



Rapid Magnetic Solid-Phase Extraction Based on Graphene Oxide/Magnetite Nanoparticles for the Determination of Non-Steroidal Anti-Inflammatory Drugs and Bisphenol-A in Tap Water

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A simple and rapid magnetic solid-phase extraction based on reduced graphene oxide/ferroferic oxide (rGO/Fe₃O₄) coupled with high performance liquid chromatography-diode array detector (HPLC-DAD), was proposed for the determination of naproxen and diclofenac sodium of non-steroidal anti-inflammatory drugs (NSAIDs) and bisphenol-A residue in tap water sample. Fe₃O₄ was assembled with graphene oxide by facile method. Several important parameters, such as extraction time, desorption time and desorption solvent were studied and optimized. Optimal conditions were then used for the extraction of NSAIDs and bisphenol-A from real samples, prior to HPLC-DAD analysis using C₁₈ column. The proposed method provides good linearity in the range of 1.0-5.0 mg L⁻¹ (R²_{naproxene} = 0.9983, R²_{diclofenac} = 0.9960) for NSAIDs and R² = 0.9919 for bisphenol-A; limit of detection of 0.031 mg L⁻¹ and 0.023 mg L⁻¹ for naproxen and diclofenac, respectively and 0.1785 mg L⁻¹ for bisphenol-A; limit of quantification for naproxen and diclofenac were 0.102 and 0.076 mg L⁻¹, respectively and 0.5949 mg L⁻¹ for bisphenol-A; relative recoveries between 66.21 and 105.60 % for diclofenac and naproxen, respectively and 72.74 % for bisphenol-A; and acceptable reproducibility relative standard deviation (RSD, n = 3) of 6.79 and 11.32 % for diclofenac and naproxen, respectively and 0.93 % for bisphenol-A. The method was successfully applied to tap water sample and the results indicates that small residue of bisphenol-A is present the sample while no traces of NSAIDs was detected.

Keywords: Magnetite graphene oxide, Naproxen, Diclofenac, Bisphenol-A, Adsorption.

INTRODUCTION

Magnetic solid-phase extraction (MSPE) is a new method where it employed the magnetic material as a sorbent and an application of external magnetic field, for the preconcentration of the target analyte from the large volume of sample [1]. The major advantage of MSPE is simplicity, avoiding the use of filtration and centrifugation steps as in traditional solid-phase extraction (SPE), which subsequently promotes faster analysis [2,3]. Nowadays, the use of carbon nanoparticles sorbent in SPE is becoming more prevalent in analytical chemistry. One of the most common carbon nanoparticles used is graphene oxide (GO).

Graphene oxide (GO) is a single layer atoms of carbon with functional groups containing oxygen in variable ratios attached to both planes and edges of the flake. This excellent sorbent material received its popularity due to its high thermal and chemical stability, large surface area, good adsorption

capacity, high hydrophobicity and good acid/alkali resistance [4]. To date, graphene oxide has been used extensively to develop new microextraction method for the extraction and purification of analyte such as pesticide, organic contaminant or metal ions from various sample [5,6]. In this study, graphene oxide was combined with ferroferic oxide (Fe₃O₄) to form a new magnetic nanosorbent; rGO/Fe₃O₄. Fe₃O₄, commonly called as magnetite, is the most acknowledged inorganic magnetic material as it is inexpensive, biocompatible and easy to synthesize [4]. Owing to its magnetic properties, this newly synthesized magnetic nanosorbent can be easily separated from a medium using a magnetic field. Hence, this nanosorbent was expected to provide an efficient extraction technique for the analysis of the selected nonsteroidal anti-inflammatory drugs (NSAIDs) and bisphenol-A in tap water.

NSAIDs are among the most regularly consume medications worldwide. Over the past 50 years, NSAIDs have become

a choice by western community as a type of pain reliever that treat various symptom such as pain, inflammation and stiffness [7]. The extensive use of NSAIDs occur by the availability of these products in over the counter settings, such as petrol station and supermarket which lead to the dangerous public opinion of NSAIDs as safe and carrying trivial risk. While it has long been known that NSAIDs have possibility to cause harm to gastrointestinal tract including nausea, vomiting and diarrhoea, there are several reports from high level publications which had proved that these heavily consumed drugs are not as safe as may be perceived by the public [8]. Thus, in order to protect the health of public community, the maximum residue limits (MRL) for NSAIDs have been defined by Commission Regulation (EU) 37/2010 for a range of food products.

Accordingly, bisphenol-A is a hazardous organic compound that has colourless crystalline solid which is belonging to the group of diphenylmethane derivatives and bisphenols, with two hydroxyphenyl groups. This material is broadly use in the manufacture of polycarbonate plastics and epoxy resins which can normally be found in water bottles, food container, water supply pipes, kitchen utensils and other commercial product of common personal and industry use [9,10]. Bisphenol-A is known as endocrine disruptor which would interrupt regulation or development function of an organism [11]. Numerous studies have found that bisphenol-A exposure may cause several adverse health effects in human and animals such as liver damage, disruption of multiple metabolic mechanisms, disruption of thyroid hormone and obesity promoting effect [12,13]. For this reason, U.S Environmental Protection Agency has established the recommendation acceptable dose of bisphenol-A in tap water at 0.05 mg L⁻¹ per day [14].

The introduction of rGO/Fe₃O₄ as a sorbent has several advantages over conventional solid phase extraction as it avoided time consuming and tedious on column SPE procedures. Instead, it will provide a fast, simple, economical and environmentally friendly method for NSAIDs determination in water samples. According to Xiao *et al.* [15] because of its extremely high surface area, excellent chemical stability and great thermal stability, graphene oxide is being used as the sorbent in sample preparation. In addition, in MSPE, magnetic Fe₃O₄ nano-particles are usually coated the graphene sorbent to create magnetic graphene oxide composite for better separation after adsorption. Fe₃O₄ material are the most regularly used as they are low toxicity, has good biocompatibility and easy to prepare [16]. Thus, the aim of the study is to prepare, optimize and validate MSPE using rGO/Fe₃O₄ nanoparticles sorbent coupled with high performance liquid chromatography-diode array detector (HPLC-DAD) for the quantification of NSAIDs, namely naproxen and diclofenac and bisphenol-A in tap water.

EXPERIMENTAL

Graphene oxide (GO) with purity greater than 99 % purchased from graphene oxide Advanced Solutions Sdn. Bhd (Selangor, Malaysia). Fe₃O₄, FeCl₂·4H₂O, nitric acid, methanol, isopropanol which all were of analytical grade were obtained from Sigma-Aldrich, (USA). HPLC grade acetonitrile (ACN) organic solvent was obtained from Merck KGaA (Darmstadt, Germany). Sodium acetate powder was obtained from Sigma-

Aldrich, (USA). NSAIDs standard; naproxen and diclofenac sodium salt (> 95 % purity) and bisphenol-A were obtained from Sigma-Aldrich, (USA).

Preparation of rGO/Fe₃O₄ magnetic nanoparticle (MNP): rGO/Fe₃O₄ was prepared using a facile method that has been previously employed in published literature with little modification [17]. Graphene oxide nanosheets (25 mg) was dispersed in 20 mL deionized water through ultrasonication for 3 h. Next, 25% of NH₃ solution was added by drops to make the pH solution become 11. The rGO/Fe₃O₄ nanoparticle were synthesized at 1:5 ratio, then 125 mg of FeCl₂·4H₂O was added slowly to the mixture with continuous magnetic stirring. A black suspension of rGO/Fe₃O₄ nanocomposite was obtained after 3 h of stirring. The rGO/Fe₃O₄ was separated by magnetic separation, washed with water several times and the residue was dried in an oven for further use.

Preparation of stock and working solutions: A stock solution of 1000 mg L⁻¹ of each standard was prepared by weighing 0.01 g of each analytes and diluted in 10 mL of methanol. Then from the stock solutions, a 100 mg L⁻¹ of standard mixture was prepared to be used in the subsequent experiments. Incidentally, a series of working solutions were prepared by a serial dilution of the standard mixture with methanol. To make a calibration curve in the extraction procedure, spiked samples at final concentration of 1 mg L⁻¹ were prepared by adding 1 mL of standard solution into 9 mL of water.

Extraction of NSAIDs and bisphenol-A: The extraction of NSAIDs and bisphenol-A compounds began by dispersing 20 mg of rGO/Fe₃O₄ nanoparticles in 10 mL of spiked water sample at pH 3. pH of the spiked water sample was adjusted to 3 by adding some HCl prior to the extraction. The mixture undergoes agitation by vortex to allow the diffusion of analyte through the sorbent. The optimal extraction time was 30 min. After that, the magnetic adsorbent was separated from the sample solution by using a magnet and the supernatant liquid was decanted. The magnetic sorbent was put in a 3 mL of desorption solvent and sonicated for 10 min to desorb the analyte from the adsorbent. The samples containing analyte was separated from the magnetic sorbent by using a magnet. The sample then blew with nitrogen to preconcentrate until 1 mL and filtered through PTFE syringe filter (0.45 μm). Finally, 20 μL of the solution was injected into HPLC-DAD for quantitative analysis. The schematic diagram for this method apparatus is shown in Fig. 1. In this study, several factors such as extraction time, desorption time and desorption solvent were studied and optimized to ensure the high sensitivity, precision and accuracy of the magnetic solid-phase extraction process by changing one factor at a time. Several parameters namely extraction time, desorption time and desorption solvent were studied to optimized the MSPE procedure. The extraction performance of rGO/Fe₃O₄-MSPE in determination of NSAIDs and bisphenol-A was compared with other type of sorbent from previous literature. The optimum condition of the MSPE process was applied to the subsequent analysis.

The chromatographic analysis of NSAIDs and bisphenol-A was performed using Agilent Technologies 1200 series HPLC system (Agilent Technologies, Germany), coupled with

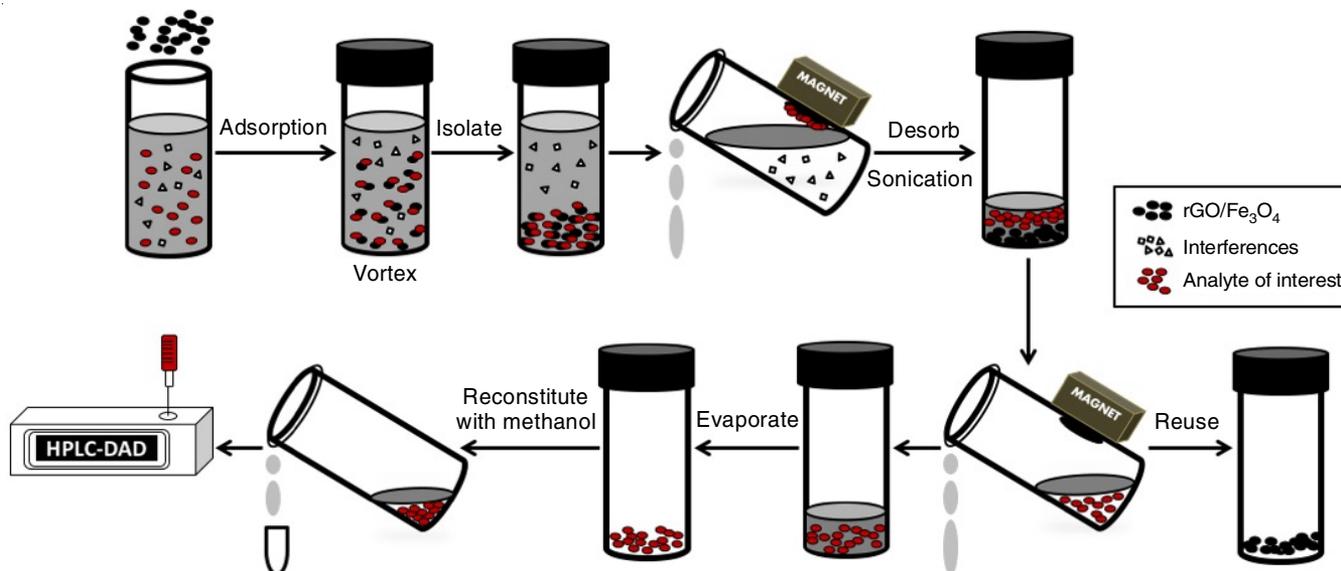


Fig. 1. A schematic diagram of the extraction procedure using rGO/Fe₃O₄ nanocomposite and analysis by HPLC-DAD instrument

diode array detector (DAD). The chromatographic separation of NSAIDs was carried out on an analytical column, which is VertiSep™ USP C18 column (5 μm particle size, 250 mm × 4.6 mm I.D.) (Vertical Chromatography Co., Ltd., Thailand) at 28 °C. For NSAIDs, the isocratic mobile phase, which is acetonitrile (A) and acetate buffer (B) were used at the composition of 60:40 (A:B). Meanwhile, the isocratic mobile phase for bisphenol-A, which is acetonitrile (A) and water (B) was also programmed at the composition of 60:40 (A:B). The detection wavelength, flow rate and injection volume were fixed at 220 nm (sample) and 360 nm (reference), 1.0 mL min⁻¹ and 20 μL. The chromatographic system was cleaned and stabilized to initialize the analysis. The quantification of NSAIDs and bisphenol-A was determined by measuring the peak area (mAU) of the chromatogram. Coefficient of determination were determined by constructing a calibration curve between the peak areas and concentrations of both of the NSAIDs and bisphenol-A standards.

Method validation: The analytical parameters such as linearity, limit of detection (LOD), limit of quantification (LOQ), accuracy and precision were determined to ensure the result of the analysis is reliable and the procedure is fit for the intended purpose. The linearity of the method was determined by plotting a calibration curve at five different standard concentrations. The precisions of the method were determined by a triplicate reading of each analyte ($n = 3$) and for the accuracies, the values were obtained in terms of recoveries where the sample was spiked with minute volume of standard solutions.

Sample preparation: The water sample was obtained from tap water in Lab M FR B304, at Institute of Science, UiTM Shah Alam. 10 mL of tap water were taken for blank sample analysis and recovery test. Each sample were triplicated ($n = 3$). For recovery identification, each sample was spiked with NSAIDs standard mixture of naproxen and diclofenac and bisphenol-A at a final concentration of 1 mg L⁻¹. The solutions were prepared by adding 1 mL standard mixture into 9 mL of tap water. The samples that were not immediately analyzed were stored at 4 °C prior to the HPLC analysis.

RESULTS AND DISCUSSION

System performance: Appropriate HPLC-DAD conditions were optimized and established. The LC conditions were optimized to obtain a maximum sensitivity for identification of NSAID and bisphenol-A. A calibration curve of both of the NSAIDs (naproxen and diclofenac) and bisphenol-A standard at 5 different concentrations were constructed by plotting the concentration of analyte (mg L⁻¹) against the peak area (mAU), to obtain the linearity of the LC system performance. A good linearity was established where the coefficient of determination (R^2), for naproxen and diclofenac is obtained at 0.9851 and 0.9734, respectively and 0.9995 for bisphenol-A. The retention time for naproxen and diclofenac were observed at 4.8 and 6.4 min, respectively, while bisphenol-A was eluted at 4.1 min. Each standard was replicated to verify a good precision. The chromatograms of both of the NSAIDs and bisphenol-A indicate that a good separation was achieved (Fig. 2).

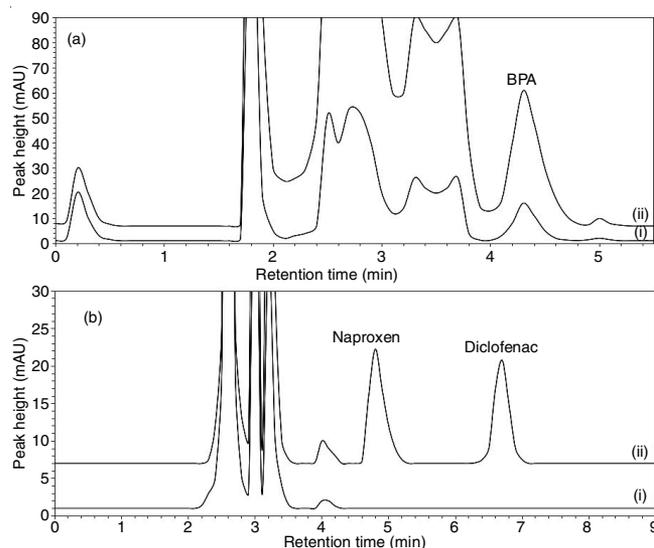


Fig. 2. Chromatogram for the extraction of (a) bisphenol-A and (b) NSAIDs in a (i) blank and (ii) spiked with NSAIDs at concentration of 1 mg L⁻¹ of tap water sample under optimum condition

Preparation of graphene oxide/Fe₃O₄ magnetic nanoparticle sorbent: In this study, Fe₃O₄ were combined with GO to support and strengthen the material to produce a stable magnetic sorbent. Graphene and graphene oxide (GO) has become the most favoured and effective sorbent in analytical chemistry since it has very large surface area, which is 2630 m² g⁻¹ [18,19]. Graphene is hydrophobic which determine its function in water system. However, its property can be modified by oxidize graphene into GO to increase its solubility in water. The presence of carboxyl and hydroxyl functional groups on the surface of hydrophilic GO make it bind with other substances specifically with drug substances. Fe₃O₄ has good magnetic properties and great biocompatibility to be used in biological application. The rGO/Fe₃O₄ nanocomposite with high superparamagnetism, were synthesized by facile method. The combination of GO and Fe₃O₄ provides extra advantages that cannot be achieved by each material on its own. A simple chemical reaction that bind these two materials in preparation of the sorbent was illustrated in Fig. 3. In this process, GO was reduced to rGO and act as an oxidizing agent, which would simultaneously aid in the oxidation of Fe²⁺ to Fe³⁺. The Fe³⁺ ions reacted in alkaline condition, also with -OH group from the GO surface, were then deposited on the surface of rGO sheets as Fe₃O₄.

Optimization of MSPE procedure: Several parameters that affect the extraction efficiency of rGO/Fe₃O₄ sorbent were studied in depth to obtain a shorter analysis of time with maximum extraction efficiency. The parameters chosen for the optimization method were extraction time, desorption time and desorption

solvent. In this procedure, deionized water was spiked with NSAIDs mixture (naproxen and diclofenac) and bisphenol-A at a final concentration of 1 mg L⁻¹ to simulate the extraction of these analyte from real sample.

Effect of extraction time on the extraction efficiency:

The effect of the extraction time was studied and optimized in order to obtain the most suitable extraction time to give the highest extraction efficiency of the selected analytes. The extraction time chosen for the optimization analysis was 10, 20, 30 and 40 min done by using a vortex at 1000 rpm. Extraction time is one of the important parameters as it represents the equilibrium time required for the target compound to be adsorbed on the adsorbent. The extraction time will increase if the adsorption equilibrium between the adsorbent and the target analyte is not reached. Fig. 4a indicates that the extraction efficiency of both naproxen and diclofenac analyte increases significantly as the extraction time increase, but after 30 min the extraction recovery of the analyte decline. Meanwhile, the adsorption equilibrium could not be reached if it is less than 30 min as the whole system has not enough time to reach equilibrium. Incidentally, the adsorption of bisphenol-A increased significantly at 15 min but showed no significant effect on the extraction recovery after 15 min. Therefore, the extraction time was selected at 30 and 15 min in the subsequent experiment for both of the NSAIDs and bisphenol-A, respectively.

Effect of desorption time on the extraction efficiency:

The effect of desorption time on the elution of NSAIDs and bisphenol-A from the rGO/Fe₃O₄ sorbent is shown in Fig. 4b. Desorption time was another important optimization parameter,

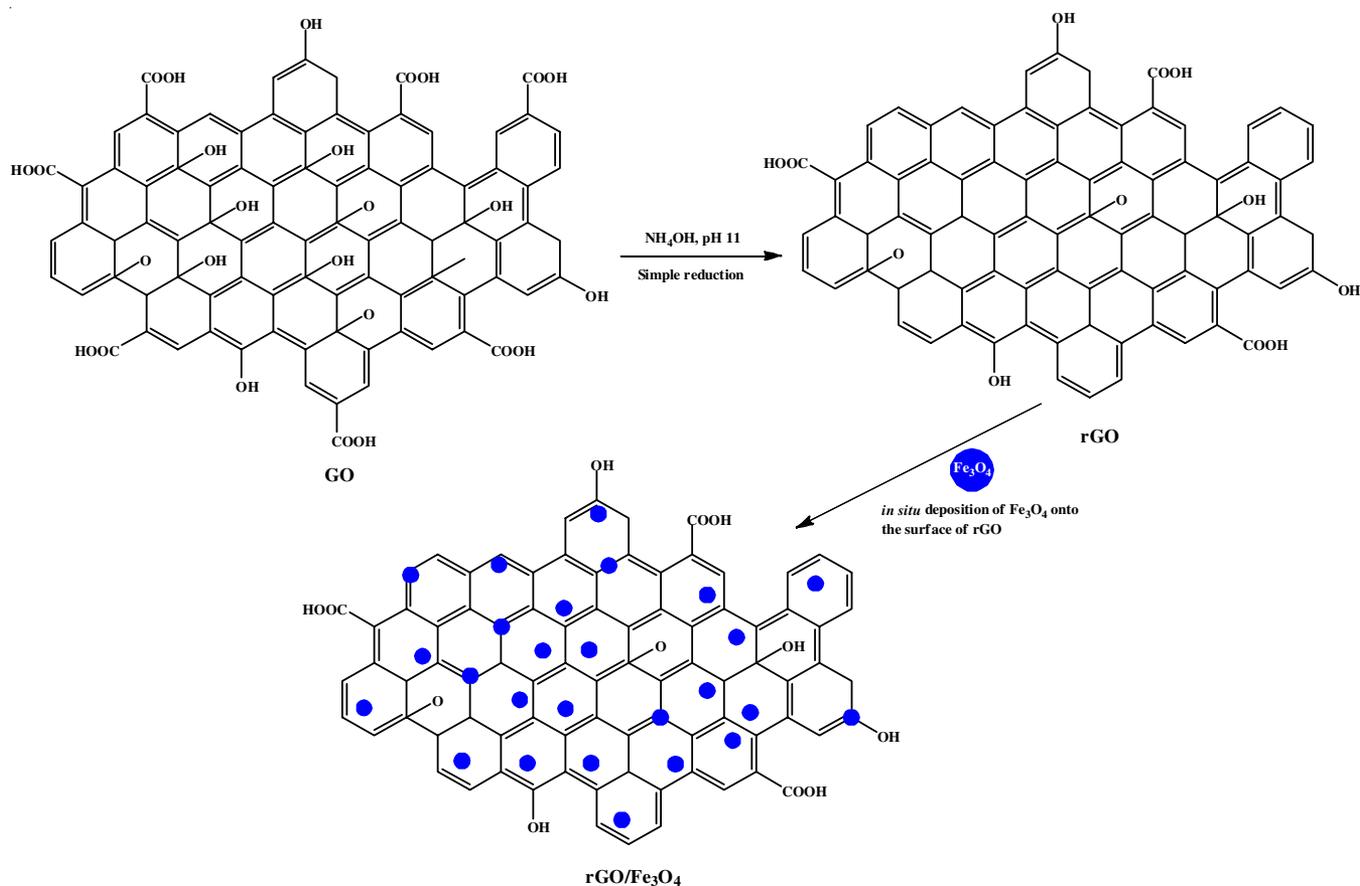


Fig. 3. Simplified chemical schemes of the formation of rGO/Fe₃O₄ nanocomposite

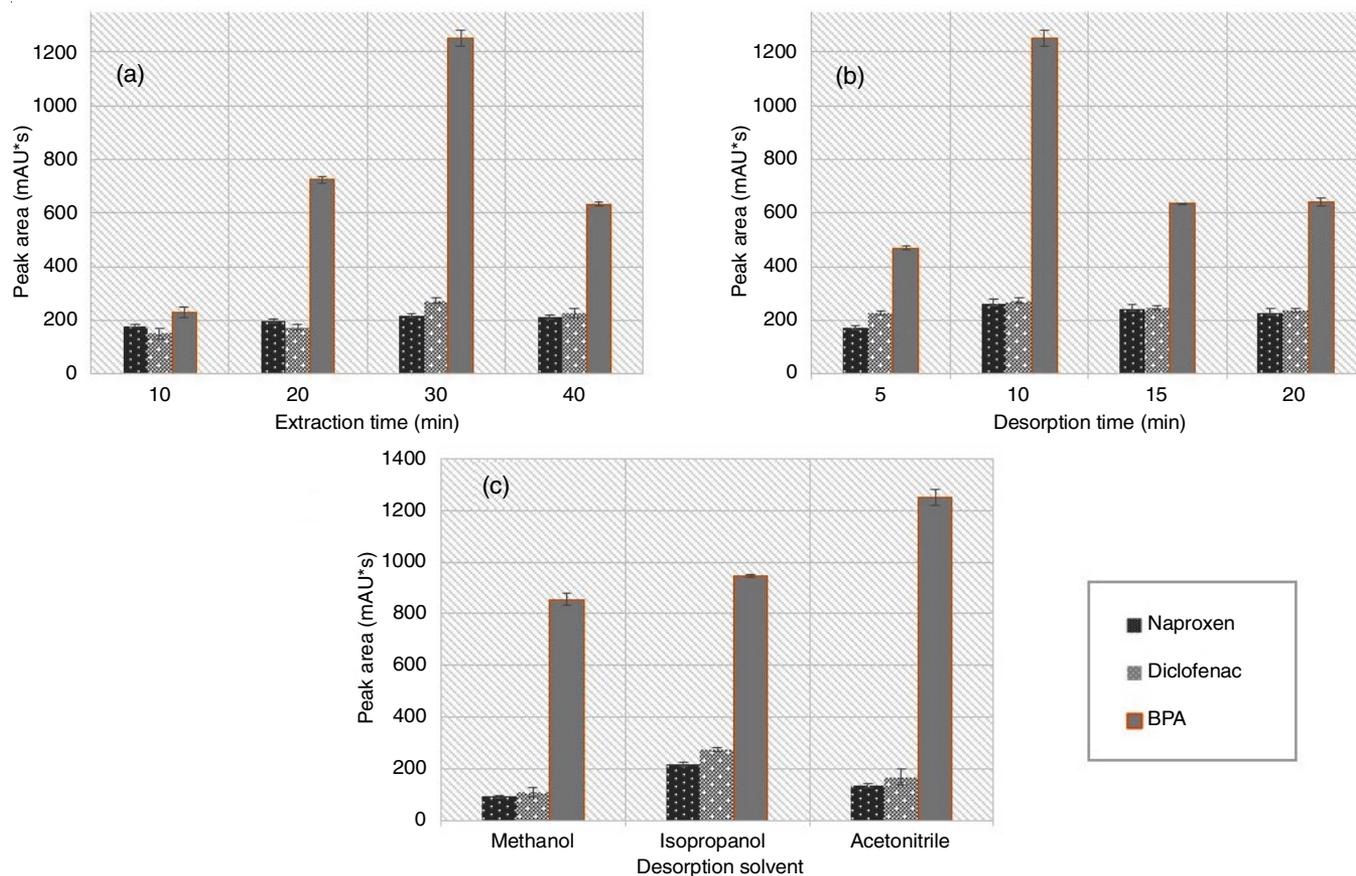


Fig. 4. Extraction performance of rGO/Fe₃O₄ with incremental effect of extraction time (a), desorption time (b) and desorption solvent (c) on the efficiency of rGO/Fe₃O₄-MSPE of NSAIDs and bisphenol-A from tap water

analyzed to shorten the sample preparation time, performed by ultrasonicate the sample at different period of times. Desorption time in the range of 5 to 20 min were studied to determine the minimum time required to elute the target compounds from the adsorbent. The results obtained indicate that the elution recoveries for both of the NSAIDs and bisphenol-A increased greatly between 5 to 10 min and subsequently showed no improvement after 10 min. The results indicate that 10 min is sufficient enough for the analyte desorption. Hence, 10 min was chosen as desorption time for both NSAIDs and bisphenol-A in subsequent experiment.

Effect of desorption solvent on the extraction efficiency:

Type of desorption solvents are also important as optimization parameter since it would affect the elution efficiency. 3 mL of desorption solvent were used to desorb the both NSAIDs (naproxen and diclofenac) and bisphenol-A from the sample matrix. The tested solvent used were isopropanol, acetonitrile and methanol. The use of a suitable solvent would elute a high recovery of target analyte. Fig. 4c illustrates that isopropanol exhibits stronger desorption ability of both naproxen and diclofenac compound compared to methanol and acetonitrile,

while acetonitrile exhibits stronger desorption ability than methanol and isopropanol for the bisphenol-A. Therefore, isopropanol and acetonitrile were selected as the optimal desorption solvent in the following experiment for both of the NSAIDs and bisphenol-A, respectively.

Method validation: Several criteria namely linearity, LOD, LOQ, precision and accuracy were assessed to evaluate the performance of the developed rGO/Fe₃O₄-MSPE method for the extraction of the selected NSAIDs and bisphenol-A from tap water. The results obtained are tabulated in Table-1. The linearity of the developed method was achieved in the range of 0.1 to 5.0 mg L⁻¹ ($R^2_{\text{naproxene}} = 0.9983$, $R^2_{\text{diclofenac}} = 0.9960$) and 0.05 to 3.0 mg L⁻¹ ($R^2 = 0.9919$) for both of the selected NSAIDs and bisphenol-A, respectively. The LOD for both of the NSAIDs, which are naproxen and diclofenac and bisphenol-A were found to be 0.031 and 0.023 mg L⁻¹ and 0.1785 mg L⁻¹, respectively. The LOQ were 0.102 and 0.076 for naproxen and diclofenac, respectively and was 0.5949 mg L⁻¹ for bisphenol-A. The precision is determined by the relative standard deviation (RSD) of the three spiked deionized water at the lowest concentration, which was 0.1 mg L⁻¹ for both naproxen

TABLE-1
ANALYTICAL PERFORMANCE OF THE DEVELOPED METHOD

Analytes		Linearity (mg L ⁻¹)	Correlation of coefficient (R ²)	LOD (mg L ⁻¹)	LOQ (mg L ⁻¹)	RSD (%)
NSAIDs	Naproxen	0.1-5.0	0.9983	0.031	0.102	1.04
	Diclofenac	0.1-5.0	0.9960	0.023	0.076	0.67
Bisphenol-A		0.05-3.0	0.9919	0.1785	0.5949	17.28

and diclofenac and 0.05 mg L⁻¹ for bisphenol-A. The RSD obtained for naproxen and diclofenac is 1.04 and 0.67 %, respectively and is 17.28 % for bisphenol-A.

Real sample analysis: Under the optimized condition, the proposed rGO/Fe₃O₄ sorbent MSPE coupled with HPLC-DAD technique were successfully applied to the tap water sample collected in UiTM Shah Alam for determination of the selected NSAIDs (naproxen and diclofenac) and bisphenol-A. The results exhibited both NSAIDs compound residues were absent in the tap water, while traces of bisphenol-A can be found at the concentration of 0.4841 mg L⁻¹, which is above the recommended exposure limit established by the U.S. Environmental Protection Agency. Acceptable recoveries of naproxen and diclofenac is ranged between 66.21 and 105.60 % while bisphenol-A is 72.74 %. Excellent RSDs were achieved for both of the NSAIDs which are 6.79 and 11.33 % for diclofenac and naproxen, respectively and bisphenol-A which is 0.93 %. The accuracy and precision of the sample analysis are tabulated in Table-2.

Analytes	Concentration (mg L ⁻¹)	Relative recovery (%)	RSD (%)
NSAIDs	Naproxen	ND	11.36
	Diclofenac	ND	6.79
Bisphenol-A	0.4841	72.74	0.93

*ND: Not detected

Comparison with other reported methods: The performance of the proposed rGO/Fe₃O₄MSPE method was compared with other recent literature for detection of NSAIDs and bisphenol-A residue in term of LOD, recovery and RSD. The results were summarized in Table-3. Although the LOD and RSD are higher than that of other established method, the developed rGO/Fe₃O₄MSPE could achieve desirable recoveries and precision which were comparable with other listed methods such as SPE, SPME, on-line and off-line SPE and IL-DLLME. It can be concluded that high relative recovery and good precision were accomplished by the developed method. The developed MSPE method was much simpler than other previous reported methods. Besides that, it provides a high

extraction efficiency due to its large surface area and has great reusability and reduces the analysis time where it simplifies the long steps in traditional procedure. Therefore, this developed MSPE method is proved to be sensitive, cost-effective and environmental-friendly in determination of NSAIDs and bisphenol-A in tap water.

Conclusion

In summary, the magnetic nanocomposite sorbent, rGO/Fe₃O₄ nanoparticle, was successfully prepared, optimized and applied to determine NSAIDs and bisphenol-A from tap water. The adsorption efficiency of NSAIDs and bisphenol-A onto the prepared sorbent can be attributed to the π - π interaction between the analyte and the graphene oxide in the sorption material. The sorbent could be easily separated from the sample matrix using an external magnet, thus, avoiding the need to centrifuge and preventing the sorbent to leech into the environment, which could possibly cause secondary pollution. Additionally, the proposed method is in line with the concept of green chemistry as it minimizes the use of organic solvents and due to the reusability of the sorbent [26]. rGO/Fe₃O₄ nanoparticle could potentially be used as an alternative adsorbent for trace analysis of NSAIDs and bisphenol-A in contaminated water.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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Sample	Method	Analytes	Detection method	LOD (µg L ⁻¹)	RSD (%)	Recovery (%)	Ref.
Tap water	MSPE	Naproxen, Diclofenac, bisphenol-A	HPLC-DAD	23-31	6.8-11.3	66.2-105.6	This study
Biological fluids	DLLME	Ketoprofen, Naproxene, Diclofenac, Ibuprofen	HPLC-DAD	0.1-6.0	2.8-8.1	36-86	[20]
Environmental water	Off-line SPE, On-line SPE	Diflunisal, Diclofenac, Fenoprofen, Ibuprofen, Naproxen, Tolmetin	HPLC-DAD, LC-MS	0.7-0.94	< 5	96-109	[21]
Water	IL-DLLME	Ketoprofen, Ibuprofen, Diclofenac	HPLC-DAD-FD	0.017-0.095	2-3	89-103	[22]
Water	d-SPE	bisphenol-A	HPLC-UV	0.3	2.5-4.5	95.0-106.2	[23]
Water	SPME	bisphenol-A	SPME-SERS	1	14.0	97-110	[24]
Drinking water treatment plants	SPE	bisphenol-A, MCBPA, DCBPA, TCBPA, TeCBPA	UPLC-ESI(t)-MS/MS(MRM)	0.001-0.03	≤ 10.00	102-109	[25]

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