

Analytical Methods

γ -Fe₂O₃ magnetic nanoparticle functionalized with carboxylated multi walled carbon nanotube for magnetic solid phase extractions and determinations of Sudan dyes and Para Red in food samples

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ABSTRACT

Hybrid nanostructures composed of γ -Fe₂O₃ (maghemite) and carboxylated-multi walled carbon nanotube (cMWCNT) were used for the magnetic solid phase extractions and determination of Sudan I, II, III, IV, Para Red, Sudan Black B and Sudan Red 7B in chili products. High performance liquid chromatography (HPLC) was employed for the measurements. Limit of quantification (LOQ) values were found in the range 0.44–2.82 ng mL⁻¹ for analytes. The best extraction parameters were determined as pH 8.0, 40 mg of magnetic nanoparticle, 4.0 min of contact time, 0.3 mL desorption by acetonitrile. The samples were dissolved in acetone-dichloromethane-methanol (3:2:1, v/v/v) and diluted with acetonitrile-methanol (v/v; 80:20) before the method was applied. Concentrations of Sudan dyes and Para Red were determined in four samples of chili powder from less than LOQ to 31.21 ± 1.6 ng g⁻¹, two samples of chili tomato sauces (lower than LOQ) and two samples of ketchup (lower than LOQ).

1. Introduction

Sudan dyes have been categorized as class 3 carcinogens by the International Agency for Research on Cancer (IARC) (Mo, Zhang, Zhao, Xiao, Guo, & Zeng, 2010; Stiborova, Martinek, Rydlova, Hodek, & Frei, 2006). Despite this, Sudan dyes are used illegally for coloring foods, like ketchup and chili products, to improve their appearance. In 2014, they were detected in approximately 580 different food products bought in EU countries, and the products were withdrawn (Food Standard Agency, 2014).

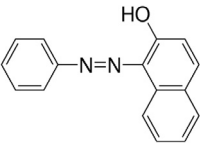
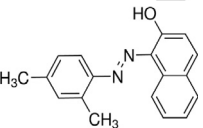
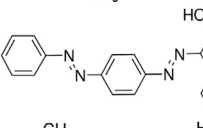
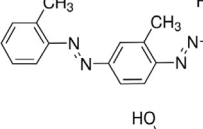
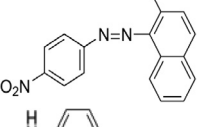
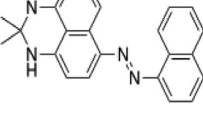
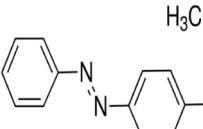
The chemical structures, molecular formulas and CAS (Chemical Abstract Service) numbers of Sudan I, II, III, IV, Para Red, Sudan Black B and Sudan Red 7B are (Table 1) were reviewed recently (Yanjala, Nainar, & Ramiseti, 2016). Membrane filtration, solid phase extraction, liquid-liquid extraction, microwave, and ultrasound-assisted extraction have been carried out to clean up samples for analysis using spectrophotometry, thin layer chromatography, liquid chromatography with and without mass spectrometry (Yanjala et al., 2016). Molecularly imprinted solid-phase extraction coupled with liquid chromatography has also been used for the determinations of Sudan I, II, III and IV in 30 bean curd products (Yan et al., 2012). LODs were in the range of 5–9 µg kg⁻¹. A SPE-alumina-N cartridge was used to clean up the

sample before HPLC analysis of Sudan I, II, III and IV in chili products. Here, however, LODs were found in the range 4.1–5.8 µg kg⁻¹ (Qi, Zeng, Wen, Liang, & Zhang, 2011). Before liquid chromatography-photodiode array determinations of Sudan I, II, III and IV in chili-containing foodstuffs, supramolecular solvent-based microextraction was applied for sample clean up. The detection limits of the method were 4.2, 2.7, 6.5 and 7.4 µg kg⁻¹ for Sudan I, II, III and IV, respectively (López-Jiménez, Rubio, & Pérez-Bendito, 2010). Using atmospheric pressure photoionization-tandem mass spectrometry, LODs ranged between 5 and 18 µg L⁻¹ and LOQ between 10 and 24 µg L⁻¹ for Sudan I, II, III and IV (Murty, Chary, Prabhakar, Raju, & Vairamani, 2009). Sudan I, II, III and IV were determined in candy using ionic liquid/anionic surfactant aqueous two-phase extraction coupled with high-performance liquid chromatography (Yu, Liu, Li, Zhang, & Yu, 2015) in 80 red chili peppers (including Para Red) (Ertas, Özer, & Alasalvar, 2007), in soft drink samples with an electrochemical detector (Chailapakul et al., 2008) and in chili products with gel permeation chromatography (GPC) clean-up (including Para Red and Sudan Red 7B) (Zhu et al., 2014). These methods are based on time-consuming procedures for pre-concentration of samples. Using magnetic nanomaterials would eliminate the need for such procedures whilst still achieving comparable LODs and LOQs.

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Table 1
Chemical informations of Sudan I, II, III, IV, Para Red, Sudan Black B and Sudan Red 7B.

Dye	Molecular formula	CAS No	Structure
Sudan I	C ₁₆ H ₁₂ N ₂ O	842-07-9	
Sudan II	C ₁₈ H ₁₆ N ₂ O	3118-97-6	
Sudan III	C ₂₂ H ₁₆ NO	85-86-9	
Sudan IV	C ₂₄ H ₂₀ N ₄ O	85-83-6	
Para Red	C ₁₆ H ₁₁ N ₃ O ₃	6410-10-2	
Sudan Black B	C ₂₉ H ₂₄ N ₆	4197-25-5	
Sudan Red 7B	C ₂₄ H ₂₁ N ₅	6368-72-5	

In this study, carboxyl functionalized MWCNT- γ -Fe₂O₃ MNPs synthesized previously (Kılınc, 2016) was used for magnetic solid phase extractions and determination of Sudan I, II, III, IV, Para Red, Sudan Black B and Sudan Red 7B in foods using HPLC. Using the magnetic nanomaterials overcame problems with traditional solid phase extraction and time-consuming procedures, such as filtration and centrifugation on SPE, were eliminated, allowing large numbers of samples to be tested for Sudan dyes.

2. Materials and methods

2.1. Reagents and standards

Multiwalled carbon nanotube (o.d. 10–15 nm, i.d. 2–6 nm, length 0.1–10 μ m), toluene, NH₄OH and Sudan I (≥ 95), Sudan II (≥ 96), Sudan III (≥ 96), Sudan IV (≥ 96), Para Red (> 95), Sudan Black B and Sudan Red 7B (≥ 96) were supplied from Sigma-Aldrich (St Louis, MO, USA). FeCl₃·6H₂O, FeCl₂·4H₂O were bought from Merck (Darmstadt, Germany) and Fluka (St Louis, MO, USA). All chemicals were of analytical reagent grade. Doubly distilled water was used for the entire experiments. Solvents were of HPLC grade and obtained from Merck. PTFE membrane filters (0.2 μ m) were supplied by Waters (CITY, Massachusetts, USA).

After the required amounts of each dye were mixed with acetone-dichloromethane-methanol (3:2:1, v/v/v), the solution was heated to

40 °C for 10 min to dissolve the dyes completely. Acetonitrile-methanol (v/v; 80/20) was used to make up the final volume (100 mL).

2.2. Instrumentation

The chromatography equipment with a Dionex Ultimate 3000 LC system consisted of a quaternary pump, degasser, auto sampler, DAD detector and thermostated column compartment (Agilent, Waldbronn, Germany). The ACE 5 C18 column (250 \times 4.6 mm stainless steel, particles size as 5 μ m) was used for separation of Sudan dyes and Para Red. Acetonitrile-methanol (v/v; 80/20) was used as a mobile phase with a flow rate of 1.0 mL min⁻¹. The targeted analytes were monitored at 504 nm.

2.3. Sample preparation for magnetic solid phase extraction

10 g of chili powder, chili tomato sauces and ketchup samples were weighed into a 50 mL glass bottle. After the addition of 30 mL of acetone-dichloromethane-methanol (3:2:1, v/v/v), the mixtures were ultrasonicated for 15 min at 40 °C. Then, acetonitrile-methanol (v/v; 80:20) was added to a final volume of 50 mL. The final solutions were subjected to SPE and passed through a 0.2 μ m syringe filter before injection on to the HPLC column. Spiked samples were prepared in the same way. A mixed stock standard solution was added to samples before extraction.

2.4. Magnetic solid phase extraction procedure

For the magnetic solid phase extraction, 100 μ L of methanol and 100 μ L of water were added sequentially to activate 20 mg of cMWCNT- γ -Fe₂O₃. This was dispersed in 50 mL of sample and the mixture shaken on a slow-moving platform shaker for 15 min. The magnetic adsorbent was isolated from the solution by means of a strong magnet (Nd-Fe-B; 100 mm 100 mm 20 mm) at the bottom of the beaker and most of the supernatant was poured away. Then, the magnetic nanoparticles and residual solution were transferred to a 15-mL centrifuge tube. The desorption of analytes was realized with the addition of 0.5 mL of acetone twice with vortexing for 1 min. These solutions were combined and transferred to a 2 mL microcentrifuge tube before being dried under nitrogen. The residue after drying was reconstituted in 250 μ L of acetonitrile-methanol (v/v; 80/20) and passed through a PTFE filter (0.2 μ m). 15 μ L was injected on to the HPLC system for analysis.

3. Results and discussions

3.1. Investigation of extraction parameters for magnetic solid phase extraction of Sudan dyes and Para Red

50 mL portions of double-distilled water spiked with 10 ng mL⁻¹ of each dyes (model solution) were used to determine the best experimental conditions. The effects of pH, amounts of magnetic nanoparticles, extraction time, and type and the volume of desorber were investigated. All experiments were performed in triplicate. pH is one of the most important parameters that influences adsorption and desorption behaviors (Özdemir, Okumuş, Dündar, & Kılınc, 2013). The effect of pH on magnetic solid phase extraction of Sudan dyes was investigated in the range 3–9. pH values was adjusted with NaOH and HNO₃. As shown in Fig. 1, the percentage adsorption increased with increasing pH to a maximum at pH 8. Adsorption decreased at higher pH values, suggesting the negatively charged sites on the adsorbent surface did not favor Sudan dyes.

The amount of magnetic nanoparticle, as a solid phase adsorbent, was examined for quantitative extraction and removal of Sudan dyes in the range 10–100 mg of cMWCNT- γ -Fe₂O₃. Maximum adsorption capacities were achieved when 40 mg of cMWCNT- γ -Fe₂O₃ was in contact with a model solution for 15 min. The results are shown in Fig. 2.

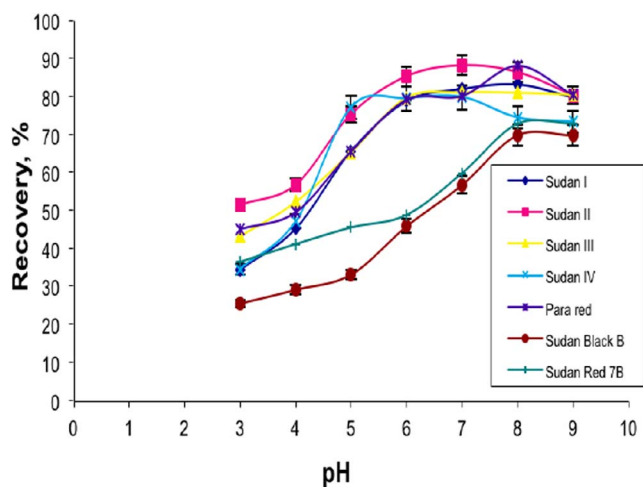


Fig. 1. Effect of pH on the extraction of Sudan dyes and Para Red by cMWCNT- γ -Fe₂O₃.

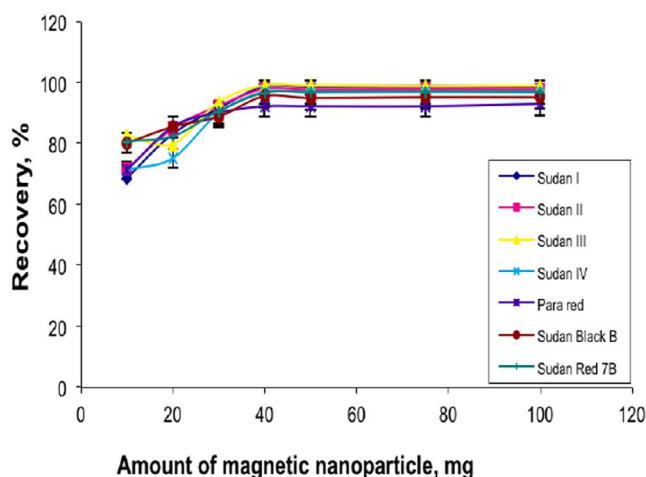


Fig. 2. Effect of amount of magnetic nanoparticle on the extraction of Sudan dyes and Para Red by cMWCNT- γ -Fe₂O₃.

The effect of extraction time on the magnetic solid phase extraction of Sudan I, II, III, IV, Para Red, Sudan Black B and Sudan Red 7B was investigated in the range 1–20 min (Fig. S1). The adsorption of Sudan dyes increased up to 4 min and then remained constant. The results can be attributed to shorter diffusion route for MNPs. In other words, rapid extraction and desorption of the analytes from the MNPs could shorten the analysis time considerably. Therefore, 4 min was chosen as the best extraction time.

Different solvents were used to find the best for quantitative extraction of targeted analytes. Water, methanol, ethanol, 1-propanol, acetonitrile and acetonitrile-methanol (v/v; 80/20) were applied, but the highest quantitative extraction efficiency was obtained with acetonitrile.

The influence of solvent volume on desorption efficiency of the

analytes was also investigated. As a result, quantitative desorption of the analytes was achieved with 0.3 mL of acetonitrile (two desorptions steps with 0.15 mL, respectively). Due to the dilution effect, the peak areas of the analytes decreased when the volume of desorption solvent was greater than 0.3 mL. The results are presented in Fig. S2.

3.2. Analytical figures of merit

Analytical characteristics of the developed method (Table 2) were evaluated for the best extraction conditions (pH 8.0, 40 mg of the magnetic nanoparticle, 0.3 mL of acetonitrile as the elution solvent, 4.0 min extraction time). The results indicated that wide linear ranges and high correlation coefficients (> 0.99) were achieved. The limits of detection (LOD) and quantification (LOQ) were calculated from the ratio of 3- and 10-times of the standard deviation of the lowest concentration in the linear range to the slope of the linear calibration plot, respectively. All compounds showed good linearity with determination coefficients (r^2) greater than 0.9900.

Repeatability was illustrated with the relative standard deviations (RSD), which ranged between 1.6 and 4.8%. Preconcentration factors were calculated as 167 for Sudan I, II, III, IV, Para Red, Sudan Black B and Sudan Red 7B, based on the initial and final volumes of 50 mL and 0.30 mL respectively.

LODs, based on use of different sample clean up procedures, are presented in Table 3. The magnetic nanoparticle method exhibited lower LODs than alternative methods.

To investigate the re-usability of cMWCNT- γ -Fe₂O₃ as a sorbent for magnetic solid phase extraction, it was washed with 5 mL of acetone twice and reused. The results showed that cMWCNT- γ -Fe₂O₃ could be re-used at least 20 times (each of three replicate) without a significant loss of the sorption capacity.

3.3. Real sample analysis

The method was applied for the determination of Sudan I, II, III, IV, Para Red, Sudan Black B and Sudan Red 7B in eight different food samples. Among the analyzed samples, no residues of the Sudan II, IV, Para Red, Sudan Black B and Sudan Red 7B were detected. However, Sudan I ($31.21 \pm 1.6 \text{ ng g}^{-1}$) and Sudan III ($41.5 \pm 2.2 \text{ ng g}^{-1}$), the use of which is illegal in Turkey, were detected in chili powder and chili tomato sauces samples. To demonstrate the applicability and accuracy of this method, three samples were spiked with 20 and 40 ng mL^{-1} of Sudan I, II, III, IV, Para Red, Sudan Black B and Sudan Red 7B. The results are presented in Table 4.

4. Conclusion

In this study, a novel magnetic adsorbent (cMWCNT- γ -Fe₂O₃) was used successfully as a magnetic material for solid phase extraction of Sudan I, II, III, IV, Para Red, Sudan Black B and Sudan Red 7B in food samples. Good repeatability, recoveries and high enrichment factors were obtained. The results in this study indicate that cMWCNT- γ -Fe₂O₃ may be used as a magnetic solid-phase extraction material and applied to detect the illegal addition of Sudan dyes in foods. Using the magnetic

Table 2
Analytical figures of merit.

Parameter	Sudan I	Sudan II	Sudan III	Sudan IV	Para Red	Sudan Black B	Sudan Red 7B
Linear range, ng mL^{-1}	3.0–60	3.0–60	6.0–60	3.0–60	3.0–60	3.0–60	3.0–60
r^2	0.9996	0.9936	0.9961	0.9975	0.9998	0.9949	0.9987
RSD ¹ , %	1.6	2.9	4.8	1.6	2.6	1.8	2.3
LOD, ng mL^{-1}	0.16	0.26	0.84	0.13	0.24	0.16	0.21
LOQ, ng mL^{-1}	0.54	0.88	2.82	0.44	0.81	0.53	0.69

¹ RSD values were calculated for the lowest concentration in linear range ($n = 7$).

Table 3
Comparison of LODs of different methods for Sudan dyes and Para Red.

Method and matrices	LOD, ng mL ⁻¹								Ref.
	Sudan Red 7B								
	Sudan I	Sudan II	Sudan III	Sudan IV	Para Red	Sudan Black B	Sudan Red 7B		
Ultra-high performance supercritical fluid chromatography, chili-containing samples	0.21 ¹	0.23 ¹	0.45 ¹	0.30 ¹	0.30 ¹	–	0.60 ¹	–	Khalikova, Satinsky, Solich, and Novakova (2015)
Extraction by magnetic molecularly imprinted polymers before HPLC-UV; chili powder samples	1.8 ²	2.6 ²	3.4 ²	5.7 ²	–	–	–	–	Xie, Chen, Pan, and Wang (2015)
Microwave-assisted homogeneous ionic liquid microextraction before HPLC; red wine	0.19	0.18	0.24	0.16	–	–	–	–	Song, Wu, Li, Hu, and Wang (2015)
LC-MS/MS; chili powders and syrup-preserved fruits	0.01 ^{1,3}	0.005 ^{1,3}	0.025 ^{1,3}	0.05 ^{1,3}	0.01 ^{1,3}	–	0.001 ^{1,3}	–	Tsai, Kuo, and Shih (2015)
Organic solvent-free air-assisted liquid-liquid microextraction before HPLC; spices, cosmetics and human bio-fluid samples	7.7 ²	8.3 ²	12.1 ²	10.6 ²	–	–	–	–	Barfi, Asghari, Rajabi, and Sabzalain (2015)
Extraction by magnetic titanium dioxide nanoparticles before HPLC; environmental water	7.3 ⁴	3.9 ⁴	2.1 ⁴	6.8 ⁴	–	–	–	–	Li, Chen, and You (2014)
SPE before HPLC; preserved beanscurds	0.005 ¹	0.005 ¹	0.009 ¹	0.009 ¹	–	–	–	–	Yan et al. (2012)
SPE before HPLC; chili foods	5.6 ²	4.2 ²	4.1 ²	5.8 ²	–	–	–	–	Qi et al. (2011)
Multi-wavelength HPLC fingerprint; chili products	1.58 ¹	–	–	0.76 ¹	0.37 ¹	0.51 ¹	0.58 ¹	–	Zhu et al. (2016)
Supramolecular solvent-based microextraction; chili-containing foodstuffs	4.2 ²	2.7 ²	6.5 ²	7.4 ²	–	–	–	–	López-Jiménez et al. (2010)
Atmospheric pressure photoionization-tandem mass spectrometry; chili containing food products	5	8	15	18	–	–	–	–	Murty et al. (2009)
Ionic liquid/anionic surfactant aqueous two-phase extraction before HPLC; candy samples	5.45 ²	4.66 ²	3.68 ²	4.20 ²	–	–	–	–	Yu et al. (2015)
Solvent extraction before HPLC; red chili pepper	1.2 ²	3.9 ²	1.2 ²	5.4 ²	3.6 ²	–	–	–	Ertaş et al. (2007)
Evaporating and diluting with mobile phase before HPLC; soft drink samples.	0.001 ²	0.001 ²	0.005 ²	0.025 ²	–	–	–	–	Chailapakul et al. (2008)
Gel permeation chromatography clean-up before HPLC; chili products	13	13	10	11	11	–	15	–	Zhu et al. (2014)
SPE before HPLC; chili products and ketchup	0.16	0.26	0.84	0.13	0.24	0.16	0.21	–	This study

¹ µg g⁻¹,

² µg kg⁻¹,

³ LOQ,

⁴ ng L⁻¹.

Table 4
Determination of Sudan I, II, III, IV, Para Red, Sudan Black B and Sudan Red 7B in food samples.

Sample	Concentration, ng g ⁻¹						
	Sudan I	Sudan II	Sudan III	Sudan IV	Para Red	Sudan Black B	Sudan Red 7B
Chili powder 1	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ
Chili powder 1 ^a	19.45 ± 0.99	19.39 ± 0.85	19.48 ± 0.85	18.99 ± 1.2	19.01 ± 1.15	19.45 ± 0.86	19.06 ± 0.75
Chili powder 1 ^b	38.92 ± 1.1	39.09 ± 1.25	39.92 ± 1.31	38.99 ± 0.75	39.60 ± 1.25	39.31 ± 1.21	39.65 ± 1.25
Chili powder 2	< LOQ ^c	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ
Chili powder 3	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ
Chili powder 4	31.21 ± 1.6	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ
Chili powder 4 ^b	70.67 ± 5.42	38.67 ± 2.12	39.20 ± 3.1	38.99 ± 1.70	39.65 ± 2.43	39.99 ± 3.23	38.56 ± 2.01
Chili tomato sauces	< LOQ	< LOQ	41.5 ± 2.2	< LOQ	< LOQ	< LOQ	< LOQ
Chili tomato sauces	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ
Ketchup	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ
Ketchup	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ

^a Spiked with required amount of standard to give final concentrations as 20 ng g⁻¹ of Sudan I, II, III, IV, Para Red, Sudan Black B and Sudan Red 7B.

^b Spiked with required amount of standard to give final concentrations as 40 ng g⁻¹ of Sudan I, II, III, IV, Para Red, Sudan Black B and Sudan Red 7B.

^c Lower than LOQ.

nanomaterials, problems with traditional extraction procedures were overcome. Time-consuming procedures, such as filtration and centrifugation on SPE, were eliminated, meaning the analysis of large numbers of food samples could be achieved. cMWCNT- γ -Fe₂O₃ magnetic nanomaterials are expected to be widely applicable for detecting other trace organic pollutants in different samples.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.foodchem.2017.09.039>.

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