

Analytical Methods

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Determination of 10 photo-initiators residues in food plastic packaging by gel permeation chromatography extraction coupled with gas chromatograph-mass spectrometry

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An analytical method using gel permeation chromatography (GPC) coupled with gas chromatography-mass spectrometry (GC-MS) was established for the determination of 10 photo-initiators - Benzophenone (BP), 2-Methyl Benzophenone (2-MBP), 1-Hydroxycyclohexyl phenyl ketone (Irgacure 184), 3-Methyl Benzophenone (3-MBP), 4-Methyl Benzophenone (4-MBP), Ethyl 4-dimethylaminobenzoate (EDB), 2,2-Dimethoxy-2-phenylacetophenone (Irgacure 651), o-Methyl-benzoyl Benzoate (OMBB), 2-Ethylhexyl-4-dimethylamino Benzoate (EHA) and 2-Isopropylthioxanthone (ITX) - in plastic food packaging. The plastic samples were treated with ultrasound in cyclohexane/ethyl acetate (1:1, v/v). The extract was cleaned up by gel permeation chromatography. The final solution was subjected to GC-MS using an external standard method for quantitation under selected ion monitoring mode. The 10 target photo-initiators showed linear recovery relationships over the concentrations of 0.005 to 0.2 mg/L, with $R^2 > 0.999$. The recoveries from spiked samples (0.07×10^{-3} , 0.17×10^{-3} and 0.33×10^{-3} mg/dm² of standard) for each of the 10 photo-initiators ranged from 77.0%-120.6%, with relative standard deviations (RSD) of 0.5%-13.9%. The limits of detection (LOD) were from 0.03×10^{-3} mg/dm² to 0.12×10^{-3} mg/dm². The developed extraction method is simple and yields a sufficiently pure test sample, while the analytical method meets the sensitivity and precision requirements for photo-initiator detection. This method could be routinely used to determine the levels of photo-initiator residues in plastic food packaging.

Introduction

Photo-initiators are one of the key components of UV-cured inks used for labeling food cartons. These light-sensitive reaction initiators work through a free-radical mechanism and promote rapid drying of the ink, leading to a faster curing process.¹ Due to the many advantages of UV-cured inks, such as high production efficiency, energy savings, and low environmental pollution,²⁻³ they are widely used in the printing of food packages. Several recent studies have indicated that residual photo-initiators from the ink on

the package surface have the potential to migrate into food,⁴⁻⁶ resulting in food contamination and endangerment of human health. In 2005, the Federal Institute for Risk Assessment (BfR) recommended that the **specific migration limit (SML)** be set at 0.5 mg/kg for the photo-initiators Isopropylthioxanthone (ITX) and 2-Ethylhexyl-4-dimethylamino Benzoate (EHA) in food contacting materials.⁷ In 2009, the Standing Committee on the Food Chain and Animal Health (SCFAH) declared that the overall migration limit must be less than 0.6 mg/kg for Benzophenone (BP) and 4-Methyl Benzophenone (4-MBP) in food packaging materials.⁸ In 2010, the European Printing Ink Association (EuPIA) published a clear migration limits for 105 kinds of photo-initiators.⁹ With recent technological advances, the numbers and varieties of photo-initiators used in UV-cured inks have increased considerably. In addition, a variety of photo-initiators are often combined to improve the curing rate and product quality. Thus, it is important to develop effective analytical methods for simultaneous determination of several photo-initiators in food packaging materials.

Currently, the main methods used for detecting photo-initiators include HPLC,¹⁰⁻¹¹ liquid chromatography-mass spectrometry,¹²⁻¹³

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gas chromatography-mass spectrometry,¹⁴⁻¹⁵ and liquid chromatography-tandem mass spectrometry.¹⁶⁻¹⁷ Due to the complex matrix of plastic packaging materials, pre-processing methods such as conventional liquid-liquid extraction or solid phase extraction are unable to completely remove the interfering components.

technique for detection of photo-initiators in plastic food packaging materials has yet to be reported.

In this study, ten widely used photo-initiators selected (Fig. 1, Table 1) were purified by GPC and then analyzed by gas chromatography-mass spectrometry (GC-MS). This method is shown to be fast, accurate, and highly sensitive, thus providing a useful and practical technique to detect photo-initiators in plastic packaging materials.

Table 1 The specific migration limit (SML) of 10 photo-initiators.

Compound	MRL (mg/kg)
BP	0.6*
2-MBP	0.6*
Irgacure 184	<0.01
EDB	0.05
3-MBP	0.6*
4-MBP	0.6*
Irgacure 651	<0.01
OMBB	0.05
EHA	2.4
ITX	0.05

*as sum of all these four benzophenone photo-initiators

Gel permeation chromatography (GPC) is widely used for sample purification, especially for samples that are rich in fats or pigments. The technique has excellent separation efficiency, can be automated, and can be applied to a broad variety of matrices and products.¹⁸ GPC is a widely used technique in large-molecule samples due to its effective removal of interfering components and has been used in the detection of pesticide residues and antioxidants in eggs,¹⁹ honey,²⁰ and pharmaceuticals products.²¹ The U.S. Environmental Protection Agency (EPA), the U.S. Food and Drug Administration (FDA), the American Association for Analytical Chemistry (AOAC) and the European Commission (EN) all consider GPC as an official method. However, the application of the GPC

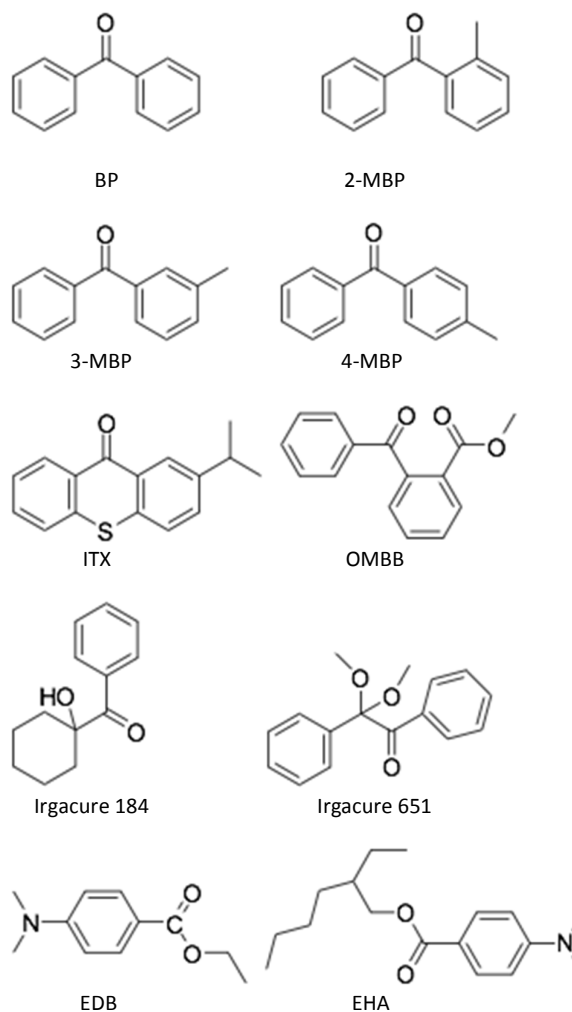


Fig. 1 Structures of ten photo-initiators.

Materials and methods

Standards

Photo-initiator standards for 2,2-Dimethoxy-2-phenylacetophenone (Irgacure 651, 99%), 4-Methyl Benzophenone (4-MBP, 99%), 2-

Ethylhexyl-4-dimethylamion Benzoate (EHA, 99%), 4-(Dimethylamino)-benzoic acid ethylester (EDB, 99%), Benzophenone (BP, 99%), 1-Hydroxycyclohexyl phenyl ketone (Irgacure 184, 99%), 2-Isopropylthioxanthone (ITX, 99%), and o-Methyl-benzoyl Benzoate (OMBB, 99%) were supplied by Adamas Reagent Company (Shanghai, China) and for 2-Methyl Benzophenone (2-MBP, 99%) and 3-Methyl Benzophenone (3-MBP, 99%) were purchased from Sigma Aldrich (St. Louis, MO, USA). HPLC-grade cyclohexane, ethyl acetate, acetonitrile, hexane, and acetone were obtained from Tedia Corporation (USA).

Samples

Five types of plastic food packaging materials were obtained from the manufacturers: polypropylene (PP), polyethylene terephthalate/cast polypropylene (PET/PP), biaxially oriented polypropylene/cast polypropylene (BOPP/PP), biaxially oriented polypropylene/low density polyethylene (BOPP/LDPE), and biaxially oriented polypropylene/polyethylene terephthalate/cast polypropylene (BOPP/PET/PP). The samples included ones that were printed and ready to use and ones not yet printed with UV ink.

Sample preparation

Plastic food packaging materials were weighed (AB204-S, accurate up to 0.1 mg, Mettler Toledo, Switzerland) and 0.3 g was cut into 0.5 cm × 0.5 cm pieces and placed in a 10 mL test tube. After addition of 5 mL of either acetonitrile, cyclohexane-ethyl acetate (1:1, v/v), ethyl acetate, hexane, or acetone to the test tube, the mixture was subjected to ultrasonication twice for 30 min (CNC ultrasonic cleaner, Kunshan Ultrasonic Instrument Co., Ltd., China). The resulting supernatant was collected, filtered by 0.45 μm membrane, and then purified by the GPC before analysis by GC-MS.

Gel chromatographic conditions

GPC ULTRA gel permeation sample purification system (LCTech Corporation, Germany) was used for the raw sample processing. The GPC column was a 320 mm × 25 mm (inner diameter) glass

column packed with polystyrene divinyl benzene Bio-beads (S-X3) (200-400 mesh). The mobile phase was cyclohexane- ethyl acetate (1:1, v/v) at a flow rate of 4.5 mL/min. The sample injection volume was 5 mL. Initial samples were collected at 1 min intervals, and a total of 20 samples were collected.

GC conditions

The TSQ QUANTUM gas chromatography-mass spectrometer (Thermo Scientific, Waltham, MA, USA) was used for sample analysis. The GC column was a 50%-diphenyl-50%-dimethyl polysiloxane capillary column (30 m × 0.25 mm × 0.25 μm, Thermo Scientific, Waltham, MA, USA). The inlet temperature was set at 250 °C. The carrier gas was high-purity helium (99.999%) at the constant flow rate of 1 mL/min. All injections were split and the volume was 1 μL. The column temperature increased from 60 °C (1 min) to 200 °C at 40 °C/min, then increased at 4 °C/min to 280 °C, which was then held for 3.5 min.

MS conditions

Data were acquired in EI mode (energy at 70 eV, ion source temperature at 230 °C, transfer line temperature at 280 °C, solvent delay time at 6 min) using the selected ion monitoring (SIM) mode. The known physical and chemical properties as well as the mass parameters for the ten photo-initiators are shown in Table 2.

Standards preparation

Standards for the 10 photo-initiators were each weighed (0.1 g), and dissolved in acetonitrile in a brown 100 mL volumetric flask to prepare a 1 mg/mL stock standard solution. A mixed photo-initiator standard solution at 2 mg/L was prepared by adding 0.2 mL of each of the 10 standard stock solutions to acetonitrile in a brown 100 mL volumetric flask, which was stored at -20 °C in the dark. The mobile phase solvent mixture of cyclohexane-ethyl acetate (1:1, v/v) was used as the solvent to prepare a series of standard solutions by successive dilutions. The mixed standard solution was added to blank samples. Linear equations were fit using origin 8.1. The detection limit was calculated by 3 times the signal-to-noise ratio (S/N ≥ 3).

Table 2 MS parameters for 10 photo-initiators in SIM mode.

Nr	Compounds	CAS nr	Octanol-water partition Coefficient (logKow)	Retention time (min)	SIM ion (m/z)	Fragmentor (ev)	Time window (min)
1	BP	119-61-9	3.214	7.14	182.1*, 104.9, 77	70	6.00-7.80

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2	2-MBP	131-58-8	3.784	7.33	195.1* , 91 , 119	70	6.00-7.80	
3	Irgacure 184	947-19-3	2.175	7.43	99* , 81 , 77	70	6.00-7.80	
4	EDB	10287-53-3	2.511	7.57	193.1* , 148 , 164	70	6.00-7.80	
5	3-MBP	643-65-2	3.857	7.95	196.1* , 119 , 181	70	7.80-8.60	
6	4-MBP	134-84-9	3.874	8.21	196.1* , 119 , 181	70	7.80-8.60	
7	Irgacure 651	24650-42-8	4.75	9.65	151* , 105 , 77	70	8.60-10.40	
8	OMBB	606-28-0	2.608	11.23	163* , 240 , 209	70	10.40-17.00	
9	EHA	21245-02-3	5.41	14.06	165* , 148 , 277.3	70	10.40-17.00	
10	2-ITX	5495-84-1	5.113	18.10	254* , 239 , 281	70	17.00-29.00	

* Quantitative ion

Results and discussion

Optimization of extraction solvent

According to the logKow values in Table 1, the solubility of each photo-initiator is poor in water. Therefore, organic reagents were used as extraction solvents. The extraction efficiencies of acetonitrile, cyclohexane-ethyl acetate, ethyl acetate, hexane, and acetone were investigated. The mixed standard solution containing the ten photo-initiators was added into blank samples and then processed as described. The recovery results revealed that the extraction ability of the five organic solvents decreased in the following order: cyclohexane-ethyl acetate > acetonitrile > ethyl acetate > acetone > hexane (Fig. 2). Therefore, 1:1 cyclohexane-ethyl acetate was chosen as the extraction solvent in this study.

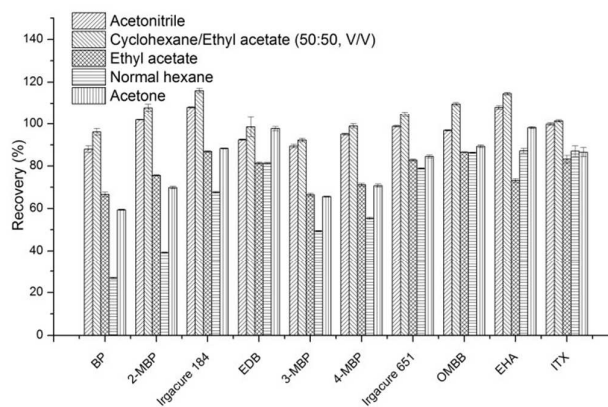


Fig. 2 Effect of different solvents on extraction and recovery of known amounts of 10 photo-initiators.

Optimization of GPC conditions

The ink on the surface of packaging materials is mainly composed of prepolymer resin, active monomer, additives and pigments, which can interfere with detection. GPC separates target and impurity molecules based on their relative molecular masses. However, the

structures and molecular masses of the ten photo-initiators are not similar. Hence, GPC was only used to purify samples from the matrix. MS analysis of the 20 samples collected at 1 min intervals demonstrated that no photo-initiators were detected in the first 4 min or in samples collected after 17 min, while photo-initiators were detected in the samples collected from 4 to 17 min. In addition, the photo-initiator recovery rate in the 4-17 min samples was 93.5-106.3%, which was in accordance with the testing requirements of photo-initiators. Therefore, the sample collection time was chosen to be 4-17 min. Most of the macromolecular impurities were collected in the first 4 min, which indicates that this purification method achieved effective separation of impurities and photo-initiators.

Purification results

The efficiency of GPC purification was examined. The samples were filtered by 0.45 μm membrane after ultrasound extraction. The filtrate was then divided into two parts: one part was directly analyzed by GC-MS while the other was purified by the optimized GPC and then analyzed by GC-MS. The results show that the response intensities of the hetero peaks in the GPC purified sample (Fig. 3b) were smaller than in samples without GPC purification (Fig. 3a). GPC purification effectively removed various impurities, which should improve the qualitative accuracy and quantitative sensitivity of the detection. Therefore, the combination of ultrasonic extraction and GPC purification was chosen as the appropriate preparation method.

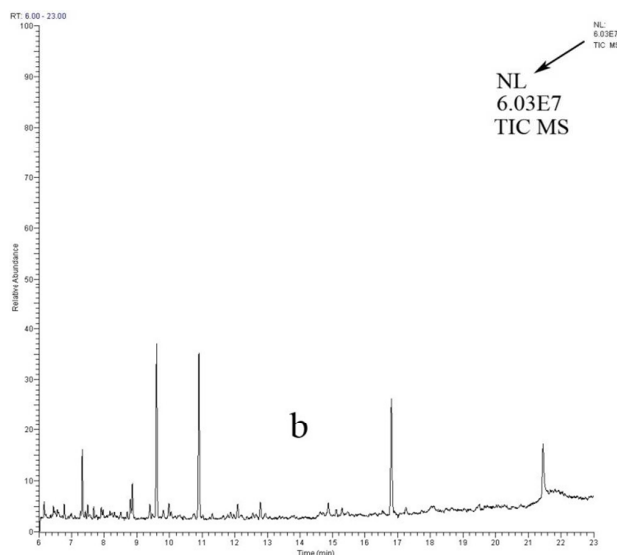
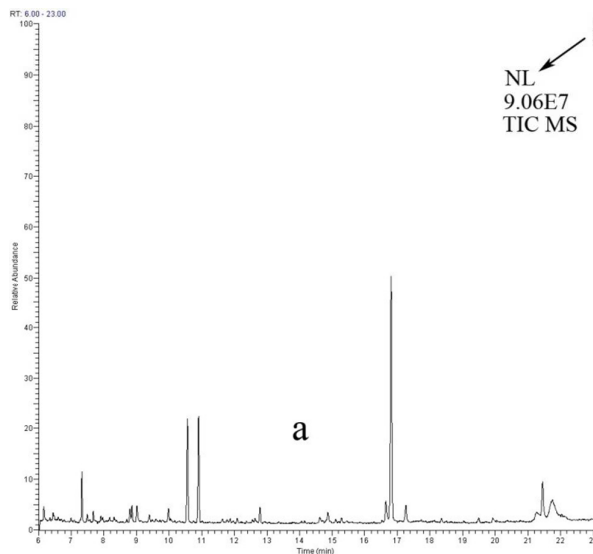


Fig. 3 Chromatograms obtained by the developed method in composite sample (trace a, after ultrasonic processing; trace b, after ultrasonic and GPC processing)

Methods Evaluation

Standard curve, linear range, correlation coefficient and detection limit. A series of mixed photo-initiator standard solutions was prepared at the following concentrations: 0.005, 0.01, 0.02, 0.05, 0.1, and 0.2 mg/L (for each standard). Standard curves were generated by plotting the peak area (y -axis) versus the mass concentration (x -axis). The blank sample for each plastic was used to determine the limit of detection (LOD). There was a linear correlation ($r^2 > 0.999$) between the response of each photo-initiator and its concentration in the concentration range of 0.005-0.2 $\mu\text{g/L}$ (Table 2). The LOD ranged from 0.03×10^{-3} - 0.12×10^{-3} mg/dm^2 .

Recoveries and relative standard deviation. The mixed standard solution was added into the blank samples at the concentrations of 0.07×10^{-3} , 0.17×10^{-3} and 0.33×10^{-3} mg/dm^2 in 1 mL solvent. The extraction began after volatilization of the solvent. Six samples were analyzed at each concentration in parallel (Table 3). The average recoveries of the 10 photo-initiators from the mixed standard solution were found to be 77.0%-120.6%, with relative standard deviations of 0.5% to 13.9%.

Determination of samples. The detection methods described above were used to investigate five different types of plastic food packaging materials, namely PP, OPP, PET, LDPE and composite membrane bags. The results indicated that 2-MBP, 3-MBP, Irgacure 184, and EHA were present in the sampled plastic food packaging materials (Table 4). The highest photo-initiator level in the practical samples was EHA in the BOPP/CPP sample, which was 2.64×10^{-2} mg/dm^2 . If 6 dm^2 of packing material is required to package 1 kg of food, and all the photo-initiator in the plastic

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packaging migrated into the food, the maximum amount of migration for EHA would be 0.16 mg/kg, which is below the SML of photo-initiators according to the EuPIA. Other photo-initiators were also below the SML of photo-initiators in EuPIA. The GC-MS chromatograms from the mixed standard solution and test samples are shown in Figure. 4. Two photo-initiators, Irgacure 184 and EHA, were clearly detected in the test samples.

Table 3 Linear equations, correlation coefficients (r^2) and limits of detection (LOD) of the ten PIs

Photo-initiator	Linear equation	r^2	LOD/(mg/ dm ²)
BP	$y=5.076 \times 10^6 x - 1.11 \times 10^4$	0.9992	0.03×10^{-3}
2-MBP	$y=1.022 \times 10^6 x - 1.01 \times 10^4$	0.9991	0.09×10^{-3}
Irgacure 184	$y=6.262 \times 10^6 x - 1.71 \times 10^4$	0.9992	0.12×10^{-3}
EDB	$y=4.346 \times 10^6 x - 1.09 \times 10^4$	0.9991	0.06×10^{-3}
3-MBP	$y=5.049 \times 10^6 x - 1.10 \times 10^4$	0.9992	0.06×10^{-3}
4-MBP	$y=4.439 \times 10^6 x - 1.36 \times 10^4$	0.9991	0.09×10^{-3}
Irgacure 651	$y=1.743 \times 10^7 x - 3.08 \times 10^4$	0.9993	0.06×10^{-3}
OMBB	$y=1.113 \times 10^7 x - 1.83 \times 10^4$	0.9994	0.06×10^{-3}
EHA	$y=1.525 \times 10^7 x - 2.27 \times 10^4$	0.9993	0.09×10^{-3}
ITX	$y=5.358 \times 10^6 x - 1.28 \times 10^4$	0.9992	0.06×10^{-3}

Table 4 Recoveries and relative standard deviations (RSDs) of the ten photo-initiators spiked at three levels in unprinted plastic packaging materials ($n=6$).

Photo-initiator	0.07×10^{-3} mg/dm ²		0.17×10^{-3} mg/dm ²		0.33×10^{-3} mg/dm ²	
	Recovery/%	RSD/%	Recovery/%	RSD/%	Recovery/%	RSD/%
BP	80	11.2	90.3	4.5	110.5	4.7
2-MBP	101.7	8.4	100.3	6.4	118.7	0.5
Irgacure 184	108.3	6.7	90.3	6.2	117	1.8
EDB	86.7	13.9	84.7	6.8	118	3.0
3-MBP	82.5	6.3	85.6	3.4	116.3	4.6
4-MBP	95.8	12	93.0	5.5	114.8	2.5
Irgacure 651	80.8	8.1	100.3	1.9	77.0	2.3
OMBB	115	3.9	91.3	3.8	118.5	0.8
EHA	120.6	6.1	114	1.8	118	4.2

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ITX	83.3	4.9	84.7	6.9	118.7	1.6

Table 5 Concentrations of the 10 photo-initiators in the consumer-ready plastic food packaging samples (mg/dm²)

sample	material	BP	2-MBP	Irgacure 184	EDB	3-MBP	4-MBP	Irgacure 651	OMBB	EHA	ITX
1	PE	ND	1.67×10^{-3}	4.76×10^{-3}	ND	0.43×10^{-3}	ND	0.13×10^{-3}	ND	0.91×10^{-3}	ND
2	PET/PPP	ND	0.77×10^{-3}	3.89×10^{-3}	ND	ND	0.27×10^{-3}	0.43×10^{-3}	ND	1.09×10^{-3}	ND
3	BOPP/PPP	0.23×10^{-3}	3.55×10^{-3}	5.66×10^{-3}	0.43×10^{-3}	0.91×10^{-3}	ND	ND	ND	2.64×10^{-2}	ND
4	BOPP/LDPE	0.90×10^{-3}	6.32×10^{-3}	0.23×10^{-3}	0.67×10^{-3}	1.09×10^{-3}	ND	ND	ND	2.66×10^{-3}	ND
5	BOPP/PET/PPP	ND	ND	1.43×10^{-3}	ND	ND	ND	ND	ND	1.66×10^{-3}	ND

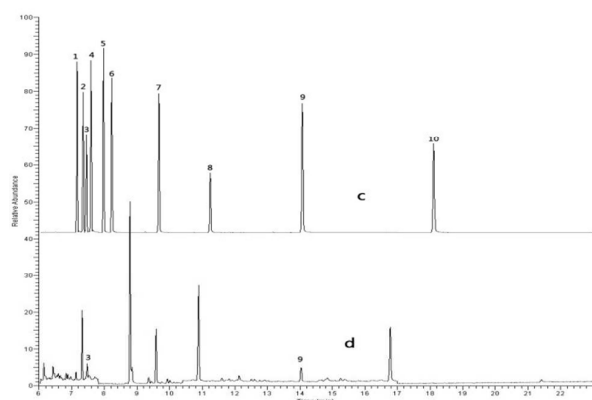


Fig. 4 Chromatograms obtained by the developed method in composite sample (trace c, blank plastic sample spiked at 200 µg/L level; trace d, real sample). For peak identification see Table 1.

Conclusions

This study established the feasibility of a GPC-GC-MS method to simultaneously measure ten photo-initiator residuals in plastic food packaging materials. In addition, five different kinds of packing materials, namely PP, PET/PPP, BOPP/PPP, BOPP/LDPE, BOPP/PET/PPP, were tested by this method. Residues of 4 photo-initiators (2-MBP, 3-MBP, Irgacure 184, and EHA) were found in consumer-ready materials, from where they could potentially migrate into food.

The method determined here is simple, accurate, easy to automate, and can be used to effectively monitor the commonly used photo-initiators in packaging materials. This method is a valuable tool for enabling future research in related areas.

Acknowledgements

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