# EFFECT OF USING SOME RICE BY-PRODUCT ON THE PHYSICOCHEMICAL PROPERTIES OF HEATED SUNFLOWER OIL DURING DEEP FRYING

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#### **ABSTRACT**

Rice hulls are inexpensive silica-rich by-product of the cereal industry. They are capable of binding lutein from soybean oil in hexane at room temperature. The objective of this study was to use rice hulls (RH) and rice straw (RS) as adsorbents for improving the physicochemical properties of heated sunflower oil for 36 h during deep fat frying of potato chips at 180° C. Results revealed that physicochemical parameters of the heated oil were markedly improved due to the use of RH and RS. Color was improved by 11.43, 56.94, 61.81 and 41.72& with RHB white improved by 9.35, 50.99, 53,21, 48.92 and 33.65% with RSB at the same heating time of 8, 16, 24, 32 and 36 h, respectively. The free fatty acids improved by 42.79, 49.21, 64.18, 47.19 and 40.59 with RHB and improved by 37.83, 42.45, 51.31, 40.16 and 32.09% with RSB. The peroxide value improved by 46.15, 63.64, 69.29, 46.29 and 14.11 with RHB, and also improved by 30.77, 56.82, 56.14, 31.48 and 14.04% with RSB. Total polar compounds of heated sunflower oil improved by 4.97, 61.79, 72.91, 59.72 and 23.84% with RHB and improved by 3.58, 55.39, 69.31, 49.39 and 14.74% with RSBat the same heated time of 8, 16, 24, 32 and 36 h, respectively. The results obtained indicated that treatments with RHB and RSB could improve oil quality as measured by changes in color, FFAs, PV and TPC. Both adsorbent materials were most effective when used in the early stages of deterioration. Results also revealed that while the initial treatments with RHB and RSB improved the oil quality up to 24 h, subsequent frying showed that useful life of the oil was not markedly extended.

#### INTRODUCTION

Deep-fat frying is one of the oldest and commonly used procedures for the cooking of foods (Valera,1988). This is due to its convenience, rapidity, economy, and the appealing flavour, odour, and texture of fried foods (Liu and White, 1992).

As frying continuos, the concentration of degradated products gradually increases until the oil is unfit for use. At the same time, the quality of the fried decreases and become unacceptable for human consumption. It is common practice in restaurants to filter oil used for deep-fat frying to remove food debris, which may accelerate the decomposition of the oil. However, filtration alone doesn't remove oil soluble degradated products. Simultaneous removal of particulate by filtration and soluble imparities by adsorption is desirable as it will extend the life of the oil and provide higher quality fried foods. Several materials have been reported to adsorb the polar degradated heated oil. Among these are synthetic magnesium silicate (Allen, 1978), synthetic calcium silicate (Artman, et al., 1988), aluminum oxides diatomaceous earth, bleaching earth (Artman, 1969), activated carbon (Bailly,

1950; Bast, et al., 1991) and various forms of silica (Billek, et al., 1978; and Bracco, et al., 1981).

It was estimated that the total consumed vegetable oils in Egypt in 1997 were 950 thousand tons (Agric. Economic, 1998). If even 5% of this oil were wasted owing to improper frying techniques or because of faulty knowledge of when the oil should be discarded, the total loss could reach about 190 million L.E. Recently, the waste estimated were placed at 25-30%, which could extrapolate these monetary losses into millions of L.E. (Hunter and Applewhite 1992). Rice hull ash can adsorb the soy oil carotenoid lutien (Proctor and Palaniappan, 1989) and free fatty acids from soy oil miscall under ambient conditions (Proctor and Palaniappan, 1990). Rice hull ash could be invaluable in third-world economies where energy and mineral resources are limited, and it could also be a useful supplement to montmorillonite clays in the developed world.

The present study, aimed to investigate the effect of heating time on the quality of sunflower oil during deep-fat frying of potato chips at 180°C for 36h, effect of using some natural waste by products (rice hull and rice straw) as adsorbing materials for regeneration the used sunflower oil in frying process and study the oil quality of the regenrated sunflower oil.

# MATERIALS AND METHODS

#### **Materials**

Oils

Refined Sunflower oil (a total of 30 L) was obtained from Alexandria Oils and Soap Company, Alexandria, Egypt during the year of 2000.

#### **Potatoes**

Potatoes (Solanum tuberosum) (50 Kg) were purchased from the local market of Alexandria city, Egypt.

#### Chemical materials

Silica gel (SG), Citric acid (CA) were purchased from (Sigma, Co. U.S.A.) and Carboxy Methyl Cellulose (CMC) was Sodium salt from BDH chemical, England.

### Experimental procedures

#### Preparation of rice hulls and rice straw:

Rice hull was collected from the Rice Mills (Alexandria Mills Company, Alexandria), while the rice straw was collected from the Faculty Farm (Faculty of Agriculture, Saba-Basha, University of Alexandria). The rice hulls and rice straw were, separately, prepared according to Barbary et al., (1999). Therefore, two blends of rice hulls or rice straw were prepared containing 0.5% (CA), 10% (SG) and 10% (CMC) and assigned as rice hull blend (RHB) and rice straw blend (RSB), respectively. Different levels of RHB or RSB in terms of 5; 7.5; 10 and 15% of oil weight were also prepared and examined separately on the used heated sunflower oil. The preliminary investigation revealed that RHB or RSB in level of 10% oil weight showed the

best result with the heated oil. This blend (10%), therefore, was further used with heated oil as filtering material (adsorbent). These two blends were added to the heated oil at 180°C for 5 min stirring time followed by filtration process.

#### Preparation of potato chips

Potatoes were peeled, cut into 2 mm thick slices using a rotary slicer (Edelstahl, Rostfrel, England), washed and dewatered before to frying.

#### Frying performance

Frying performance was carried out according to Barbary *et al.*, (1999) using the deep-fat fryers (Moulinex, France). Samples of potato chips (200 g) were fried at 180°C for 10 min every an hour up to 36 h. RHB or RSB as filtering powders (10 % of oil weight) was added to the whole oil at 180°C for 5 min after (8; 16; 24; 32; and 36 h of heating), then filtered with Wattman paper No (1) under pressure.

Oil samples (50 g) were collected every 8 hours before and after adding the RHB or RSB and analyzed for their physicochemical characteristics.

# Physicochemical analysis of heated oils Colour index

Color index was determined according to official methods (1989) (Method Cc Bc-50) using spectrophotometer (Safos Moraco, 1900) and absorbance was read at the following wavelengths, 460; 550; 620; and 670 nm. The spectrophotometer colour index was calculated from the following equation:

Colour index = 1.29 (A<sub>460</sub>) + 69.7 (A<sub>550</sub>) + 41.2 (A<sub>620</sub>) - 56.4 (A<sub>670</sub>).

#### Peroxide value (PV)

Peroxide value (PV) was determined according to AOCS official methods (1989) (Method Cd 8–53) by titration with standard sodium thiosuphate (0.1 N) and was calculated as millequivalent peroxides per kilogram oil (meq/Kg oil).

# Acidity (free fatty acids)

The free fatty acid content was determined by titration with standard potassium hydroxide solution 0.1 N and calculated as oleic acid percentage according to the AOCS official methods (1984) (Method CD3a – 94).

# Total polar compounds (TPC)

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Total polar compounds were determined according to Christie (1989). Accurately weighed sample of 100 mg oil was dissolved in 2 ml hexane / diethyether, 90: 10 (v/v). The non-polar fraction was eluted with 30 ml of methanol. All solvents were rotary evaporated, the residue was weighed and the percentages of the total polar compounds (TPC) were determined (Romero et al., 1995).

#### Separation of polar and non-polar fraction from the filter cake

Samples of filter cakes (RHB and RSB) were separately placed in Soxhlet extractors and serially extracted with increasingly polar organic solvents to remove both adsorbed oil and polar compounds according to Yates and Caldwell (1993). Extraction # 1 was petroleum ether to remove the oil that coated the particles or was trapped in the pores and spaces between particles. Extraction # 2 w as diethyl ether, whereas the extraction # 3 w as methanol (progressively polar solvents). The final extraction # 4 was methanol with sodium hydroxide (0.1N. NaOH) these solvents were intended to remove any polar compounds adsorbed at the active filter cake. All solvents were carefully removed by evaporation on a steam bath.

#### Analysis of fatty acid methyl esters using GC

Fatty acid methyl esters (FAMEs ) were performed according to the procedure of Radwan (1978).

Analysis of fatty acid methyl ester (FAMEs) was carried out using a gas chromatography (GC) (Shimadzo GC - 4CM - PFE) equipped with stainless steel columns with 3 % diethylene glycol succinate on chromosorb w 80/100 and flame ionization detector (FID). The oven and detector temperatures were 180-°C isothermal and 270°C, respectively. The carrier gas was nitrogen at a flow rate of 20 ml/min.

## **RESULTS AND DISCUSSION**

# Physicochemical properties of heated oil:

#### 1. Colour Index

Figure (1) shows the photometeric colour indices (PCI) of heated oil and effects of using the (RHB) or (RSB) as filtering powders (FP) on PCI of sunflower oil during deep-fat frying of potato chips at 180°C for 36 h. Data revealed that the PCI increased (oil deteriorated) as the heating time continued for 36 h. The PCI increased from 9.85 for the unused oil (fresh oil) to 93.21 for that heated for 36 h. Using the RHB and RSB during the corresponding heating time for 36 h markedly decreased the PCI of the heated sunflower oil. The PCI decreased from 22.63; 42.11; 60.46; 91.37; and 93.21 for heated oil to 20.93; 23.74; 29.18; 46.19; and 58.43 after using the RHB and to 21.19; 25.66; 33.53; 51.49; and 65.16 after using the RSB after 8, 16, 24, 32, and 36 h of heating, respectively.

The pigment removal percentages due to using the RHB and RSB increased as the heating time continued up to 24 h then decreased during the following heating for 36 h. The pigment removal percentages increased from 6.39 % after 8 h to 43.62; 51.74 % after 16 and 24 h with using the RHB then declined to 49.45 and 37.31 % after 32 and 36 h, respectively. Using the RSB showed less pigment removal percentages as compared with the RHB at any corresponding heating time. These results suggest that both RHB and RSB are more effective in removing the pigments from the heated oil during the first 24 h then become less effective during the following heating time.

Data revealed that the colour of the heated sunflower oil was improved by 11.43; 56.94; 61.81; 55.42 and 41.72 % with RHB after 8; 16; 24; 32; and 36 h of heating. However, it improved by 9.35; 50.99; 53.21; 48.92 and 33.65 % with RSB at the same heating time, respectively. These results are in agreement with those found by Mc Neil *et al.*, 1986, Yoon *et al.*, 1987, Mazza and Qi, 1992 and Xu *et al.*, 1999.

## 2. Acidity (FFAs)

The percentage of free fatty acids (FFAs%) as a function of frying time is shown in Figure (2). The concentration of the FFAs exponentially increased from 0.246 % for the unused oil to 2.256 % after heating for 36 h. The steady rise in the formation of FFAs can be attributed partially to the hydrolysis and partially to the component carboxylic groups present in the polymeric products of frying (Engleson, 1997). The highest rate of FFAs formation, as the other parameters, occurred during the first 8 h of heating. The rate of formation increased by 171.95 % during the first eight hours then retarded to 31.84 %; during the second eight hours, then increased to 57.6 % during the third eight hours; followed by a decrease of 38.27% during the fourth eight hours and finally to 17.38 % during the last four hours. Certainly, it is true that some of the FFAs are volatile (Melton, et al., 1994). Therefore, during frying or heating, the FFAs are degraded to more volatile compounds and lost or react to form nonvolatile compounds. Using high frying temperature as 180C, however, exacerbated these breakdown reactions; hence the process was irreversible and had a cascading effect (Lake and Scholes, 1997). Therefore, whether or not FFAs levels are indicative of frying oil degradation, they are related to fried food quality most likely depending upon the type of oil / fat used for frying (Melton et al., 1994).

The formation of the FFAs % of heated oil, however, decreased due to addition of FP (RHB or RSB) as compared to the heated oil. The FFAs % decreased to 0.488; 0.569; 0.688; 1.131 and 1.440 %, respectively, with RHB and to 0.509; 0.612; 0.803; 1.249 and 1.611 %, respectively with RSB after 8; 16; 24; 32; and 36 h of heating as compared to the untreated oil. Results also revealed that the heated oil was improved by 42.79; 49.21; 64.18; 47.19; and 40.59 % with the RHB and improved by 37.83; 42.45; 51.31; 40.16 and 32.09 % with RSB after 8; 16; 24; 32; and 36 h of heating, respectively.

#### 3. Peroxide Value (PV)

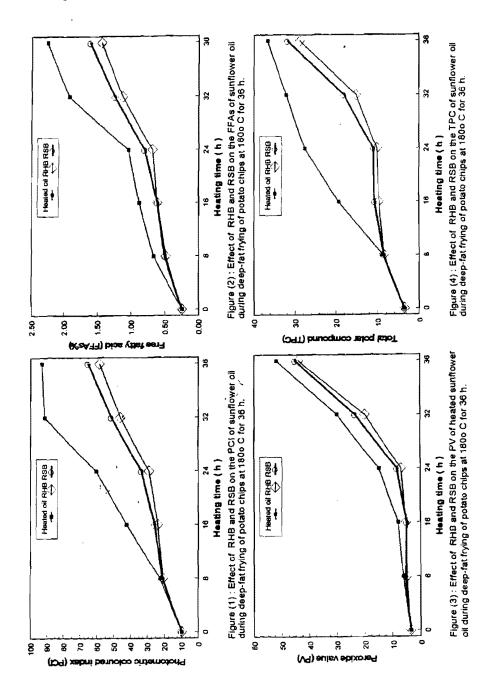
Figure (3) shows the peroxide value (PV) of heated oil and the effect of RHB and RSB on the PV of sunflower oil during deep-fat frying of potato chips at 180°C for 36 h. Data showed that the PV increased as the heating time increased up to 36 h. The PV increased from 3.6 (control) to 52.0 after heating for 36 h. Data revealed that the PV of the heated oil had improved by 46.15; 63.64; 69.29; 46.29 and 14.11 %, respectively with RHB and improved by 30.77; 56.82; 56.14; 31.48 and 14.04 % respectively, when RSB after 8; 16; 24; 32; and 36 heating, respectively. Similar results were obtained by Engelson, (1997). The PV is a measure for autoxidation of oils. However, this measure is deceptive in heated oils that contain polyunsaturated fatty acids (PUFA) (Tyagi and Vasishtha, 1996). In PUFA – rich oils, the formation of

peroxides takes place throughout the oxidation of free radicals. These peroxides decompose much faster by, means of labile hydrogen, which causes free radical polymerization. The higher the temperature of the deep – frying, the stronger the tendency for the decomposition.

# 4. Total polar compounds (TPC)

Figure (4) shows TPC % of heated oil and the effect of RHB and RSB on the TPC of sunflower oil during deep-fat frying of potato chips at 180°C for 36 h. Data revealed that the TPC increased as the heating time increased up to 36 h. The TPC increased from 3.76 for fresh oil to 8.79; 19.36; 27.68; 32.21 and 36.73 % after heating for 8, 16, 24, 32 and 36 h, respectively. Similar results were obtained by Cuesta et al., (1993) on sunflower oil when it was used to fry potatoes 75 times. The TPC increased in quantity during frying oil and defined as the sum total of the materials that were not triglycerides (TG) (Blumenthal, 1991). Therefore, TPC in the unused frying oil included tocopherols, mono-and diglycerides, FFAs and other oil-soluble components that are more polar than triglycerides. However, the TPC in used frying oil are composed of the total breakdown products from the process (Lake and Scholes, 1997). Several workers believed that at a concentration of 25.27 % TPC caused frying oil to deteriorate and should be discarded (Lake and Scholes, 1997). Levels and composition of TPC, however, is still a subject of controversy. Melton, et al (1994) reported that the TPC had not been related to the life of a frying oil but the resulting quality of fried food for enough oil / fat was the recommendation for discarding. They added that the level of TPC might be dependent not only on use history but also on the composition of the frying oil. Lumley, (1988) reported that the more unsaturated the oil, the greater the tendency to from polymeric rather than polar degradation products. Perhaps certain polar components were better related to oil degradation and quality of fried food than TPC. On the other hand, others (Lumlay, 1988; Pokorny, 1989 and Firestone, et al., 1991) believed that the concentration of TPC was related to frying oil life. They reported that for unused oil and induction period, typically heated at 180° C (2 %< TPC < 7 %); for optimum frying and flavor development, depending upon the food fried (7 % < TPC < 20 %); for deteriorated and rejected oil (TPC > 25 - 27 %) were expected.

The present results revealed that the formation of the TPC in heated oil, however, decreased by using either RHB or RSB as compared to the untreated oil. The TPC% decreased to 8.54, 9.72, 10.24, 15.22 and 28.87 %, respectively, using the RHB. While with RSB, the TPC % decreased to 8.61; 10.72, 11.10, 18.16 and 31.87 %, at the same heating time, respectively, as compared to the corresponding TPC of the heated oil. Data, also, revealed that the heated sunflower oil had improved by 4.97, 61.79, 72.91, 59.72 and 23.84 %, respectively with using RHB and improved by 3.58, 55.39, 69.31, 49.39 and 14.74 % respectively, using RSB after 8 16 24 32 and 36 h of heating. Some possible causes of these adverse effects could be due to increased pH, soap formation and Millard reaction products.



#### Effect of heating time on sunflower oil fatty acids composition

Relative percentages of fatty acids composition of sunflower oil continuously heated for 36 h during deep-fat frying of potato chips is shown in Table (1). Data revealed that the fatty acids composition of the fresh oil (1.53 %  $C_{14:0}$ , 8.82 %  $C_{16:0}$ , 4.63 %  $C_{18:0}$  and 0.84 %  $C_{20:0}$ ). The total saturation was 15.82 %. Total monoenes of 22.63 % in terms of 0.04 %  $C_{16:1}$  and 22.59 %  $C_{18:1}$ . Dienes fatty acid in terms of linoleic acid (C18:2) represents (61.55 %) the major component of the sunflower oil.

Table (1): Relative fatty acid composition (%) of heated sunflower oil during deep-fat frying of potato chips at 180°C for 36 h.

Fatty acid	Control	p-fat frying of potato chips at 180°C for 36 h.  Heated time (h)						
		8	16	24	32	36		
C <sub>14:0</sub>	1.53	1.55	1.64	1.67	1.69	1.73		
C <sub>16:0</sub>	8.82	9.54	9.56	9.91	10.11	10.45		
C <sub>18:0</sub>	4.63	5.73	6.89	7.78	8.35	8.78		
C <sub>20:0</sub>	0.84	0.92	0.99	1.03	1.05	1.09		
T. sat.	15.82	17.79	19.08	20.39	21.20	22.05		
Increase %		12.14	20.60	28.88	34.00	39.38		
C <sub>16:1</sub>	0.04	0.07	0.11	0.15	0.21	0.25		
C <sub>18:1</sub>	22.59	23.02	24.76	25.99	28.31	29.84		
T. monoenes	22.63	23.09	24.87	26.14	28.52	30.09		
Increase %	•	2.03	9.89	15.51	26.03	32.96		
C <sub>18:2</sub>	61.55	59.17	56.05	53.47	50.28	47.86		
T. dienes	61.55	59.17	56.05	53.47	50.28	47.86		
Decrease %		-3.87	-8.94	-13.13	-18.31	-32.24		
T. sat.	15.82	17.74	19.08	20.39	21.20	22.05		
T. unsat.	84.18	82.26	80.92	79.61	78.80	77.95		
Decrease %		-2.28	-3.87	-5.42	-6.39	-7.40		

The total saturated and total unsaturated fatty acids were 15.82 and 84.18 %, respectively. Generally, the fatty acids composition, however, showed little marked changes due to heating under deep-fat frying conditions used at 180°C for 36 h. The fatty acids composition has been gradually affected by time of heating. The total saturates progressively increased from 15.82% for fresh oil to 22.05% after heating for 36 h with a change percent of 39.38%. Also, the total monoenes increased from 22.63% for unused oil to 30.09% after 36 h with 32.96% a change. On the other hand, the linoleic acid was the most affected component due to heating conditions. It decreased from 61.55% to 47.86% with a change percent of 32.24% after heating for 36h. The total unsaturated fatty acids, also, decreased from 84.18% to

77.95% with a change percent of 7.40% as compared to unused oil. These results suggested that the frying oil became more hydrogenated and exhibited no marked changes with deep-fat frying time up to 36 h. However, the saturated fatty acids, monoenes indicated that there was a slight but definite relative increase of these fatty acids with heating time and temperature used. These results are in agreement with those reported previously by Mazza and Qi, 1992; Tyaje and Vassisltha, 1996 and Barbary et al., 1999. Similar results were reported by Wolff (1993).

Addition of RHB or RSB to the heated oil improved the fatty acid composition as well the oil quality (Table 2). The total saturation decreased when the RHB was used with the heated oil up to 36 h. The maximum decrease % (24.42 %) was observed when the RHB was used with the heated oil after 24 h. The total monoenes and diene, on the other had, increased with using the RHB up to 36 h. The maximum increase percents in the monoenes and diene were observed at 24 h and found to be 8.15% and 5.33%, respectively. The total improvement percent in the heated oil due to using the RHB showed that this improvement was the highest (152.60%) during the first 8 h of heating then gradually decreased to 125.15, 108.97, 61.34 and finally to 44.94% after 16, 24, 32, and 36 h of heating.

Table (2): Effect of using RHB on relative fatty acid composition (%) of heated sunflower oil during deep-fat frying of potato chips at 180°C for 36 h.

180°C for 36 n.								
Fatty acid	Control	Heating time (h)						
composition		8	16	24	32	36		
T. sat.	15.82							
Heated oil		17.74	19.08	20.39	21.20	22.05		
RHB		14.81	15.00	15.41	17.90	19.25		
Decrease %		-16.52	-21.38	-24.42	-15.56	-12.69		
T. monoenes	22.63							
Heated oil		23.09	24.87	26.14	28.52	30.09		
RHB		24.12	26.25	28.27	30.19	31.76		
Increase %		4.46	5.55	8.15	5.85	5.55		
T. dienes	61.55							
Heated oil		59.17	56.05	53.47	50.28	47.86		
RHB		61.07	58.75	56.32	51.91	48.99		
Increase %		3.21	4.82	5.33	3.24	2.36		
Improvement %		152.60	125.15	108.97	61.34	44.94		

Improvement (%) = Heated oil - Treated oil x 100
Heated oil - Control

Using RSB with the heated oil showed the same trends (Table 3) but with less improvement as compared to RHB. The RSB improved the heated

oil by 135.94, 100.92, 94.75, 51.12 and 34.83% after 8, 16, 24, 32 and 36 h of heating.

The high level of alteration with respect to fatty acids observed in the sunflower oil is, also, in agreement with Dobarganes *et al.*, (1984), who indicated that alterations of used frying fats are related to the unsaturation fatty acid content of the fat as well as to the heat treatment and the number of fryings. Several auther (Nawar, 1984, Guillaumin, 1988 and Blumenthal, 1991) have pointed out that heat treatment of fats induces modifications of fatty acids with two and three double bonds, as was found in this study.

Table (3): Effect of using RSB on relative fatty acid composition (%) of heated sunflower oil during deep-fat frying of potato chips at 180°C for 36 h.

at 180°C for 36 n.								
Fatty acid composition	Control	Heating time (h)						
		8	16	24	32	36		
T. sat.	15.82							
Heated oil		17.74	19.08	20.39	21.20	22.05		
RSB		15.13	15.79	16.06	18.45	19.88		
Decrease %		-14,71	-17.24	-21.24	-12.97	-9.84		
T. monoenes	22.63							
Heated oil		23.09	24.87	26.14	28.52	30.09		
RSB		24.01	26.17	27.98	30.03	31.49		
Increase %		3.98	5.22	7.04	5.29	4.65		
T. dienes	61.55							
Heated oil		59.17	56.05	53.47	50.28	47.86		
RSB		60.86	58.04	55.96	51.52	48.63		
increase %		2.86	3.55	4.60	2.47	1.61		
mprovement %		135.94	100.92	94.75	51.12	34.83		
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Improvement (%) = Heated oil - Treated oil | x 100 | Heated oil - Control

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تأثير إستخدام مخلفات الأرز الثانوية على الخواص الطبيعية والكيميائيسة لزيت عباد الشمس خلال عملية التحمير

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تعتبر دراسة استخدام بعض المخلفات النباتية الطبيعية كمواد أدمصاص ذات أهمية قصوى من الناحية البيئية والإقتصادية للتخلص من تلك المخلفات. لذا فقد تم استخدام قشر الأرز وقش الأرز في اعاده تجديد زيت عباد الشمس المستخدم في تحمير البطاطس الشيبسي عند درجة جرارة ١٨٠٠م.

أدى استخدام قش أو قشر الأرز الى تحسنا ملحوظا في انخفاض نسبة المصواد القطبيسة الكلية . ففي حالة قشر الأرز كانت نسبة التحسن كما يلسي : ٢٩,٥، ،٩١,٧٩، ،٩١,٧١ ،٩٢، ،٩٢٩، ،٩٢٨ ، ،٠٤٠ الأرز كما يلسي : ٢٠,٥٠ ، ،٩٠٥، ،٩٠٥، ،٩٠٩، ،٩٠٩، ،٩٠٩، ،٩٠٩، ،١٠ ، ٤٠٠ ، ٢٠ ، ٢٠ ، ٢٠ ساعة تسخين على الترتيب. لذلك يمكن القول من كل هذه النتائج إمكانية استخدام قش أو قشر الأرز كمخلفات طبيعية نباتية في إعادة تجديد زيت عبد الشمس بكفاءة عالية. وإطالة عمر استخدامه في عملية التحميسر بدون أى مخططر صدية أو تكولوجية .