Original Article
Food Science and Technology

ISSN 0101-2061 (Print)
ISSN 1678-457X (Online)

DOI: https://doi.org/10.1590/fst.14721

Determination of migrated formaldehyde from kitchenware using gas chromatography-mass spectrometry

Salman ALOTAIBI[1], Zeid Abdullah ALOTHMAN[2]*, Ahmad Yacine BADJAH[2], Masoom Raza SIDDIQUI[2], Saikh Mohammad WABAIDUR[2], Majed M. ALMUTAIRI[1], Mohammad Saad ALHUSSAIN[1]

Abstract

A new GC-MS method is developed for the determination of formaldehyde. Formaldehyde exposure to the food materials may lead to its migration into food. The formaldehyde is reported to be toxic to human health if taken or exposed in higher than recommended quantity. This papers reports analytical method to get the information about the content of the formaldehyde in the kitchenware. Prior to the determination process, simulation study was performed to get the information about the migration of formaldehyde. Method was applied to the real samples for formaldehyde determination. It has been observed that the all the simulated real sample witnessed migration of formaldehyde from the kitchenware. Alongside the validation study was performed too, which suggest that the method is linear in the concentration range of 2.5 to 30 mg l^{-1}. The precision study suggests a % RSD in the range of 0.568- 3.77 for all concentration points. The recovery study highlights an excellent recovery of the method which was found in the range of 97.64-99.43%. LOD and LOD of the method was found to be 0.35 and 1 ml l^{-1}, which is equivalent to 0.05 and 0.142 mg l^{-1} of formaldehyde, respectively.

Keywords: formaldehyde; determination; GC-MS; migration; simulation; analytical methods.

Practical Application: A method to get the information about the content of the HCHO in the kitchenware.

1 Introduction

In the field of polymer or material science, resins could be defined as viscous or solid substances either of plant or animal origin that can typically be converted into polymers. These resins are the basic materials for organic coating, lacquers and plastics (Horie et al., 2004). These resins are also converted into various products which are extremely useful for humans. Amongst the most used resin products is melamine formaldehyde or simply melamine. It is a thermosetting plastic material which is prepared from two precursor materials i.e. melamine (chemical formula $C_3H_6N_6$) and formaldehyde (chemical formula CH_2O) (Bauer, 1986). The chemical formula of resulting melamine formaldehyde is $C_4H_8N_6O$. One of the widely used application of this polymers is its application as kitchenware precursor material. Different varieties of melamine-formaldehyde kitchenware are widely available in the market. Melamine-formaldehyde resins are hard, very durable, and versatile thermosetting aminoplastics with good fire and heat resistance. These materials are rather inexpensive and they are also dishwasher friendly and thus are popular materials which can be used by all age groups. Additionally, melamine kitchenware does not impart its odor and taste to the food stuffs that comes in contact with the beverage or the food stuffs. These properties make melamine-formaldehyde a widely used material for production of kitchenware throughout the world (Brydson, 1999). Eyeing on the customers' demand of melamine formaldehyde made kitchenware, many companies are involved in the production of melamine-formaldehyde products. Although there are several manufacturing units which follow proper norms, use high quality raw materials and release quality assured products into the market, yet there are many production units which do not follow proper production norms and use substandard raw materials for melamine kitchenware production. This practice makes some of the finished products risk of releasing chemicals which are used during the synthesis, one of them is formaldehyde, which may diffuse into the food that are served or stored in these type of kitchenware. There are several studies highlighting the fact that this migration happens at acidic pH and at certain temperatures (Tyan et al., 2009; European Food Safety Authority, 2010; Ebner et al.,2020).

Formaldehyde is classified as human carcinogen by International Agency for Research on Cancer (IARC) (International Agency for Research on Cancer, 2018); Li et al., 2008). Besides IARC, U.S. Department of Health and Human Services, also listed formaldehyde as "known to be carcinogen" in its Twelfth Report on Carcinogens in 2011 (National Toxicology Program, 2016). Thus, food we take for our health may turn into toxic substances when taken regularly in melamine-formaldehyde utensils and in amounts which exceeds the permissible limits. Commission Regulation (EU) No. 10/2011 (Article 3(1)(b) of Regulation (EC) 1935/2004) which is concern about the quality of the food contact materials, recommends that the plastic food contact materials should not release 10 mg of substance per 1 dm^2 surface area of

Received 11 Mar., 2021
Accepted 24 Apr., 2021
[1] *Saudi Standards, Metrology and Quality Organization, Al Imam Saud ibn Abdul Aziz Road, Al Mohammadiyah, Riyadh, Saudi Arabia.*
[2] *Advanced Materials Research Chair, Chemistry Department, College of Science, King Saud University, Riyadh, Saudi Arabia.*
*Corresponding author: zaothman@ksu.edu.sa

plastic material and overall migration limit (OML) of 60 mg/kg of total constituent of released food or simulant, and specific migration limits (SML) for migration of formaldehyde into foods of 15 mg/kg food the can be equivalent to 2.5 mg/dm^2 (European Union, 2011).

As mentioned above, the plastics made from the polymerization of urea/melamine with formaldehyde are widely used throughout the world in the preparation of food contact articles and kitchen utensils. There is a possibility that these food contact, plastics and kitchen utensils may release chemicals once they attain stress conditions. For this several methods were developed and reported in the literature, some methods reported the presence of melamine while other reported formic acid other reported both of them. High performance liquid chromatography (HPLC) was used to study the migration of melamine from the kitchenware and food packaging materials (Ibarra et al., 2016; Lu et al., 2009). HPLC with DAD detection was used for the analysis of melamine and formaldehyde to check their possible migration; the results were found to comply with the European standards (Lund & Petersen, 2006). Bradley et al. performed a survey of the migration of both melamine and formaldehyde from melamine kitchenware by HPLC where the formaldehyde migration exceeded SML (T) in 5 of the 50 units tested (Bradley et al., 2005). Spectrophotometry has always been an important tool for analytical chemists; the same technique was used to analyze the migration of melamine and formaldehyde from tableware (Ishiwata et al., 1986). In addition to kitchenware, formaldehyde was analyzed by spectrophotometry in toys and fabrics (Lynch, 2009), paper, food packaging materials (Dogan & Sanci, 2015) and plastic (Xiao-yan et al., 2009). Another spectrophotometric study of formaldehyde migration was performed by Potter et al. where 3% acetic acid was used as simulant and FT-IR was used for the characterization to confirm the plastic material (Potter et al., 2010). Colorimetric and reflectometric methods were also used to determine formaldehyde in fishery products (Rehbein & Schmidt, 1996). Gas chromatography is one of the important tools for determination of formaldehyde. The technique has been used for formaldehyde detection and quantification in different matrices (Daoudy et al., 2018; Dojahn et al., 2001; Sandler & Strom, 1960; Luong et al., 1996; Dumas, 1982). Among the wide range of analytical instruments GC-MS are important for both quantitative and qualitative analysis of food residues, additives and contaminants as it offers fast and sensitive procedure along with high peak capacity (Vazquez-Roig & Pico, 2012). The reported article also considers it as an important tool for analysis of thermally stable and volatile compounds. There are several article reported, where GC-MS is reported for food analysis they include; analysis of Polycyclic aromatic hydrocarbons (Siddique et al. 2020), fruits (Sun et al., 2020) and roasted coffee (Oliveira et al., 2005). Additionally, GC-MS technique coupled with HS-SPME was also employed to determine different volatile compounds (Cui et al., 2020). Gas chromatography coupled to mass spectrometry (GC-MS) has also been used for the determination of formaldehyde in various samples (Kenessov et al., 2011; Lobo et al., 2015; Yeh et al., 2013; del Barrio et al., 2006). In another important study, time-domain nuclear magnetic resonance (TD-NMR) was employed to get the quantitative information of formaldehyde in milk sample, chemometric method was used for the same. The results suggest excellent correlation with the official method and the procedure offers an alternate to the dairy industry owing to its non-invasive features (Coimbra et al., 2020).

In the literature there are several other reports dealing with the formaldehyde analysis, in the quality assessment during frozen storage of blue shrimp, formaldehyde was analyzed using colorimetric and spectrometric methods (Valencia-Perez et al., 2015). Another study in shrimp mentions that formaldehyde along with dimethylamine can be formed by decomposition of trimethylamine oxide which may lead to degraded quality of the products (Cintra et al., 1999).

Looking at the risk associated with the formaldehyde and to check the possible presence of substandard melamine material a GC-MS method is developed. The developed method is also aim at studying the possible migration of formaldehyde from the plastic kitchenware. The study involves simulation of the formaldehyde migration prior to its analysis.

2 Material and methods

2.1 Chemicals and reagents

Formaldehyde-2-4-dinitrophenylhydrazone (FA-DNPH) standard solution was purchased from Dr. Ehrenstorfer GmbH (Augsburg, Germany). Acetonitrile (ACN) used for diluting the FA-DNPH was obtained from LGC Promochem Optigrade SO-9184. Ammonia and acetic acid were procured from Avonchem (Cheshire, UK) while toluene was purchased from Acros Organics.

Solution preparation

The stock solution of 2-4-DNPH for the migration studies was prepared by adding 0.01 g of DNPH + 0.1 ml concentrated H_2SO_4 in a 50 mL flask and filled with ACN up to the mark. Preparation of the standard samples: A series of standard solutions of FA-DNPH were prepared by diluting the FA-DNPH reference standard. The working standards were prepared in the range of 2.5 to 30 ppm.

Sample collection

Different melamine samples of different qualities based on price were purchased from different stores in Riyadh, Saudi Arabia. The samples were stored at the same storage conditions as that of kitchen.

Method of simulation

Simulation of the sample was performed using 3% w/v acetic acid solution. Since several soup bowls of different quality, sizes, shapes and manufacturers were used during the present study, two different test conditions were tried: 2 and 4 hours (-/+5 minutes) simulation period at 70 °C measured in the simulant itself (+/- 5 °C).

Steps involved in the study of formaldehyde migration

- Different qualities of melamine-formaldehyde dishes were taken and filled with 3% acetic acid solution and kept inside the oven at 70 °C for different time intervals (2h and 4h) for the simulation of formaldehyde migration.
- 0.5 mL of the simulated solution was transferred into 15 mL falcon tube (BD falcon 352096)
- 0.1 mL of the derivatizing reagent (2-4-DNPH) was added to the falcon tube containing the sample and the mixture was vortexed for 30 seconds.
- 0.175 mL of aqueous NH_3 (32%, d=0.88, VWR 153312K) was added to the mixture and the sample was vortexed for 20 seconds.
- 1 mL of toluene (Across Organics 332070025) was added to allow *in situ* liquid/liquid extraction of the FA-DNPH and the mixture was then vortexed for 60 seconds
- An aliquot of the upper organic phase (toluene layer) was transferred to an auto sampler vial and then injected into the GC-MS system and the resulting chromatogram was recorded.

Instrumentation and experimental parameters

Gas chromatographic analysis was performed using Agilent 7890B GC system coupled to 5977B mass spectrometric detector. The GC condition include; Column: DB-5MS 30m (length) x 250µm (inner diameter) x 0.25 µm (film thickness), Inlet temperature: 260 °C, Carrier gas: He (99.999%), Injection volume: 1µl, Flow-rate: 1.7 mL/min; Split ratio: 5:1; Split flow: 10mL/min. Column temperature programming and the MS conditions are mentioned in Table 1 and Table 2.

Table 1. Column oven temperature program.

	Rate °C/ min	Temperature (°C)	Hold Time (Min)	Run Time (Min)
Initial	0	70	0	0
	15	260	6	20

Table 2. MS Parameters for the detection and determination of formaldehyde.

Ionization Mode	Positive ionization
Detection mode	Single ion monitoring
Mass range	50-450 Da (m/z)
SIM	152,180, 210m/z
Transfer line temperature	300 °C
Ion source temperature	300 °C

3 Results and discussion

Different brands of kitchenware samples, purchased from different local markets in Riyadh were subjected to stress conditions generally occurring in kitchen (by simulation of formaldehyde migration). The simulation method was optimized during the experiments and 3% of acetic acid was used for the same throughout the experiments (Conditions: temperature: 70 °C, time: 2 h and 4 h). The migrated formaldehyde concentrations were measured using the developed method based on gas chromatography coupled with mass spectrometry after post-derivatization of formaldehyde into 2-4-dinitrophenylhydrazone. The final concentration of formaldehyde was calculated using the following equation.

1. Obtained conc. of FA–DNPH (ppm) in simulant/ mL = Obtained conc. of FA–DNPH (ppm) in simulant (0.5 mL) x 2 (dilution factor)
2. Total conc. of FA–DNPH in the dish, mg/Kg = (Conc. of FA–DNPH(mL) x Volume of migration solution (L)) / Weight of dish (Kg)
3. Total conc. of FA in the dish, mg/Kg = (Conc. of FA–DNPH(mg/Kg) x Molecular weight of FA / Molecular weight of FA–DNPH

3.1 Method Validation

Every developed analytical method is considered as unauthentic unless it is validated using different parameters to make sure that there is sufficient accuracy, reproducibility and reliability. In our study several validation parameters as recommended by the regulatory authorities were studied. These parameters include system suitability, accuracy and precision, linearity and range, limit of detection and quantitation and recovery.

3.2 System Suitability

This parameter aims to check and ensure proper running of the instrumental system during the experiments. It is one of the integral parts of the chromatographic system to ensure the reproducibility of the system for accurate analysis. System suitability was ascertained using 17.5 $mg.L^{-1}$ standard solution. The concentration of the standard sample was selected as it lies at concentration between the linear range. The outcome of the system suitability shows that the RSD of the retention time was found to be 0.011% while the same for the peak area and corresponding concentration was found to be 1.98% and 1.77%. The mean concentration value obtained from the selected sample was based on peak area of six different runs and was found to be 17.43 $mg.L^{-1}$, while the recovery value was found to be 99.58%.

3.3 Specificity and confirmation of identity of the target analyte

Before proceeding for the experiments in a method development process it is very important to make sure that the signals obtained during the experiments are due to the target analyte and not due to some other interfering compounds. In order to confirm the identity of the specific compound

(formaldehyde), its identification and characterization was based first on its retention time which was compared to that of the certified standard sample in the same conditions. Then, the prepared FA-DNPH derivative was subjected to MS in full scan mode and the obtained spectrum revealed that the obtained m/z profile corresponded to both the mass spectrum of the standard target analyte and the reference spectrum reported in several spectral libraries, as shown in Figure 1.

3.4 Linearity and range

In this study the linearity study was performed in the concentration range of 2.5 mg.L^{-1} to 30 mg.L^{-1}. Calibration curve was prepared by plotting the area *vs* the individual concentration. The concentration points studied were 2.5 mg.L^{-1}, 4 mg.L^{-1}, 10 mg.L^{-1}, 15 mg.L^{-1}, 20 mg.L^{-1}, 25 mg.L^{-1} and 30 mg.L^{-1}. Linear regression equation obtained by plotting peak area vs concentration was found to be A= -32603 +17234 C where A is the peak area and C is the concentration of the analyte. Corresponding correlation coefficient R^2 was found to be 0.9974.

3.5 Accuracy and Precision

In our investigation the precision was carried out as intraday and inter day precision where the former was done on a single day and later was investigated for three days. In our study three-point precision was performed taking lower middle and the higher concentration point i.e. 4.5 mg.L^{-1},15 mg.L^{-1} and 30 mg.L^{-1}. The accuracy of the study was ascertained considering the recovery studies. The result of the accuracy and precision studies are mentioned in Table 3.

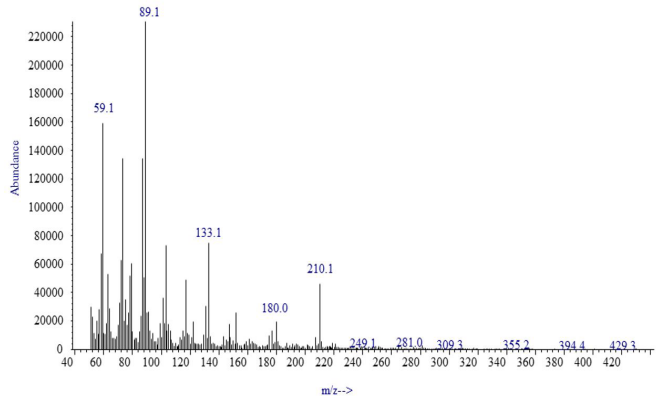

Figure 1. Full ion spectra of FA-DNPH in EI mode.

3.6 Recovery studies

The recovery studies were performed using both liquid-liquid extraction The results of the recovery study are mentioned in Table 4. From the table it can be summarized that the study was performed at three concentration points, 3.0, 7.5 and 22.5 mg l^{-1} and the recovery obtained in the study ranged from 97.64% to 99.43%. The % relative standard deviation at all points in the study was found in the range of 1.41-1.71.

3.7 Limit of Detection (LOD) and Limit of Quantitation (LOQ)

In the current study the LOD and LOD was determined using signal to noise ratio method, where the 3 × signal/noise ratio represents LOD while LOQ can be calculated as 10 × signal/noise. To investigate LOD and LOQ six blank sample were injected followed by six replicates of 0.35 mg.L^{-1} and six replicates of 1 mg.L^{-1}. The analysis of the response reveals that at 0.35 mg.L^{-1} the response of the signal was three times that of noise while the response at 1 mg.L^{-1} was found to be 10 time the response of the noise thus the LOD and LOQ of the current investigation was found to be 0.35 mg.L^{-1} and 1 mg.L^{-1}respectively the same corresponding to formaldehyde was found to be 0.05 mg.L^{-1} and 0.142 mg.L^{-1}.

3.8 Analysis of the real samples

Twenty-two real samples of melamine-formaldehyde resin soup dishes were tested for migration of formaldehyde. The simulation and the analysis procedures were adopted as mentioned in the experimental section. Post analysis, the results indicates that all the samples have shown formaldehyde migration. The migration of formaldehyde in each case was found to be higher when the samples were subjected to simulation for higher time (4h). The concentration of formaldehyde simulated at specified condition for 2 h was found in the range of 0.39 - 8.18 mg/kg, while the concentration of formaldehyde simulated at specified conditions for 4 h was found in the range of 0.44 - 9.53 mg/kg. Thus it can be concluded that the concentrations of the formaldehyde obtained after the migration studies are within the permissible limits of 15 mg/kg (Table 5) and chromatogram obtained from the real sample along with the with corresponding mass spectra is mentioned in Figure 2.

3.9 Comparison of the result with published methods

There are several analytical procedures present in the literature for the determination of formaldehyde in different matrix. Spectrophotometric analysis (Bradley et al., 2005) was

Table 3. Results of the accuracy and precision studies.

Precision	Amount of formaldehyde (mgL^{-1})		RSD (%)	Standard Analytical Error	Confidence Limit	Recovery %
	Taken	Found ± SD				
Intra Day	4.5	4.49 ± 0.04	0.959	0.019	0.05	99.72
	15.0	15.21 ± 0.202	1.32	0.090	0.251	101.41
	30.0	29.517 ± 0.167	0.568	0.075	0.208	98.39
Inter Day	4.5	4.41 ± 0.166	3.77	0.07	0.206	98.03
	15.0	14.87 ± 0.311	2.088	0.138	0.386	99.17
	30.0	30.48 ± 0.596	1.955	0.267	0.75	101.62

Table 4. Recovery studies post liquid-liquid extraction.

Amount of formaldehyde (mgL^{-1})		RSD	Standard analytical Error	Confidence Limit	%, Recovery
Taken	Found ±SD				
3.0	2.92±0.04	1.41	0.634	1.76	97.64
7.5	7.40±0.108	1.46	0.04	0.134	98.66
22.5	22.37±0.38	1.71	0.171	0.474	99.43

Table 5. Analysis of migrated formaldehyde in samples using GC-MS.

Sample No	Conc. of HCHO –DNPH (ppm)/0.5mL		Conc. of HCHO –DNPH (ppm)/mL (x2)		Wt. of Dish, Kg	Volume of simulant/ L	Total conc. of HCHO –DNPH (mg/Kg) in the dish/2H	Total conc. of HCHO –DNPH (mg/Kg) in the dish/4H	Total conc. of HCHO (mg/Kg) in the dish/2H	Total conc. of HCHO (mg/Kg) in the dish/4H
	2h	4h	2h	4h						
1	6.50	7.21	13	14.42	0.237	750 ml	41.14	45.63	5.88	6.52
2	6.44	7.50	12.88	15	270 g	1200 ml	57.24	66.67	8.18	9.53
3	6.88	7.20	13.76	14.44	262 g	1000 ml	52.52	54.96	7.50	7.85
4	6.88	7.10	13.76	14.2	269g	860 ml	43.99	45.40	6.29	6.49
5	8.39	9.14	16.78	18.28	123g	123 ml	16.78	18.28	2.40	2.61
6	6.72	7.51	13.44	15.02	94.1g	200 ml	28.56	31.92	4.08	4.56
QC 25 ppm	24.50									
7	6.50	7.12	13	14.24	94g	180 ml	24.89	27.27	3.56	3.90
8	7.10	7.44	14.2	14.88	91.8g	160 ml	24.75	25.93	3.54	3.7.
9	6.86	7.56	13.72	15.12	92.2g	160 ml	23.81	26.24	3.40	3.75
10	6.58	7.12	13.16	14.24	115g	200 ml	22.89	24.77	3.27	3.54
11	7.03	7.91	14.06	15.82	111.2g	280 ml	35.41	39.83	5.06	5.69
12	6.96	7.83	13.92	15.66	90.6g	160 ml	24.58	27.66	3.51	3.95
13	6.74	7.50	13.48	15	76.1g	160 ml	28.34	31.54	4.05	4.51
14	7.36	8.27	14.72	16.54	94g	160 ml	25.06	28.15	3.58	4.02
15	2.68	2.96	5.36	5.92	0.179	0.2 ml	5.99	6.62	0.86	0.95
16	2.98	3.42	5.96	6.84	0.207	0.16 ml	4.61	5.29	0.66	0.76
17	23.3	24.5	46.6	49.0	0.188	0.21 ml	52.05	54.74	7.44	7.82
18	3.21	3.62	6.42	7.24	0.365	0.75 ml	13.19	14.88	1.89	2.13
19	3.34	3.70	6.68	7.40	0.153	0.50 ml	21.83	24.18	3.12	3.46
20	2.78	3.08	5.56	6.16	0.299	0.180 ml	3.35	3.71	0.48	0.53
21	2.66	2.98	5.32	5.96	0.309	0.16 ml	2.76	3.08	0.39	0.44
22	3.57	3.98	7.14	7.96	0.296	0.40 ml	9.65	10.76	1.38	1.54

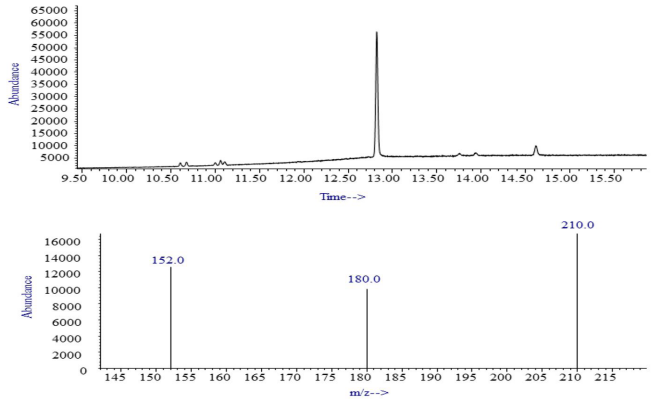

Figure 2. Chromatogram of target analyte in real sample and its corresponding mass spectra.

reported for formaldehyde migration, the sample measurement involved tedious process of heating the reaction sample at 60 ± 2 °C for 10 minutes and the absorbance measurement is completed in 35 and 60 minutes and the recovery is also 90%. Another colorimetric procedure reported (Dogan & Sanci, 2015) involves several pretreatment procedures including Carrez I and Carrez II reagents. The drawback of the procedure includes the precipitation which need to be removed by filtration which might interfere in the determination process. Sandler and strom (Rehbein & Schmidt, 1996) reported GC method for formaldehyde the analysis of the sample requires heating of the real samples at 125 °C and then to 180 °C for 24-hour period which means a longer and complex pretreatment prior to analysis. In the current analysis, there is minimal pretreatment procedure of simulation and the run time was 18 minutes. The LOD and LOQ of the current investigation was found to be 0.35 and 1.0 mgL^{-1} FA-DNPH corresponding to 0.05 mgL^{-1} and 0.143 mgL^{-1} of formaldehyde, respectively.

4 Conclusions

In this paper an analytical method based on gas chromatography-'mass spectrometry was developed for determination of formaldehyde traces, the complete determination

process also involve extraction and derivatization too. Different qualities of melamine materials were taken up for checking the possible migration of formaldehyde from melamine kitchenware. This melamine kitchenware was subjected to simulation using 3% acetic acid solution at 70 °C for 2 hours and 4 hours. It was observed that the melamine resin, when subjected to simulation for longer time resulted in higher migration of formaldehyde. The method was found to be linear in the concentration range of 2.5 mgL^{-1} to 30 mgL^{-1}. The precision studies suggest that the RSD at all the concentration level studied was found to be in the range of 0.96-3.77 while the accuracy measured in terms of the percent recovery was found to be in between 98.03% -101.62%. Recovery studies too were conducted and it was found to be in range of 97.64-99.43. Limits of detection and quantitation were found to be 0.35 mg. L^{-1} and 1 mg. L^{-1}respectively the same corresponding to formaldehyde concentration was found to be 0.05 mg. L^{-1} and 0.142 mg. L^{-1}. The analysis results showed that the level of migrated formaldehyde was 0.39 - 8.18 mg/kg when simulated for 2 hours and 0.44 - 9.53 mg/kg when simulated for 4 hours. These values establish that the levels of formaldehyde migrated into food are below the allowed specific migration limit (15 mg/kg food).

Acknowledgement

The authors are grateful to the Saudi Standards, Metrology and Quality Organization for funding this work. The authors are grateful to the Deanship of Scientific Research, king Saud University for funding through Vice Deanship of Scientific Research Chairs.

Reference

Bauer, D. R. (1986). Melamine/formaldehyde crosslinkers: characterization, network formation and crosslink degradation. *Progress in Organic Coatings*, 14(3), 193-218. http://dx.doi.org/10.1016/0033-0655(86)80001-2.

Bradley, E. L., Boughtflower, V., Smith, T. L., Speck, D. R., & Castle, L. (2005). Survey of the migration of melamine and formaldehyde from melamine food contact articles available on the UK market. *Food Additives and Contaminants*, 22(6), 597-606. http://dx.doi.org/10.1080/02652030500135243. PMid:16019835.

Brydson, J. A. (1999). Amino Plast. In J. A. Brydson. Plastic Materials (7th ed., pp. 668-693). London: Butterworth-Heinemann.</bok>

Cintra, I. H. A., Ogawa, N. B. P., Souza, M. R., Diniz, F. M., & Ogawa, M. (1999). Decomposition of trimethylamine oxide related to the use of sulfites in shrimp[1]. *Food Science and Technology (Campinas)*, 19(3), 314-317. http://dx.doi.org/10.1590/S0101-20611999000300003.

Coimbra, P. T., Bathazar, C. F., Guimarães, J. T., Coutinho, N. M., Pimentel, T. C., Neto, R. P. C., Esmerino, E. A., Freitas, M. Q., Silva, M. C., Tavares, M. I. B., & Cruz, A. G. (2020). Detection of formaldehyde in raw milk by time domain nuclear magnetic resonance and chemo metrics. *Food Control*, 110, 107006. http://dx.doi.org/10.1016/j.foodcont.2019.107006.

Cui, L., Xu, J., Feng, Z., Yan, M., Piao, X., Yu, Y., Hou, W., Jin, Y., & Ying-Ping, W. (2020). Simultaneous determination and difference evaluation of volatile components generated from ginseng fruit by HS-SPME Coupled with GC-MS according to fruit color. *Food Science and Technology (Campinas)*, 40(3), 532-536. http://dx.doi.org/10.1590/fst.26718.

Daoudy, B. D. A. D., Al-Khayat, M. A., Karabet, F., Al-Mardini, M. A. (2018). A robust static headspace GC-FID method to detect and quantify formaldehyde impurity in pharmaceutical excipients. *Journal of Analytical Methods in Chemistry*, 2, 1-8. https://doi.org/10.1155/2018/4526396.

Oliveira, A. L., Cruz, P. M., Eberlin, M. N., & Cabral, F. A. (2005). Brazilian roasted coffee oil obtained by mechanical expelling: compositional analysis by GC-MS. *Food Science and Technology*, 25(4), 677-682. http://dx.doi.org/10.1590/S0101-20612005000400009.

del Barrio, M.-A., Hu, J., Zhou, P., & Cauchon, N. (2006). Simultaneous determination of formic acid and Formaldehyde in pharmaceutical excipients using headspace GC/MS. *Journal of Pharmaceutical and Biomedical Analysis*, 41(3), 738-743. http://dx.doi.org/10.1016/j.jpba.2005.12.033. PMid:16464557.

Dogan, C. E., & Sanci, R. (2015). Formaldehyde migration in aqueous extracts from paper and cardboard food packaging materials in Turkey. *Food Additives and Contaminants-B*, 8, 221-226.

Dojahn, J. G., Wentworth, W. E., & Stearns, S. D. (2001). Characterization of formaldehyde by gas chromatography using multiple pulsed-discharge photoionization detectors and a flame ionization detector. *Journal of Chromatographic Science*, 39(2), 54-58. http://dx.doi.org/10.1093/chromsci/39.2.54. PMid:11245226.

Dumas, T. (1982). Determination of formaldehyde in air by gas chromatography. *Journal of Chromatography. A*, 247(2), 289-295. http://dx.doi.org/10.1016/S0021-9673(00)85952-X.

Ebner, I., Haberer, S., Sander, S., Kappenstein, O., Luch, A., & Bruhn, T. (2020). Release of melamine and formaldehyde from melamine-formaldehyde plastic kitchenware. *Molecules (Basel, Switzerland)*, 25(16), 3629. http://dx.doi.org/10.3390/molecules25163629. PMid:32784987.

European Food Safety Authority – EFSA. (2010). Scientific opinion on melamine in food and feed. *EFSA Journal*, 8(4), 1573.

Horie, K., Baron, M., Fox, R. B., He, J., Hess, M., Kahovec, J., Tikayama, T., Kubisa, P., Marechal, E., Mormann, W., Stepto, R. F. T., Tabak, D., Vohlidal, J., Wilks, E. S., & Works, W. J. (2004). Definitions Of Terms Relating To Reactions of Polymers and to Functional Polymericmaterials. *Pure and Applied Chemistry*, 76(4), 889-906.

European Union. The European Comission. (2011, January 1). Comission Regulation No 10/2011 of 14 January 2011 on plastic materials and articles intended to come into contact with food. Official Journal of the European Comission. Retrieved from: https://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2011:012:0001:0089:EN:PDF.

International Agency for Research on Cancer – IARC (2018). Formaldehyde. Retrieved from: https://monographs.iarc.fr/wp-content/uploads/2018/06/mono100F-29.pdf.

National Toxicology Program – NTP (2016, November 3). Formaldehyde. Retrieved from: https://ntp.niehs.nih.gov/ntp/roc/content/profiles/formaldehyde.pdf.

Ibarra, V. G., Bernaldo de Quirós, A. R., & Sendón, R. (2016). Study of melamine and formaldehyde migration from melamine tableware. *European Food Research and Technology*, 242(8), 1187-1199. http://dx.doi.org/10.1007/s00217-015-2623-7.

Ishiwata, H., Inoue, T., & Tanimura, A. (1986). Migration of Melamine and formaldehyde from tableware made of melamine resin. *Food Additives and Contaminants*, 3(1), 63-70. http://dx.doi.org/10.1080/02652038609373566. PMid:3956795.

Perkinelmer. Application note. (2009). Retrieved from: https://www.perkinelmer.com/lab-solutionsresources/docs/APP/FormaldehydeInToysUVVis.pdf.

Kenessov, B., Sailaukhanuly, Y., Koziel, J. A., Carlsen, L., & Nauryzbayev, M. (2011). GC–MS and GC–NPD Determination of Formaldehyde

Dimethylhydrazone in Water Using SPME. *Chromatographia*, 73(1-2), 123-128. http://dx.doi.org/10.1007/s10337-010-1820-6. PMid:21423319.

Li, Q., Sritharathikhum, P., Oshima, M., & Motomizu, S. (2008). Development of novel detection reagent for simple and sensitive determination of trace amounts of formaldehyde and its application to flow injection spectrophotometric analysis. *Analytica Chimica Acta*, 612(2), 165-172. http://dx.doi.org/10.1016/j.aca.2008.02.028. PMid:18358862.

Lobo, F. A., Santos, T. M. O., Vieira, K. M., Osório, V. M., & Taylor, J. G. (2015). Determination of formaldehyde in hair creams by gas chromatography-mass spectrometry. *Drug Testing and Analysis*, 7(9), 848-852. http://dx.doi.org/10.1002/dta.1808. PMid:25929824.

Lu, J., Xiao, J., Yang, D.-J., Wang, Z.-T., Jiang, D.-G., Fang, C.-R., & Yang, J. (2009). Study on migration of melamine from food packaging materials on markets. *Biomedical and Environmental Sciences*, 22(2), 104-108. http://dx.doi.org/10.1016/S0895-3988(09)60030-1. PMid:19618686.

Lund, K. H., & Petersen, J. H. (2006). Migration of formaldehyde and melamine monomers from kitchen- and tableware made of melamine plastic. *food additives and contaminants*, 23(9), 948-955. http://dx.doi.org/10.1080/02652030500415660. pmid:16901863.

Luong, j., sieben, l., fairhurst, m., & de zeeuw, j. (1996). determination of low levels of formaldehyde and acetaldehyde by gas chromatography - Flame Ionization Detection with A Nickel Catalyst. *Journal of High Resolution Chromatography*, 19(10), 591-594. http://dx.doi.org/10.1002/jhrc.1240191013.

Potter, E. L. J., Bradley, E. L., Davies, C. R., Barnes, K. A., & Castle, L. (2010). Migration of Formaldehyde from Melamine-Ware – UK 2008 Survey Results. *Food Additives and Contaminants*, 27(6), 879-883. http://dx.doi.org/10.1080/19440041003636638. PMid:20486003.

Rehbein, H., & Schmidt, T. (1996). A rapid and simple method for the determination of formaldehyde in fishery products. *Inf. Fischwirtseh*, 43(1), 37-39.

Sandler, S., & Strom, R. (1960). Determination of formaldehyde by gas chromatography. *Analytical Chemistry*, 32(13), 1890-1891. http://dx.doi.org/10.1021/ac50153a053.

Siddique, R., Zahoor, A. F., Ahmad, S., Ahmad, H., Mansh, A., Zahid, F. M., Faisal, S., & Aadil, R. M. (2020). GC-MS analysis of PAHs in charcoal grilled rabbit meat with and without additives. *Food Science and Technology (Campinas)*. http://dx.doi.org/10.1590/fst.34720.

Sun, H., Chen, W., Jiang, Y., He, Q., Li, X., Guo, Q., Xiang, S., Xi, W., & Liang, G. (2020). Characterization of volatiles in red- and white-fleshed loquat (Eriobotrya japonica) fruits by electronic nose and headspace solid-phase microextraction with gas chromatography-mass spectrometry. *Food Science and Technology*, 40(Suppl. 1), 21-32. http://dx.doi.org/10.1590/fst.27318.

Tyan, Y. C., Yang, M. H., Jong, S. B., Wang, C. K., & Shiea, J. T. (2009). Melamine Contamination. *Analytical and Bioanalytical Chemistry*, 395(3), 729-735. http://dx.doi.org/10.1007/s00216-009-3009-0. PMid:19669733.

Valencia-Perez, A. Z., Soto-Valdez, H., Ezquerra-Brauer, J. M., Márquez-Ríos, E., & Torres-Arreola, W. (2015). Quality changes during frozen storage of blue shrimp (Litopenaeus stylirostris) with antioxidant, α-tocopherol, under different conditions. *Food Science and Technology*, 35(2), 368-374. http://dx.doi.org/10.1590/1678-457X.6666.

Vazquez-Roig, P., & Pico, Y. (2012). Gas chromatography and mass spectroscopy techniques for the detection of chemical contaminants and residues in foods In D. Schrenk. *Chemical Contaminants and Residues in Food*. Philadelphia, PA: Woodhead Publishing

Xiao-yan, Z., Shao-hong, C., Zai-mei, L., & Guo-zhou, C. (2009). Acetylacetone Spectrophotometric determination of migration of formaldehyde and Hexamethylenetetramine from plastics into food simulants. *Shipin Kexue*, 30, 172-175.

Yeh, T.-S., Lin, T.-C., Chen, C.-C., & Wen, H.-M. (2013). Analysis of free and bound formaldehyde in squid and squid products by gas chromatography–Mass spectrometry. *Yao Wu Shi Pin Fen Xi*, 21, 190-197.