division 6 Effect of agitation time on the final oil yield. The effect of agitation time on the FY of WSSO is shown in Figure 2. The increased agitation time from 15 to 25 min significantly increased the FY of WSSO. However, further prolonged agitation time significantly decreased the FY of WSSO. This may be caused by prolonged heating time (see "*Division 4*", "*Division 5*" and "*Division 7*"). Agitating for 25 min should be reasonable.

Division 7 Effect of agitation temperature on the final oil yield. The influence of agitation temperature on the FY of WSSO is shown in Figure 2. The FY of the WSSO increased significantly as the agitation temperature rose from 25 to 65 °C. However, the FY of WSSO reduced greatly as the agitation temperature further increased from 65 to 70 °C. Among all the agitation temperatures studied, 65 °C gave the highest FY of WSSO. The reason for these results may be as follows: An increase in temperature reduced the viscosity of the oil, which is advantageous to the separation of oil. The reduced viscosity of oil facilitated its aggregation and escape from the retention of solid particles. Increases in the intense molecular motion of the oil and hydrophilic compounds at an adequately high temperature promoted the aggregation of oily phases or of hydrophilic compounds. The mechanism for reducing the final oil yield at a temperature of > 65 °C may be similar to the one described above in "*Division 4*".

Division 8 Effect of particle size of white sesame seed slurry on the final oil yield. The effect of particle size on the FY of WSSO is shown in Figure 2. The FY of WSSO dramatically increased when the pore size of the sieve through which the slurry passed decreased from 180 to 154 μm. This indicates that grinding white sesame seeds to a certain extent (154 μm) is essential for the decomposition of the oil body. The reduction in the solid particle size of the WSSS can increase its contact surface with water and therefore facilitate the extraction of oil. However, as the solid particle size further decreased to < 154 μm, the FY of WSSO significantly decreased. A solid particle which is too fine may result in an increase in the hydrophobic groups which are exposed to the surface of solid particles so that the FY of WSSO is decreased.

Division 9 Final yield of white sesame oil from the aqueous method as finally established by single-factor experimentation. The single factor experiments described in the above sections established a new aqueous method for extracting WSSO which was very different from traditional aqueous methods which use large quantities of water. The exact operating procedure and parameters of this new aqueous method were as follows: The white sesame seed with peel was baked at 115 °C for 1 min. The baked seed was ground into continuous slurry by passing it through a sieve of 154 μm pore size. An aqueous salt solution (2.00 ml; 6.00% (w/w)) was added to 10.00 g seed slurry in a 20 ml centrifuge tube. Then, the mixture of the salt solution and the seed slurry was agitated for 25 min at 65 °C. Finally, free oil was collected by centrifugation three times at 4000 r/min for 30 min, followed by one cold screw pressing (Ai Bang Agricultural and Horticultural Machinery Plant, China). The DBSSM was quantitatively collected and dried at 60 °C. By using this new aqueous method, the FY of WSSO obtained was as high as 95.76%.

3.3. Response surface method to further optimize aqueous extraction conditions

The design and results from the response surface experiment and the analysis of variance are shown in Tables 2 and 4. The experimental results

TABLE 2. Design and results of surface response experimentation

Run	Factor 1 A: Concentration oncentration of aqueous salt solution (%)	Factor 2 B: Amount of aqueous salt solution added (ml)	Factor 3 C: Agitation temperature (°C)	Factor 4 D: Agitation time (min)	Response Final oil yield (%)
1	6.00	1.95	66.00	25.00	96.04
2	6.00	2.05	66.00	25.00	94.17
3	6.50	2.00	65.00	26.00	91.58
4	6.00	2.05	65.00	26.00	94.51
5	6.00	2.00	64.00	24.00	95.59
6	6.00	2.00	65.00	25.00	95.71
7	6.00	2.00	64.00	26.00	95.68
8	6.00	2.00	65.00	25.00	95.89
9	6.00	2.00	66.00	24.00	95.69
10	5.50	1.95	65.00	25.00	94.63
11	6.00	2.00	65.00	25.00	95.98
12	6.00	1.95	64.00	25.00	96.06
13	5.50	2.00	65.00	24.00	94.14
14	6.00	2.00	65.00	25.00	95.98
15	6.00	2.05	65.00	24.00	94.81
16	6.00	1.95	65.00	26.00	96.03
17	6.00	2.00	66.00	26.00	95.68
18	6.00	2.05	64.00	25.00	94.72
19	6.50	2.00	66.00	25.00	91.49
20	5.50	2.00	65.00	26.00	94.32
21	5.50	2.00	64.00	25.00	93.98
22	6.00	1.95	65.00	24.00	96.06
23	6.50	2.00	65.00	24.00	91.16
24	6.50	2.00	64.00	25.00	91.71
25	5.50	2.05	65.00	25.00	93.04
26	6.50	1.95	65.00	25.00	92.36
27	5.50	2.00	66.00	25.00	93.55
28	6.50	2.05	65.00	25.00	90.01
29	6.00	2.00	65.00	25.00	95.76

TABLE 3. ANOVA for Response Surface Quadratic Model Analysis of variance table [Partial sum of squares - Type III]

Source	Sum of Squares	df	Mean Square	F Value	P-Value Prob > F	
Model	88.03	14	6.29	124.59	< 0.0001	significant
A-Concentration of aqueous salt solution	19.64	1	19.64	389.07	< 0.0001	-
B-Amount of aqueous salt solution added	8.20	1	8.20	162.49	< 0.0001	-
C-Agitation temperature	0.10	1	0.10	2.07	0.1721	-
D-Agitation time	0.010	1	0.010	0.20	0.6598	-
AB	0.14	1	0.14	2.86	0.1129	-
AC	0.011	1	0.011	0.22	0.6474	-
AD	0.014	1	0.014	0.29	0.6016	-
BC	0.070	1	0.070	1.39	0.2578	-
BD	0.018	1	0.018	0.36	0.5575	-
CD	2.500E-003	1	2.500E-003	0.050	0.8271	-
A2	57.52	1	57.52	1139.72	< 0.0001	-
B2	1.14	1	1.14	22.57	0.0003	-
C2	0.21	1	0.21	4.12	0.0618	-
D2	0.030	1	0.030	0.59	0.4547	-
Residual	0.71	14	0.050			-
Lack of Fit	0.64	10	0.064	4.15	0.0913	not significant
Pure Error	0.062	4	0.016	-	-	-
Cor Total	88.74	28		-	-	-

Note: "*" indicates significant, $p < 0.05$; "**" indicates extremely significant, $p < 0.01$; "-" indicates not significant, $p > 0.05$; R^2=0.9920, R^2Adj=0.9841.
The Model F-value of 124.59 implies the model is significant. There is only a 0.01% chance that a "Model F-Value" this large could occur due to noise.
Values of "Prob > F" less than 0.0500 indicate model terms are significant.
In this case A, B, A++2+-, B++2+- are significant model terms.
Values greater than 0.1000 indicate the model terms are not significant.
If there are many insignificant model terms (not counting those required to support hierarchy), model reduction may improve the model.
The "Lack of Fit F-value" of 4.15 implies there is a 9.13% chance that a "Lack of Fit F-value" this large could occur due to noise. Lack of fit is bad -- we want the model to fit. This relatively low probability (<10%) is troubling.

TABLE 4. Production efficiency of white sesame oil by the advanced aqueous method and other methods as well as the characteristics of the oil produced as compared with Chinese National Standard (CNS; GB1536-2004) for 1st class refined oil (Only bold items are mandatory while others are not.)

Items	CNS	Aqueous	Enzyme-assisted aqueous	Hexane	Cold pressing
Extraction yield of oils (%)	—	96.02	69.33[a]	99.03	74.43
Smell, taste[b]	b	b	—	b	b
Transparency	C, T[c]	C, T[c]	—	C, T[c]	C, T[c]
AV (mg KOH/g)	**0.6**	0.19	—	1.93	1.60
PV (mmol/kg)	**6.0**	3.25	—	5.74	5.13
Residual hexane (mg/kg)	**ND[d]**	ND[d]	ND[d]	50	ND[d]
Residual oil content in de-oiled meal (%)	—	3.98	—	1.01	21.37
Final yield of oils (%)	—	96.02	65.71[e]	95.18	72.26

[a]The highest extraction yield of white sesame oil by enzyme-assisted aqueous method reported in the literature (Wang *et al*., 2015); [b]Having the inherent smell and taste of sesame oil, no adverse smell; [c]C,T-Clarify, transparent; [d]ND- not detectable.

indicated that the concentration of aqueous salt solution (A) and amount of aqueous salt solution added (B) significantly affected the FY of WSSO ($p < 0.01$). The cross-effect of these four factors (AB, AC, AD, BC, BD and CD) on the FY of WSSO was not significant ($p > 0.05$). The results from the response surface analysis using Design Expert 8.0.6 software are shown in Figures 3-5. These figures show similar results with respect to the effect of variables and their cross-effect on the FY of WSSO.

A regression equation was obtained by using the SAS RSREG program to perform regression fitting on response values and various factors. The final equation established in terms of coded factors was as follows:

$$\text{FY of WSSO} = + 95.86 - 1.28*A - 0.83*B - 0.093*C + 0.029*D - 0.19*A*B + 0.053*A*C + 0.060*A*D - 0.13*B*C - 0.067*B*D - 0.025*C*D - 2.98*A^2 - 0.42*B^2 - 0.18*C^2 - 0.068*D^2$$

In the equation, A represents the concentration of the aqueous salt solution; B represents the amount of aqueous salt solution added; C represents agitation temperature; D represents agitation time. The optimum combination of operating parameters for obtaining the highest FY of WSSO was as follows: A = 6.00% (w/w), B = 1.95 ml, C = 64 °C, and D = 25 min. The highest FY of WSSO obtained by using these optimum operating parameters was 96.02%, which was not significantly different from that (95.76%) obtained from using those established by the single factor experiment.

Therefore, the advanced aqueous method for extracting WSSO followed the same procedure as the one described in "*Division 9*." except for the fact that 1.95 ml aqueous salt solution instead of 2.00 ml were used. This method resulted in a much higher FY of WSSO as compared to direct cold pressing or the enzyme-assisted aqueous method using large quantities of water (Table 4). Although a FY of WSSO obtained by enzyme-assisted aqueous method of 87.58% has been reported, the authors

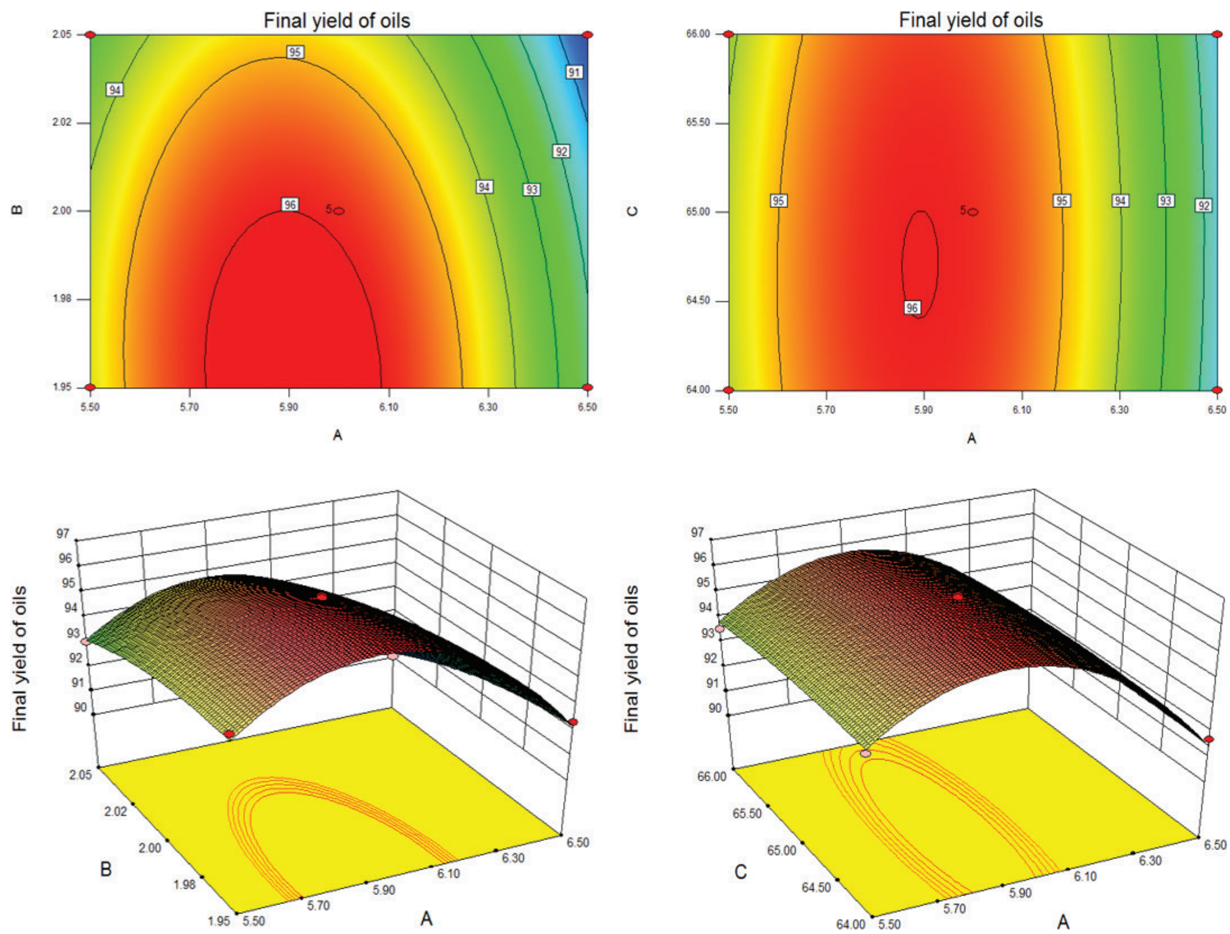

FIGURE 3. Response surface interactions of AB and AC. A-Concentration of aqueous salt solution, B-Amount of aqueous salt solution added and C-Agitation temperature.

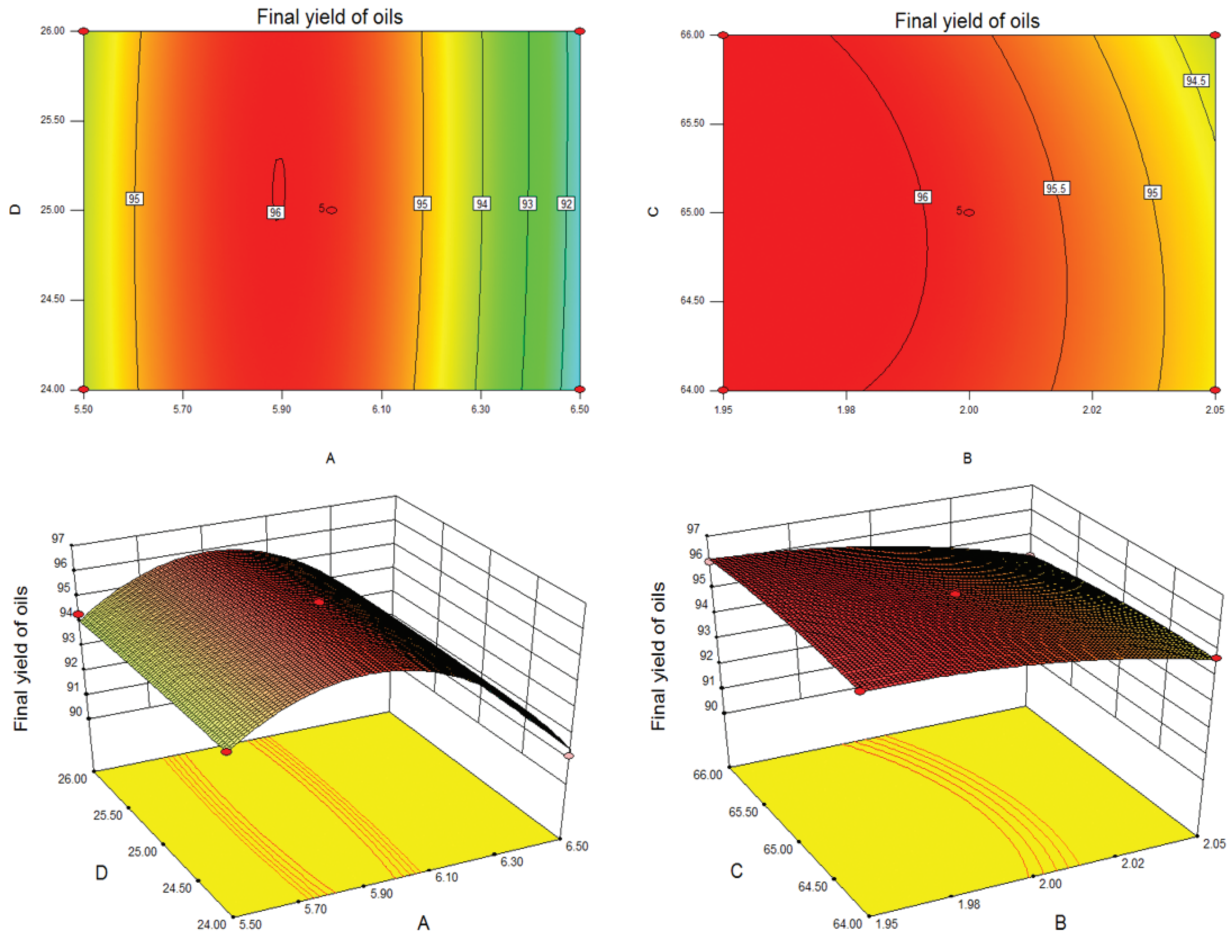

FIGURE 4. Response surface interactions of AD or BC. A-Concentration of aqueous salt solution, B-Amount of aqueous salt solution added, C-Agitation temperature and D-Agitation time.

did not specify whether the study material was white or black sesame seed (Hou *et al.*, 2013). Although the aqueous method developed in this study produced a slightly lower extraction rate of WSSO, it's FY of WSSO was comparable to that of hexane extraction. The reason for this may likely be that the oil produced by hexane extraction cannot be consumed directly and should therefore be refined. The refining process of crude oil can lead to a large amount of neutral oil loss (Stalker and Wilson, 2015).

3.4. Quality of white sesame oil and de-oiled meal produced by the new aqueous extraction method

The WSSO produced by the advanced aqueous method finally established in this study was clear and transparent. Table 4 indicates the chemical and physical indices of the oils produced. The acid value (AV, 0.19 mg KOH/g) and the peroxide value (PV, 3.25 mmol/kg) produced by the advanced aqueous method were better than the ones required by the Chinese national standard for first class edible sesame oil and that of the oil produced by hexane extraction or cold pressing.

The color indices for the oil produced by the advanced aqueous method were as the follows: L* = 57.76, a* = -5.68, and b* = 86.01; while those produced by hexane extraction were L* = 40.63, a* = -12.16, and b* = 51.79; and those obtained from direct cold pressing were L* =60.67, a* =15.97, and b* =97.44. The oil obtained by the advanced aqueous method had a lighter color than the oil produced by hexane extraction.

The DBSSM obtained from the advanced aqueous method showed 45.52% protein content and 3.98% residual oil content. Although the color of DBSSM from the advanced aqueous method was slightly deeper than that obtained by hexane extraction, it may be directly applied to the food industry or food recipes since whole sesame seeds with their peel are edible.

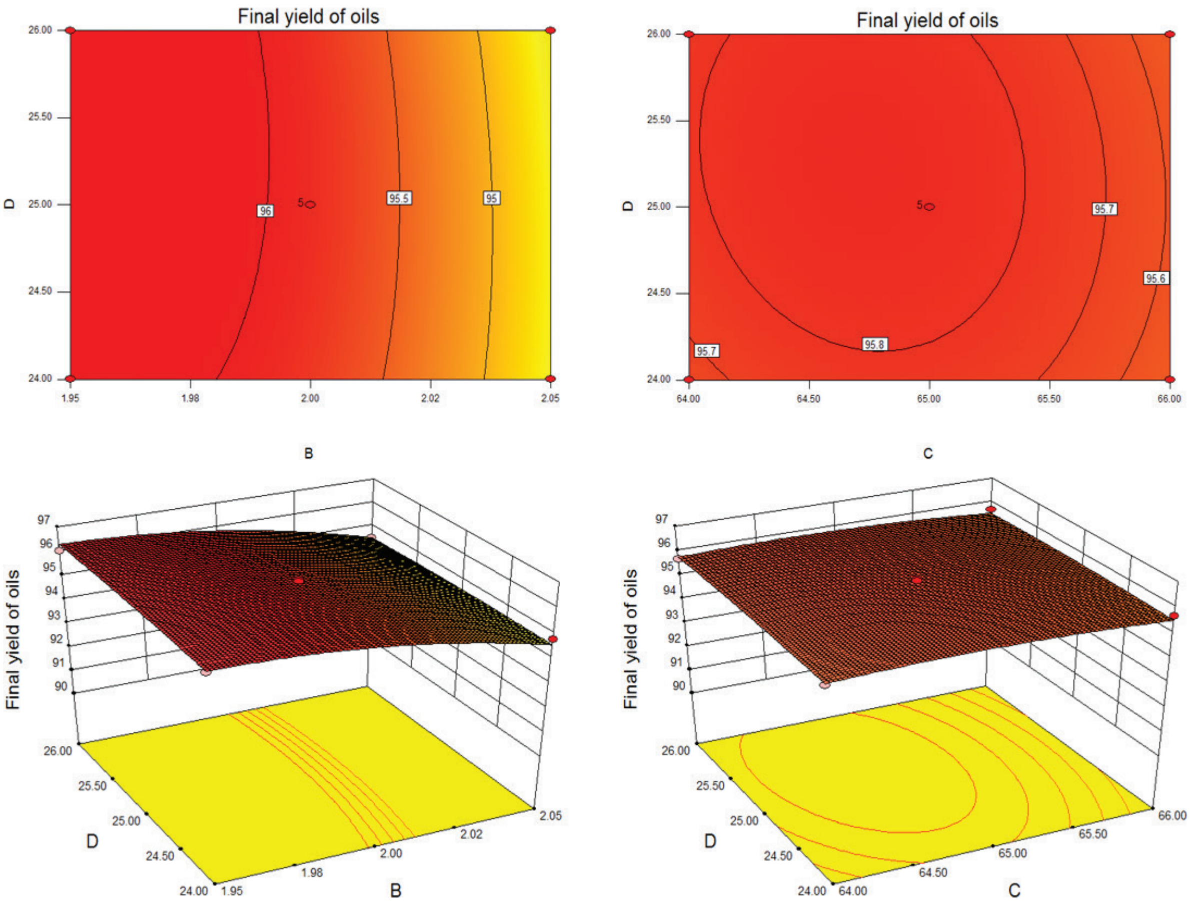

FIGURE 5. Response surface interactions of BD and CD. B-Amount of aqueous salt solution added, C-Agitation temperature and D-Agitation time.

3.5. Advancement of the new aqueous method established in this study compared to the traditional aqueous method

The traditional aqueous method using large quantities of water (usually liquid:raw material ratio > 2) relies on the solubilization of hydrophilic compounds such as proteins, free fatty acids, or free amino acids and the formation of emulsion for the efficient extraction of oil. By this method, it is difficult to obtain clear free oil because of serious emulsion though a high extraction rate can be achieved. Therefore, the recovery rate of oil from sesame seeds is quite low, i.e. less than 90%. Another disadvantage of this method is that it produces a large quantity of waste water, causes a loss in water soluble compounds and difficulty in drying the de-oiled residue.

The aqueous method established in this study uses a small amount of water (liquid:raw material ratio being only1.95:10). Our method is based on a new theory of separating oil. Its mechanism is associated with the work of cohesion of oil or hydrophilic compounds as well as the work of adhesion of oil to solid particles containing proteins, saccharides, free amino acids and free fatty acids via a hydrophobic interaction. The aggregation of hydrophilic groups of compounds such as proteins, saccharides, free amino acids, free fatty acids and phospholipids (with or without hydrophobic groups) through hydrogen bonds is promoted by adding the proper amount of water which can avoid their solubilization or dispersion as a prerequisite. This process results in the work of cohesion becoming larger than the work of adhesion so that release of free oil is carried out. Therefore, the mechanism of this new aqueous method is completely different from that of traditional aqueous method using large amounts of water and relying on the solubilization or dispersion of hydrophilic compounds such as proteins, saccharides, free amino acids, free fatty acids and phospholipids.

The recovery rate of oil from white sesame seeds is as high as 96.06%. No waste water is produced during the extraction of the oil. The water content of the de-oiled meal is quite low so that it can be easily dried. Full utilization of white sesame seeds is achieved.

4. CONCLUSIONS

It was concluded that the advanced aqueous method eventually developed in this study recovered 96.02% of WSSO. The acid value at 0.19 mg KOH/kg and peroxide value at 3.25 mmol/kg were quite low, and better than that required by the Chinese national standard for first-class edible sesame oil and that of oil produced by hexane extraction. The oil obtained from the advanced aqueous method had a lighter color than the oil produced by hexane extraction. The residual oil content in the DBSSM was 3.98%, while its protein content was 45.52%. No wastewater was discharged during the aqueous extraction of oil. The results of this study should provide valuable scientific guidance for the future development of production technology of high-quality WSSO and DBSSM with high-quality and low cost on an industrial scale.

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