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Frying Oil Refreshing Capacity of a New Adsorbent Mixture

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ABSTRACT

The aim of this study was to evaluate frying oil refreshing capacity of a new adsorbent mixture composed of diatomaceous earth, natural zeolite and lime, and to compare it with a commercial adsorbent, Magnesol XL. Each adsorbent constitutive was individually tested first, and then a 1:1:1 (w/w/w) mixture was used to refresh frying oils at various levels. The new mixture at 10% addition level had around 8.1, 18.9, 58.2, 39.7, 21.7, and 18.0% improvements in viscosity, turbidity, free acidity, peroxide value, conjugated diens and total polar materials, respectively. At this level, Magnesol XL had improvements in viscosity (1.2%), turbidity (65.2%), free acidity (56.3%), and peroxide value (16.4%). In conclusion, the new mixture can be a promising adsorbent material for frying oil recovery.

Key Words: Natural zeolite, Lime, Diatomaceous earth, Magnesol XL, Frying, Refreshment.

Yeni Bir Adsorban Karışımının Kızartma Yağını Yenileme Kapasitesi

ÖZET

Bu çalışmanın amacı Diatome Toprağı, Doğal Zeolit ve Sönmüş Kireçten oluşan yeni bir adsorban karışımının kızartma yağlarını yenileme kapasitesinin ölçülmesi ve ticari bir adsorban olan Magnesol XL ile karşılaştırılmasıdır. Yeni karışımdaki her bir adsorban madde önce tek başlarına analiz edilmiş ve daha sonra 1:1:1 (ağırlıkça) karışımı seçilmiştir. Yeni adsorbanın yağda %10'luk katım seviyesindeyken, viskozite, bulanıklık, serbest asitlik, peroksit sayısı, konjuge dienler ve toplam polar maddede sırasıyla %8.1, 18.9, 58.2, 39.7, 21.7 ve 18.0'lık iyileştirme sağladığı bulunmuştur. Aynı katım seviyesinde, Magnesol XL ise sırasıyla viskozitede (%1.2), bulanıklıkta (%65.2), serbest asitlikte (%56.3) ve peroksit sayısı değerinde (%16.4) iyileştirme sağlayabilmiştir. Sonuç olarak yeni adsorban madde karışımının kızartma yağlarının yenilenmesi için ümitvar olduğu değerlendirilmiştir.

Anahtar Kelimeler: Doğal zeolit, Sönmüş kireç, Diatom toprağı, Magnesol XL, Kızartma, Yenileme.

INTRODUCTION

Frying is a very old and popular process utilized in food preparation due to its significant sales and vast quantity products. It has been used traditionally in many cuisines as well as in large food industry applications. Fried foods have usually more palatability related to unique sensory characteristics, including flavor, texture and appearance. In addition, frying has also accelerated Western cultures to accept ethnic foods, bolder flavors and traditional meals. On the other hand, the increased awareness of the consumers to the relationship between food, nutrition and health has emphasized the need to limit oil consumption, calories originating from fat, and cholesterol among others [1].

Frying is immersing and cooking foods in hot oil which includes complex phenomena like heat and mass transfers. Frying yields fast cooking, crust surface and unique flavor and texture. During the process, the presence of high temperature, oxygen, moisture, and leaching materials cause a variety of reactions to deteriorate oil, and reduce the frying oil quality and healthiness properties of the fried products [2]. Therefore, some cautions like regular cleaning and maintenance of equipment, good quality frying oil selection, utilization of proper frying conditions and use of some adsorbents for removing soluble degraded compounds have been in practice [3, 4].

Regeneration of frying oils by selective adsorption and removal of undesirable polar compounds by active filtration has been searched extensively. Commercially, the adsorbent materials are mixed with used frying oil, circulated through a filter and collected back in the fryer for subsequent uses. This treatment has been shown to maintain frying life of oil by controlling the buildup of total polar materials and free fatty acids without any adverse effect on the safety and sensory quality of the fried products. Many different adsorbent materials including activated carbon, clay, aluminum hydroxides, charcoal, celite, silica gel, silicon dioxide, oyster shell, ceramic plate, calcium oxide and others have been searched for this purpose. There are commercial adsorbent products like Magnesol®, Britesorb™, Frypowder (Miroil) and OilFresh™ around markets [1, 5, 6].

The aim of this study was to determine the effectiveness of a new adsorbent mixture to refresh used frying oils. The new adsorbent mixture was made of equal weight mixtures of diatomaceous earth, natural zeolite, and lime. In addition, effectiveness of this new mixture was compared with one of the commercial adsorbent product, Magnesol® XL in this study.

MATERIALS and METHODS

Materials

The canteen of the engineering faculty kindly provided the used frying oils for this study. The oil was used for a week in potatoes and chicken frying. The frying oil was refined-winterized sunflower oil. Lime was purchased from local stores. Natural zeolite (10-50 µm) (Türkzeolit Madencilik Co., Balıkesir), Diatomaceous Earth (5-20 µm) (CMM Chemicals Co., Istanbul) and Magnesol® XL (synthetic magnesium silicate) (Yılmaz Gıda A.Ş., İzmir) were gifted by the companies. All chemicals used for the analyses were of analytical grade (Merck Co., Darmstadt, Germany).

Development of the new adsorbent mixture

In order to determine the effects of the selected adsorbent materials, at first, each adsorbent material was mixed separately with 500 mL used frying oil (10% w/w, each) at 150°C and stirred for 30 min. Then the slurry was vacuum filtered through Whatman no. 41 filter paper. Treated oil samples were kept in amber colored screw cap glass bottles in fridge until the analyses. After examination of the results of these samples, we decided to mix the three adsorbent materials by weight ratio of 1:1:1 to create the new mixture.

In the second part of the study, the most efficient addition level of adsorbent mixture into frying oil was determined. The adsorbent mixture was added into the used frying oil by 1, 3, 5, 7, and 10%, and collected in the same way. The samples were evaluated as the same.

In the third part of the study, comparison of the new adsorbent mixture and commercial Magnesol XL was studied. The used frying oil was treated by the same technique with 1, 3, 5, 7, and 10% added level of the adsorbents. The samples were collected and analyzed in the same manner.

Physical analyses

Viscosity measurements of the oils were carried out by placing 7.5 mL of sample in a special sample holder, and direct measuring centipoises (cP) with a Brookfield viscosimeter (model DV II+Pro with Rheocalc software, Brookfield Eng. Lab., Inc., MA, US) equipped with LV-SC4-18 spindle at 25°C. Turbidity values of the samples were measured by Micro T100 Lab Turbidimeter (HF Scientific Inc, US) calibrated with 0.2, 10 and 1000 NTU calibration liquids, according to the manual of the instrument at 25°C. Refractive indexes of the samples were measured under daylight with a 2WAJ model Abbe refractometer, calibrated against pure water at 25°C. Smoke points of the samples were measured following AOCS method Cc 9a-48 [7]. Instrumental color of the samples were measured by a Minolta CR-400 Chroma Meter (Osaka, Japan) by immersing the probe of instrument into the oil sample put in a petri dish on the white tile. Readings of the CIE L, a* and b* values are recorded.

Chemical analyses

Free fatty acids values of the samples were determined by Ca 5a-40 of American Oil Chemists Society methods [8]. Peroxide values were determined by AOCS method Cd 8-53 [9]. Conjugated diens of the samples were measured following Ti 1a-64 method [10] on a UV Mini 1240 Spectrophotometer (Shimadzu Co, Japan). Quick analysis of the total polar materials of the frying oil samples were made by sensor reading (Testo 265, Lenzkirch, Germany). The sensor functions on a capacitive basis and takes the overall level of polar materials in % as its reading. According to the usage manual, the sensor first calibrated with the incoming calibration oil, and then direct readings of the percent total polar materials values were accomplished by immersing the probe into hot oil (above 40 °C) and reading the value.

Statistics

The study was accomplished twice with all analyses performed in parallel. The statistical package programs of Minitab (ver. 14.1) and SPSS (ver.10.1) were used for all analyses. Significant differences among the means of the samples for the measurements were determined by the analysis of variance using the Duncan test at 95% of confidence [11, 12].

RESULTS

For this study, the one week fried sunflower oil was used. At the first part of the study, the recovery capacities of the individual adsorbent materials were tested (Table 1). Percent changes of the measured values was calculated over the control measurements. The viscosities of the used frying oils were decreased by 2.0, 1.5 and 1.9% by diatomaceous earth, natural zeolite and lime itself at the 10% (w/w) addition level. On the other hand, effect of lime on the turbidity was very negative, enhancing the value by 76.7%. There was no significant effect on the refractive indices of the samples. Instrumental values of the color of the samples are shown in Table 1 as well. Luminosity (L) of the oils improved by diatomaceous earth, but decreased by natural zeolite and lime. Similar to the decrease in turbidity value, lime has caused the luminosity of the samples to decrease significantly. It might be possible that lime may form some insoluble complexes with some molecules in used frying oils. All three treatments have caused the a* value to shift towards the negative direction, that is from red to green color value. Similarly yellowness, indicated by positive b* value, was decreased. The effects of each adsorbent material on the measured chemical quality indices of frying oil are also shown in Table 1. While free acidity of the oil sample has decreased by 59.7% by lime, diatomaceous earth has, on the contrary, caused the value to increase by 10.4%. It can be quite possible that lime is neutralizing the free fatty acids, while diatomaceous earth itself yields some hydrogen ions into the oil to increase the measured acidity. On the other hand, peroxide value of used frying oil was decreased by the three adsorbents significantly. Similar results were observed with the conjugated diens and total polar materials. These effects are very positive in refreshing the degraded frying oil for reuse purposes. In literature, very diverse ranges of the effects of adsorbent materials on frying oils were reported. A combination of Hubesorb600, Frypowder and Magnesol (3:3:2 w/w/w) reduced the total polar materials, color and free acidity by 30, 52 and 72%, respectively [13]. Treatment of fried oils with Magnesol XL, diatomaceous earth and kaolin at the 1, 2 and 4% addition levels were greatly improved the quality of fried oils. These findings indicate the high efficiency of the filter aids in adsorbing the products of oil degradation [14]. In another study, the maximum improvement was up to an extent of 7.0% to 8.8%, 15.2% to 18%, 11.7% to 14.6% and 4.5% to 5.9% for capacitance (C), viscosity (μ), photometric color index (PCI) and free fatty acids (FFAs), respectively. Specific gravity (γ) and color parameters (CIE L*, a*, b* and Δ E*) showed no significant difference due to treatments [15].

Table 1. The effects of individual adsorbent materials on the physical, instrumental color and chemical properties of frying oil.

Parameter	Control	Diatomaceous Earth	Natural Zeolite	Lime
Viscosity (cP,25ºC) (P=0.0019)	85.40±0.20 A*	83.70±0.10 B (2%)**	84.10±0.10 B (1.5%)	83.75±0.05 B (1.9%)
Turbidity (NTU) (P<0.0001)	1.09±0.02 B	0.96±0.0 D (12.3%)	1.03±0.0 C (6.4%)	1.94±0.01 A (- 76.7%)
Refractive index (25ºC) (P=0.999)	1.48±0.0 A	1.48±0.0 A	1.48±0.0 A	1.48±0.0 A
L value (P=0.005)	52.02±0.23 B	56.65±0.59 A (- 8.9%)	51.89±0.34 B (0.2%)	45.72±1.69 B (12.1%)
a* value (P<0.0001)	-0.40±0.06 A	-1.12±0.02 B (180%)	-2.36±0.07 D (490%)	-1.80±0.02 C (350%)
b* value (P=0.0011)	25.46±1.80 A	10.44±0.11 C (59%)	13.67±0.05 BC (46.3%)	14.51±0.06 B (43%)
FFA (% oleic acid) (P<0.0001)	0.28±0.001 B	0.31±0.001 A (-10.4%)	0.25±0.001 C (0.2%)	0.11±0.0 D (59.7%)
Peroxide value (meq/kg) (P=0.1300)	25.61±0.37 A	24.98±0.92 A (2.5%)	22.03±0.84 A (14%)	22.47±1.42 A (12.2%)
Conjugated diens (%) (P=0.011)	3.024±0.10 A	2.63±0.0 B (13%)	2.62±0.005 B (13.3%)	2.65±0.0 B (12.5%)
Total polar materials (%) (P=0.0004)	28.75±0.25 A	26.50±0.0 C (7.8%)	26.0±0.0 D (9.6%)	27.0±0.0 B (6.1%)

* Different letters within a row show significant differences among the adsorbent materials for each measurement.
** (% Change value) was calculated over the control value for each of the measurements.

Data evaluation from the first part of the study revealed that a combination of the adsorbents would be more effective to regenerate the degraded frying oils. Hence, it was decided to use a 1:1:1: (w/w/w) ratio of the diatomaceous earth, natural zeolite and lime. Another batch of used frying oil sample (one week used sunflower oil) was collected from the school canteen. Since the same oil and the same frying conditions had been used for the frying, and data comparisons were by the calculated % change value, it would not be a problem of using a second batch of sample for this part of the study. Then different weight ratios of the new adsorbent mixture were added and treated in the same way. Results of the Duncan's comparisons of the measurements were shown in Tables 2. Viscosity and turbidity of the samples were decreased continually by increased level of adsorbent addition, while no change was observed with refractive index values. At the 10 % addition level of new adsorbent mixture, viscosity and turbidity have decreased by 8.05 and 18.93% over the control sample, respectively. Although different addition levels were statistically not significant, enhanced level of adsorbent addition has caused the L and b* values to decrease. Similarly a* value has also decreased with significant differences among the addition levels. By 10% added adsorbents, the values of L, a* and b* were decreased by 7.70, 464.71 and 28.17%, respectively. A mixture of pekmez soil, bentonite and magnesium silicate was used for frying oil recovery [16] in a study. There was no significant effect of the adsorbent mixture on the viscosity and peroxide value, while the L value was decreased and the a* and b* values were increased. In another study [17], effect of frying on the dynamics of visual characteristics of oil was investigated. No significant change was observed in the refractive index of the samples. The turbidity decreased with frying time due to the increase in the fat darkness. The redness, yellowness and chroma increased gradually until 3rd. and 5th. days of frying and decreased afterwards due to increase in darkness. Lightness decreased and photometric color index increased with increase in frying time and correlated well with major degradation products. Generally findings in our study are in agreement with the published literature.

Table 2. The effects of the adsorbent mixture addition level on the measured properties of frying oil.

Parameter	Addition Levels of the Adsorbent Mixture (%)					
	Control	1	3	5	7	10
Viscosity (cP, 25ºC) (P<0.0001)	91.95±0.15 A*	91.95±0.05 A	91.75±0.15 A	90.85±0.05 B	90.40±0.10 C	84.55±0.05 D
Turbidity (NTU) (P=0.0002)	0.845±0.01 A	0.805±0.005 A	0.705±0.01 B	0.650±0.020 C	0.645±0.005 C	0.685±0.01 BC
Refractive index (25ºC) (P=0.999)	1.477±0.000 A	1.477±0.000 A	1.477±0.000 A	1.477±0.000 A	1.477±0.000 A	1.477±0.000 A
L value (P=0.43)	51.79±0.00 A	50.68±4.83 A	48.68±1.23 A	45.72±1.69 A	46.97±0.24 A	47.80±0.21 A
a* value (P=0.18)	-0.34±0.00 AB	0.84±1.79 A	-0.82±0.70 AB	-1.09±0.51 AB	-2.85±0.43 B	-1.92±0.66 AB
b* value (P=0.32)	27.26±0.00 A	17.67±1.23 A	25.71±2.05 A	21.59±3.58 A	21.83±4.96 A	19.58±3.27 A
FFA (%oleic acid) (P<0.0001)	0.361±0.004 A	0.264±0.012 B	0.245±0.003 B	0.197±0.008 C	0.167±0.000 D	0.151±0.011 D
Peroxide value (meq/kg) (P<0.0001)	25.88±0.64 B	30.97±0.57 A	24.70±0.00 B	20.505±0.28 C	25.70±0.10 B	15.615±0.49 D
Conjugated diens (%) (P=0.001)	1.396±0.02 A	1.221±0.01 B	1.371±0.04 A	1.244±0.01 B	1.162±0.04 BC	1.072±0.006 C
Total polar materials (%) (P<0.0001)	30.5±0.0 A	29.5±0.0 B	29±0.0 C	29±0.0 C	28±0.0 D	25±0.0 E

* Different letters within a row show significant differences among the addition levels tested.

Comparisons of the measured chemical quality indices for the different levels of added new adsorbent mixture for frying oil are also shown in Table 2. As the level of adsorbent addition increased, all the measured chemical indices were decreased, except peroxide value. There was no clear trend in the peroxide value changes because it is well known that peroxides in frying oils are continually forms and decomposes during the high temperature process. Dependently, in frying oils the level of oxidation is mostly measured by conjugated dien value. There were statistically significant differences among the addition levels of the new adsorbent mixture for the measured chemical indices. At the 10 % added adsorbent mixture level, the reduction in free acidity, peroxide value, conjugated diens and total polar materials were 58.17, 39.66, 21.69 and 18.03%, respectively. In one study, a combination of Britesorb, Hubesorb600, Frypowder and Magnesol was used to regenerate frying oil, and recoveries of 61.9% free acidity, 59.7% total polar materials, 38 % of food oil sensor reading and 11.3% total color difference were reported [18]. Especially for the chemical indices, results in our study agree with previous findings. More recently [6], for the recovery of thermally degraded palm oil, activated charcoal powder, aluminum oxide, aluminum hydroxide, activated clay, celite, silica gel, activated carbon, frypowder, britesorb, magnesol and their binary and quaternary mixtures were evaluated for their efficacy in regenerating the oils in terms of their effects on physicochemical parameters such as free fatty acids, total polar materials, p-anisidine value, conjugated dienes, viscosity and color. It was concluded that, binary and quaternary adsorbent mixtures were better than single adsorbents in improving the overall quality of the oils studied.

In the last part of the study, a commercial adsorbent, Magnesol XL, was evaluated at different addition levels to observe its effectiveness and to compare it with the new adsorbent mixture studied. Similar to the results of the new adsorbent mixture, Magnesol XL had no effect on the refractive index and had some effects on viscosity and turbidity of the samples (Table 3). Effect on oil turbidity was higher than that of the new adsorbent mixture. Magnesol XL reduced turbidity by 65.19% over the control sample at the 10% addition level. Added Magnesol XL's effect on the instrumental color values of frying oil samples was in the same direction as measured with the new adsorbent mixture (Table 2). That is the clearness (L value) and greenness (negative a* value) were enhanced, while the yellowness (decreasing positive b* value) was decreased by the treatment.

Effects of different levels of added Magnesol XL on the measured chemical indices of the frying oil samples are also shown in Table 3. As the addition level increased, the free acidity was reduced. By 10% addition of Magnesol XL, the free acidity of sample was decreased by 56.25% over the control sample. A similar result was observed with the new adsorbent mixture addition (Table 2). There was no statistically significant effect on the peroxide value of the samples. Most interestingly, the amounts of conjugated diens and total polar materials were increased by different levels of Magnesol XL addition compared to the control sample. Since all treatments were applied in the same way, the findings for Magnesol XL are quite unexpected and important to evaluate in further studies. In a study [15], quality of degraded frying oil using Magnasol and Filtrite adsorbents was evaluated. The improvement up to an extent of 7.0% to 8.8%, 15.2% to 18%, 11.7% to 14.6% and 4.5% to 5.9% for capacitance (C), viscosity (µ), photometric color index (PCI) and free fatty acids (FFAs) were reported.

Table 3. The effects of the Magnesol XL addition level on the measured properties of frying oil.

Parameter	Addition Levels of the Magnesol XL (%)					
	Control	1	3	5	7	10
Viscosity (cP, 25ºC) (P<0.0001)	61.00±0.00 A*	60.75±0.05 A	60.75±0.05 A	60.10±0.10 B	60.15±0.05 B	60.25±0.05 B
Turbidity (NTU) (P<0.0001)	2.70±0.01 A	2.50±0.02 B	1.27±0.01 C	0.95±0.00 E	1.20±0.00 D	0.94±0.00 E
Refractive index (25ºC) (P=0.999)	1.471±0.000 A	1.471±0.000 A	1.471±0.000 A	1.471±0.000 A	1.471±0.000 A	1.471±0.000 A
L value (P=0.004)	52.12±0.03 A	51.27±0.01 B	51.16±0.02 B	51.13±0.01 B	50.66±0.01 B	51.10±0.33 B
a* value (P<0.0001)	-1.01±0.01 A	-0.26±0.79 A	-2.91±0.01 B	-4.09±0.01 CB	-4.48±0.04 CB	-4.76±0.01 C
b* value (P=0.001)	32.39±0.03 AB	36.07±2.40 A	29.97±0.04 BC	26.29±0.01 CD	25.25±0.03 CD	22.71±0.15 D
FFA (%oleic acid) (P<0.0001)	0.480±0.005 A	0.391±0.000 B	0.338±0.003 C	0.279±0.001 D	0.238±0.014 DE	0.210±0.013 E
Peroxide value (meq/kg) (P=0.911)	2.98±0.96 A	2.99±0.01 A	2.98±0.02 A	3.01±0.01 A	2.99±0.00 A	2.49±0.00 A
Conjugated diens (%) (P=0.019)	1.095±0.003 C	1.202±0.007 A	1.190±0.026 AB	1.207±0.017 A	1.224±0.015 A	1.129±0.033 BC
Total polar materials (%) (P=0.012)	18.50±0.00 C	19.00±0.00 ABC	19.50±0.00 A	19.25±0.25 AB	18.75±0.25 BC	18.50±0.00 C

* Different letters within a row show significant differences among the addition levels tested.

CONCLUSION

This study shows for the possibility of natural zeolite as frying oil recovery adsorbent. The new adsorbent mixture composed of natural zeolite, lime and diatomaceous earth has been shown as more effective than the commercial product, Magnesol XL, for used frying oil recovery. For frying oils, total polar materials (TPM) is the most important parameter regulated by the Turkish Official Notification of the Control Criteria of Frying Fats/Oils [19]. According to the codex [19], frying oils must have TPM value equal or lower than 25%. This study has shown that used frying oil having 30% TPM value have decreased to 25% by the adsorbent mixture treatment. Hence, the prepared mixture of adsorbents can successfully be implemented for frying oil recovery and shelf life extension. Literatures indicate some other adsorbents with much higher effect on the refreshing of frying oils. The natural zeolite used in this study was a microporous material, not broken up to the nanoscale level. Natural zeolites are microporous, aluminosilicate minerals commonly used as commercial adsorbents and known as molecular sieves. The maximum size of the molecular or ionic species that can enter the pores of a natural zeolite is controlled by the dimensions of the channels. These are conventionally defined by the ring size of the aperture, where, for example, the term ‘8-ring’ refers to a closed loop that is built from 8 tetrahedrally coordinated silicon (or aluminum) atoms and 8 oxygen atoms. The pores in many natural zeolites are not cylindrical [20]. In this study, there was no prior treatment or modification on the natural zeolite used. It might be possible that some dimensional or surface modifications of natural zeolite may cause some improvements on the effects of frying oil recovery abilities.

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