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Chinese Cooking with Ionic Seasonings May Enhance Migration of Perfluorooctanic acid from Food Contact Articles

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ABSTRACT

With its desirable water and oil resistant property, perfluorooctanic acid (PFOA) has been used as a key chemical in many consumer products and food contact articles (FCA), such as non-stick pans, oil-resistant food papers, carpets, textiles and paint. PFOA has been detected at noticeable levels in a wide range of environmental matrices. The present study investigated the safety of migration of PFOA at high temperature from non-stick cooking pans (125°C) and oil-resistant food papers (100°C) under simulated Chinese cooking conditions involving food oils and ionic seasonings (salts, soybean sauce, vinegar, tomato sauce). Results indicated that, in comparison with using oil alone, Chinese cooking using ionic seasonings in addition to oil would enhance migration of PFOA at a level up to 1.2 ng/dm^2 in cooking pans and 9.2 ng/dm^2 in food papers. Using a value of cumulative estimated daily intake of 6 ng/kg BW/day and the exposure scenario (food contact factor 155 g/dm^2, body weight 60 kg, intake rate 3 kg/head) set by the U.S. Food and Drug Administration, this study suggests a regulatory limit to be set for both cooking pans and food papers at 25 and 50 ng/dm^2 for PFOA, assuming a FCA consumption fraction of 0.8 and 0.4 for high and average consumers, respectively.

Key words: food contact articles, migration, perfluorooctanic acid, PFOA

INTRODUCTION

Perfluorooctanic acid (PFOA) belongs to a large family of perfluoroalkyl carboxylates (PFAC) containing a range of carbons from 5 to 12 (C5-C12) with a functional group of COOH attached to the terminal carbon of an 8-carbon (C8) alkyl chain. The molecular structure of PFOA is shown in Figure 1. Over 600 chemical precursors may be degraded to PFOA *via* food chain or environmental transformation. The chemical property of PFOA is characterized by its refractory to degradation and resistance to water and oil. Accordingly, it has been used extensively as a surfactant in many consumer products, such as non-stick cooking pans, food packaging, carpets, textiles, paint, cleaning agents and fire retardants. PFOA is also a key component of polytetrafluoroethylene, commonly known as Teflon. It has been widely found in polluted rivers[9], in indoor air dusts[12] and in fish livers[5,6], all at noticeably detectable levels.

Based on a number of animal studies, PFOA is likely to cause liver cancer and a host of non-cancer adverse effects to the liver, endocrine, immune and reproductive system in human[18,19]. A median level of 75.7 ng/mL (n=108) was detected in the blood of residents in 2005-2006 in a mid-Ohio Valley outbreak in USA[8]. For the population, an average level of approximately 5 ng/mL (n=7,876) was reported in blood samples collected in 1999-2006 in USA[11]. In an estimated medium exposure scenario in North America, a dose of PFOA of about 10 ng/kg BW/day was found to be received by infants primarily via hand-to-mouth from carpets and dust inhalation, and about 5 ng/kg-day received by both teens and adults primarily via food intake due to migration from food packaging materials[17]. Thus, a provisional standard of 0.4 ppb of PFOA in drinking water was promulgated by the United States Environmental Protection Agency (USEPA) in 2009[21].

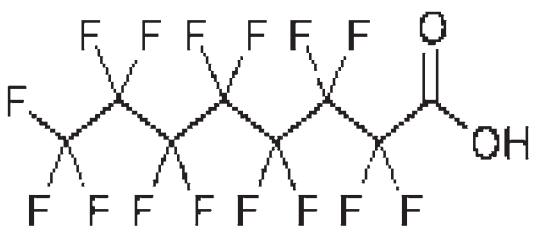

Figure 1. The molecular structure of PFOA.

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Studies have shown that, at room temperature with methanol as a food simulant, PFOA could migrate at a level up to 0.05 ng/cm^2 from non-stick cooking pans and 92.8 ng/cm^2 from food papers[20]. However, little was reported in literature on the migration of PFOA from food contact articles under Chinese cooking conditions at high temperatures. In addition, regulatory limits are currently not available for PFOA migration for cooking pans and oil-resistant food bags used in Taiwan. The purpose of this study was therefore to investigate the migration of PFOA from non-stick cooking pans and oil-resistant food papers under simulated Chinese cooking conditions involving food oils and ionic seasonings (salts, vinegar, soybean sauce) at high temperature (100-125°C). Regulatory analysis was also performed to calculate the estimated daily intake (EDI) and the specific migration limit (SML) with respect to the exposure scenarios set forth by the U.S. Food and Drug Administration[22] and by the European Commission of Standards[4], respectively, for comparison.

MATERIALS AND METHODS

I. *Migration Test at Room Temperature*

The migration test at room temperature and quality control measures developed by USEPA[20] were adopted in this study. The test was developed as a screening method for the surveillance of food contact articles (FCA) sold to consumers in retail stores. Three brands of PFOA-coated cooking pans and oil-resistant paper bags were purchased for the migration test from a major retail store in Taichung, Taiwan. These 3 brands were selected from among 15 brands for their highest migration levels in a preliminary study at room temperature. Therefore the test results of this study would represent conservative PFOA migration estimates for the food contact articles currently sold in the Taiwan market. The 15 brands of cooking pans and paper bags selected in this study had a wide range of list prices and hence represented a variety of manufacturing quality in market.

The USEPA protocol was carried out with methanol at room temperature. A volume of 100-150 mL of methanol was added into cooking pans to obtain a liquid depth of about 3 mm. A gentle agitation was provided during a total contact time of 24 h. For the coated papers, a total immersion test was performed using approximately 1 g of specimen in 50-mL polypropylene (PP) centrifuge tubes containing 45 mL of methanol. The tubes were placed on a horizontal table shaking at a speed of 100 rpm for 24 h. Several quality control measures were carried out in parallel to test the samples. Before migration, each test pan and tube was spiked with 0.1 mL of 500 ppb PFOA-$^{13}C_4$ in methanol as a recovery check surrogate standard (RCSS), and its recovery was used to check the accuracy of the test samples. A recovery of 80-120% was required as a quality criterion as suggested by USEPA[20]. Upon completion of migration, 0.1 mL of 500 ppb of perfluoro-decanoic acid PFDA-$^{13}C_2$ was added as an internal standard (IS) to each test, and its recovery was used to adjust the concentration of test samples to a recovery equivalent to 100% for the IS. A pair of split samples of the migrant solution was obtained for each batch of test and a relative percentage difference (RPD = $|\ x_1-x_2\ |\ /\ (x_1 + x_2)/2$) of 10% or less was required as test precision[20]. To check for possible contamination, a method reagent blank prior to the extraction was also processed in parallel to test samples for each batch of test.

II. *Migration Test at High Temperature*

In order to simulate typical Chinese cooking conditions, a high temperature migration protocol was developed in this study as modified from two reference test protocols, one recommended by the Center for Food Safety and Applied Nutrition (CFSAN) of USFDA[22] and another by the Committee European of Normalization[4]. A major brand of soybean oil sold in Taiwan (Taiwan Sugar, Inc.) was used as a food simulant in the migration test. The test temperature was set at 125±5°C for cooking pans and 100±5°C for paper bags, both for a total contact time of 15 min. The test temperatures and exposure times used in this study were within the ranges as reported in literature[2,3,13]. Five kinds of typical Chinese ionic seasonings were each used along with oil to simulate the Chinese cooking conditions: salt, soy sauce, vinegar, vinegar plus sugar, and tomato sauce. The five kinds of seasonings used in this study were those of major brands sold in market. A typical Chinese cooking menu of YTOWER[24] on the internet was consulted to determine the amount of each kind of seasoning used in cooking. A total of 4,164 Chinese major dishes were listed in the menu, in which 12 typical dishes made on cooking pans were selected. The five seasonings tested in this study were determined for the spiking dose by calculating the average of each seasoning used in the selected 12 dishes: salt 5 g, sugar 8 g, vinegar 14 g, soybean sauce 8 g and tomato sauce 40 g. In each test, the seasoned oil (18 mL) were first heated in a separate steel vessel to the test temperature and transferred to the pan or vessel for the test. Upon completion of migration, each test tube was spiked with 0.01 mL of 100 ppb PFOA-$^{13}C_4$ in methanol as the internal standard (IS), and its recovery was used to adjust the concentration of the test samples to an equivalent of a 100% recovery of the internal standard. For quality control, a recovery of 65-135% for the PFOA-spiked internal standard and a RPD of 35% or less for the split samples were required as a valid test[20]. The method reagent blank and standard solutions were prepared as for the migration test at room temperature.

III. *Preparation of Migrants for Instrumental Analysis*

The migrant samples collected in the test at room temperature were transferred in 170-mL polyethylene (PE) tubes and centrifuged at 4,500 rpm for 5 min[10]. The supernatants were collected in 170-mL polypropylene (PP) tubes and purged with nitrogen gas to near dryness. The final volume was adjusted to 10 mL prior to instrumental analysis. Five

standard solutions of 0.1-5.0 ng/mL were prepared to establish a calibration curve.

The oily migrant samples collected at high temperature were processed through a 4-step procedure of extraction, cleanup, solid phase extraction (SPE) and concentration to minimize interferences of instrumental analysis by impurities present in the sample matrices[16]. In the first step of extraction, 10 μL of 100 ppb PFOA-$^{13}C_4$ was added to the migrant oil as IS in a 50-mL PP centrifuge tube. Twenty-five-millilter mL of methanol: 1 M KOH de-ionized water (4 : 1, v/v) for every 15 g of oil was then added to the PP tube. The extraction was carried out on a horizontal shaking table for 30 min. In the second step of cleanup, the mixture in the PP tube was centrifuged at 5,000 rpm for 5 min. The extract was purged and concentrated with nitrogen gas to near dryness. Twenty-five-millilter of 10 mM of KOH was added to the concentrate, followed by supersonic vortex oscillation for 10 min. The sample was then centrifuged at 5,000 rpm for 15 min and the supernatant was collected for further treatment.

In the third step, a solid phase extraction (SPE) of 150 mg of Oasis WAX was sequentially activated by 4 mL of 0.1% ammonia methanol, 4 mL of methanol and de-ionized water. The supernatant after centrifugation was carefully added to the SPE column followed by washing twice with 6 mL of 25 mM ammonium acetate. The analyte was eluted with a 4 mL of 0.1% ammonia methanol to collect a volume of about 15 mL in a PP centrifuge tube. In the last step, the eluate was purged and concentrated with nitrogen gas at 60°C to near dryness. A final volume of 0.5 mL of methanol: 2 mM ammonium acetate (1 : 5, v/v) was added to the tube, followed by supersonic vortex oscillation for 10 min. Finally, centrifugation at 12,000 rpm for 10 min was done to collect the supernatant for subsequent instrumental analysis. Five standard solutions of 0.1-5.0 ng/mL were used to establish a calibration curve.

IV. *Instrumental Analysis*

Analysis of PFOA was performed using liquid chromatography tandem mass spectrometry (LC-MS/MS). An injection volume of 20 μL was introduced into an Agilent 1200 high-performance liquid chromatography system (Agilent, Germany). Separation was achieved on an Agilent ZORBAX Eclipse XDB-C18 (50×2.1 mm, 3.5 μm) analytical column kept at 40°C with an Agilent ZORBAX Eclipse XDB-C8 (12.5×2.1 mm, 5 μm) guard column. A 2 mM solution of ammonium acetate in de-ionized water and methanol were used as mobile phase solutions A and B. PFOA was chromatographically resolved using the following gradient program: 20% B at 0.25 mL/min for 3 min before injection. After injection, increasing to 50% B in 0.5 min, 95% B in 5 min, and then held at 95% B for 2.5 min, then decreasing to 20% B in 0.1 min, and then held at 20% B for 1.9 min till the end.

The liquid chromatograph was connected to an API 5000 MS/MS system (Applied Biosystems/ MDS Sciex, Canada) with a Turbo Ion Spray ion source operating in the negative electrospray mode. Samples were analyzed for PFOA and mass-labeled standards using the multiple reaction monitoring mode (MRM). Two precursor ions to product ion transitions were monitored for PFOA. The ion of a mass to charge ratio (m/z) 413→369 transition was used for PFOA quantification, whereas the 413→169 transition were used for PFOA qualification. The 417 → 372 transition was monitored for PFOA-$^{13}C_4$. The 515 → 470 transition was monitored for PFDA-$^{13}C_2$. The operating conditions used in this study were summarized in Table 1.

RESULTS AND DISCUSSION

I. *Migration from Coated Cooking Pans*

As shown in Table 2, the test results with oil alone at 125°C were not detectable (ND) with a limit of quantification (LOQ) of 0.013-0.018 ng/dm^2 for all the pans tested. The 3 brands of cooking pans tested with 5 oil and seasoning conditions (salt, soy sauce, vinegar, vinegar+sugar, and tomato sauce) gave a migration level of PFOA up to 1.2 ng/dm^2 (n=18) with a detectable rate of 56% (10/18) out of a total of 18 tests. Each type of seasoning was also analyzed for PFOA and the results showed that their concentrations were all below LOQ. The use of salt, vinegar or tomato sauce appeared to have significant effect in enhancing the migration of PFOA from cooking pans tested in this study. At 120-160°C, migration from cooking pans of about 0.1 ng/dm^2 with olive oil alone and 0.2 ng/dm^2 with olive oil plus potato stick was reported in Italy[3]. It appears that the presence of ionic seasonings would enhance the migration into oil from cooking pans. Further study is suggested to explore the quantitative relationship between PFOA migration and ionic strength of food stimulants. It should be noted that the migration could be more significant if the surfaces of food contact articles were made with poor-grade coating material or under inadequate thermal-setting pressure, temperature or holding time. The migration could be elevated and driven by dissolution physics of the PFOA coating if the cooking pans were incorrectly used by consumers, resulting in damage or scratches on the surface[22]. However, the difference in migration levels among the 5 seasoning test conditions was not significant among the 3 pans tested, even though Pan 2 had noticeably higher levels than Pans 1 and 3. The results of PFOA migration levels were comparable with those of ND-0.25 ng/dm^2 as reported by Bononi and Tatco[3] involving only oil at 120-160°C for 10 min. When tested with methanol at room temperature[20], the migration levels in this study were considerably higher at 0.9-2.1 ng/dm^2 for all the 3 brands of pans tested. Pan 2 gave the highest migration at room temperature in methanol and also gave the highest migration at 125°C in oil plus seasonings. In this study a recovery check of 88.5% and a RPD of 28.8% were achieved as a quality indicator.

II. *Migration from Food Papers*

Table 1. The operating conditions of HPLC/MS/MS used in this study for PFOA analysis

HPLC[a]

Chromato Column	Model: Agilent ZORBAX Eclipse XDB-C18 Size: 2.1 × 50 mm 3.5 μm
Protective Column	Model: Agilent ZORBAX Eclipse XDB-C8 Size: 2.1 × 12.5 mm 5 μm
Auto-sampler	Injection:10 μL
Column cont	Temperature: 40°C
Mobile pump	Mobile phase gradient:

Min	A (%)	B (%)
-3.0	80	20
0	80	20
0.5	50	50
5.5	5	95
8.0	5	95
8.1	80	20
10.0	80	20

Velocity: 0.25 mL/min
Phase A: De-ionized H_2O/2 mM NH_4Ac
Phase B: Methanol

ESI-MS/MS[b]

Ion Spray source	Collision gas(arbitrary unit (setting))	5
	Curtain gas (arbitrary unit (setting))	10
	Ion source gas 1 (arbitrary unit (setting))	45
	Ion source gas 2 (arbitrary unit (setting))	60
	Ion Spray voltage (V)	-4500
	Temperature (°C)	500
ESI/MS/MS	Ion pairs:	

Analyte	Precursor ion (m/z)	Product ion (m/z)	DP (V)	CE (eV)
PFOA	413	369*	-45	-14
		169**	-45	-23
PFOA -$^{13}C_4$	417	372	-45	-14
PFDA -$^{13}C_2$	515	470	-55	-14

DP: Declustering Potential
CE: Collision Energy
* PFOA quantification ion
** PFOA qualification ion

[a] High-performance liquid chromatography
[b] Electrospray ionization tandem mass spectrometry

Table 2. PFOA migration concentrations (in ng/dm^2) for 3 different brands of coated cook pans (125±5°C, 15 min) and food-contact papers (100±5°C, 15 min) under different cooking conditions

	Pan 1	Pan 2	Pan 3	Paper 1	Paper 2	Paper 3
Surface area (cm^2)	397	228	292	194	216	208
LOQ[a] (ng/dm^2)	0.013	0.018	0.017	0.012	0.012	0.012
Methanol (room temp)	1.2	2.1	0.90	12	22	50
Oil only	ND[b]	ND	ND	ND	ND	ND
Oil+salt	0.03	0.56	0.03	2.7	3.0	3.0
Oil+soy sauce	ND	0.73	0.25	6.1	0.8	0.9
Oil+vinegar	ND	0.65	ND	9.2	1.8	1.4
Oil+vinegar+sugar	0.07	0.76	0.04	6.5	1.7	1.2
Oil+tomato sauce	ND	1.20	ND	1.8	0.6	0.4

[a] LOQ, Limit of quantification.
[b] ND, Not detectable.

As shown in Table 2, the 3 brands of oil-resistant food papers tested at 125°C with 5 oil plus seasoning test conditions gave migration concentrations of PFOA of 0.4-9.2 ng/dm^2 with a detection rate of 100%. For Paper 1, the test with vinegar gave the highest level of migration and the test with tomato sauce gave the lowest level of migration. Seasoning with salt also resulted in a significantly higher level of migration from Papers 2 and 3. It is notable that all the 3 tests with cooking oil alone resulted in a migration of ND (LOQ=0.012 ng/dm^2) as for the cooking pans. Begley *et al.*[1,2] also reported a similar finding that, at 100°C for 15 min, oil (Miglyol) plus ionic or emulsified substances (butter, ethanol and vinegar) as a food simulant resulted in a significantly higher migration up to 12 ng/dm^2 from oil-resistant coated papers in contrast to the negligible migration with oil alone. This finding suggests that ionic substances can effectively enhance migration of PFOA from coated papers. Most Chinese cooking is likewise performed in conditions involving oil with ionic seasonings. However, the investigation of reaction chemistry on such an ionic effect was out of the scope of this study. The test with methanol at room temperature[20] on coated papers also resulted in much higher concentrations of 12-50 ng/dm^2. Although Paper 3 had the highest migration level with methanol at room temperature, Paper 1 gave a migration level of 5-6 times higher than Papers 2 and 3 at high temperature. A recovery of 88.2%

of PFOA-$^{13}C_4$ and RPD of 21.3% were obtained for the paper test as a quality indicator. Based on these results, it is suggested that the EPA protocol[20] using methanol at room temperature is most suitable as a screening method in the routine surveillance of food contact articles sold in retail stores, as the method gives more rigorous results comparing with simulated tests at high temperature and is subjected to less interference due to a cleaner matrix without oil.

III. *USFDA Regulatory Analysis*

Figure 2 is designed to compare the exposure scenarios and calculation of regulatory standard used in USA and in Europe. The USA algorithm is established with reference to "Guidance for Industry: preparation of Premarket Submissions for Food Contact Substances: Chemistry Recommendations", proposed by the Center for Food Safety and Applied Nutrition (CFSAN) of USFDA[22]. The Guidance is a non-binding recommendation, and the industries are allowed to present their own test protocols if they are proven to be more appropriate. Industries, prior to marketing food contact articles (FCAs) containing food contact substances (FCSs), permitted food additives, or unintentional impurities, must file an application for permit according to the Guidance. For FCAs of containers, a food simulant is filled into the container to perform the migration test. For plate articles, a one-sided migration cell (total immersion cell) is used. A two-sided migration cell is used if migration obtained with

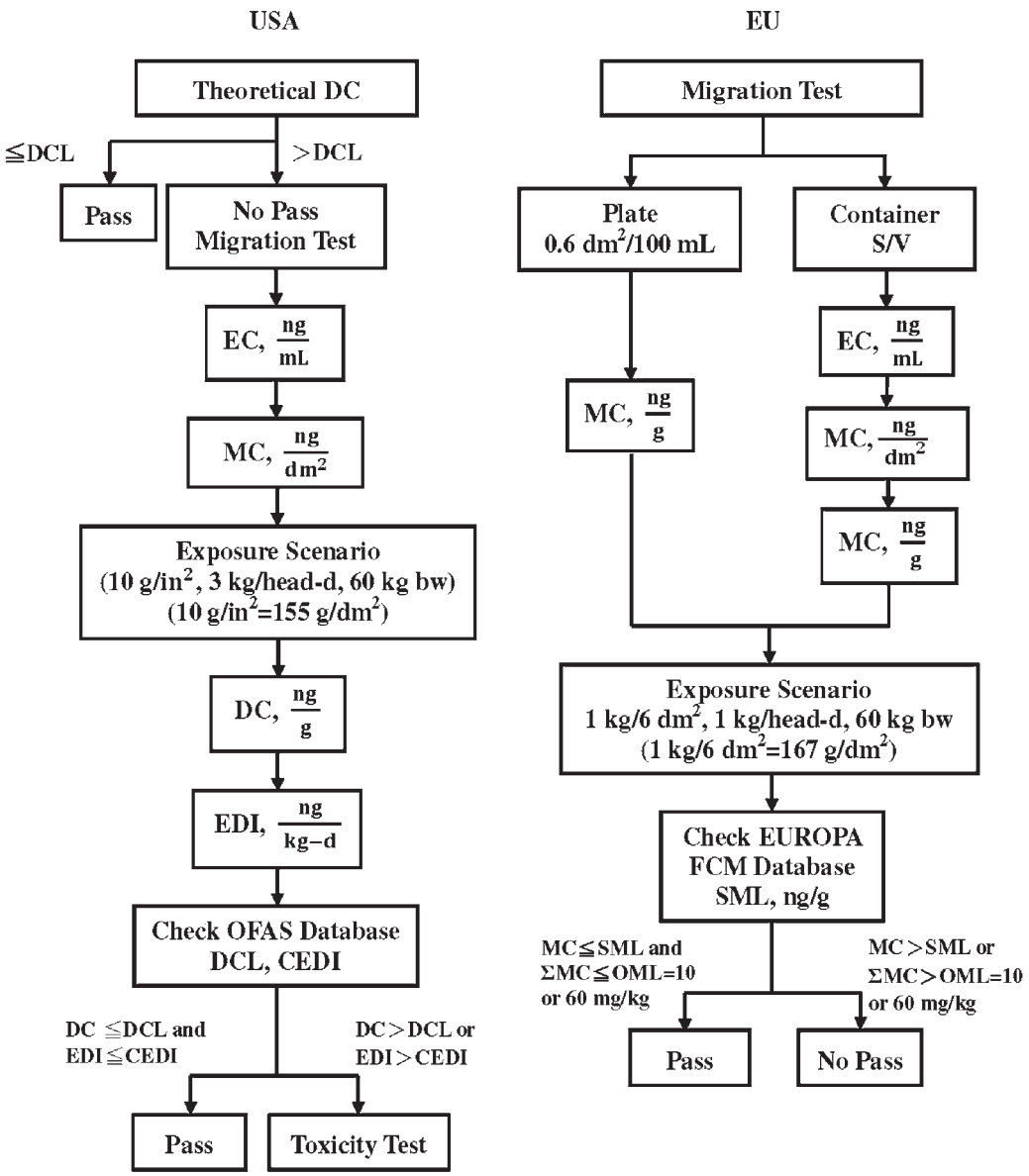

Figure 2. Comparison of the USA and European Union algorithm of migration concentration for regulatory compliance; CEDI: cumulative estimated daily intake, DC: dietary concentration, DCL: DC limit, EDI: estimated daily intake, EC: extraction concentration, MC: migration concentration, SML: specific migration limit, OML: overall migration limit.

the one-sided cell is viewed as inadequate. In migration tests, all foods are represented by three types of food simulants including: (1) 10% ethanol for aqueous and acidic foods, (2) 10 and 50% ethanol respectively for low and high alcoholic foods, and (3) food oil (e.g. corn oil), HB307, or Miglyol for fatty foods. Mild agitation of mixing is provided to prevent migration from limited dissolubility at any local spot in the test article.

As shown in Figure 2, for the USFDA algorithm, the first step is to determine the extraction concentration (EC_T, mg/L) in a specific type of food simulant under the assumption of 100% dissolution, or under the anticipated use conditions. A migration concentration (MC_T, mg/dm^2) for a specific type of food simulant is calculated as follows:

$$MC_T, \text{ ng/dm}^2=(EC_T, \text{ ng/mL})\ (V, \text{ mL})\ /(S, \text{ dm}^2) \quad (1)$$

where V is the volume of food simulant used in test, in mL and S is the surface area of the test article, in dm^2. The second step involves an calculation to obtain a sum of dietary concentration (DC, mg/kg) for all the types of food simulants with an assumption of a food contact factor of 10 g/in^2 (=155 g/dm^2) as follows:

$$DC, \text{ ng/g or ppb}=\Sigma[(MC_T, \text{ ng/dm}^2)(\text{dm}^2/155 \text{ g})\ (f_T)] \quad (2)$$

where f_T is the food-type distribution factor, specifying the fractions of the 4 types of foods utilizing certain FCA in question. For polymer-coated metal cooking pans, the f_T values found in the USFDA Guidance are 0.16 for aqueous food, 0.35 for acidic food, 0.40 for alcoholic food, and 0.09 for fatty food. For polymer-coated non-metal articles, the f_T values are 0.55 for aqueous food, 0.04 for acidic food, 0.01 for alcoholic food, and 0.40 for fatty food. For any FCA, the sum of f_T values for all types of foods is one. It should be noted that the distribution factors used in USA may not be appropriate for Taiwanese consumers.

In the third and final step, the estimated daily intake (EDI) in mg/kg BW/day is computed, assuming a body weight (BW) of 60 kg and a daily intake rate (IR) of 3 kg dietary food per head as follows:

$$EDI, \text{ ng/kg BW/day}=(DC, \text{ ng/kg})\ (3 \text{ kg/day})\ (CF)\ (1/60 \text{ kg BW}) \quad (3)$$

where CF is the fraction by weight of foods consumed (consumption fraction) which is associated with the concerned FCA relative to the total daily diet of 3 kg per head. CF values of 0.17 and 0.2 are found for coated cooking pans and coated papers, respectively[22].

To check if an EDI is acceptable in the USA, a USFDA Office of Food Additive Safety (OFAS) website database[23] can be consulted, in which regulatory standards are expressed as cumulative EDI ($CEDI$) and/or dietary concentration of cumulative DC (CDC). As of July 2011, a total of 1,267 FCS are listed in the OFSA database. A regulatory standard of 0.12 μg/kg (ppb) and 6 ng/kg BW/day can be found for CDC and $CEDI$, respectively.

Substituting Equation 2 into DC in Equation 3, rearranging the substituted equation for MC for the regulatory migration concentration limit (MCL), and setting EDI to equal $CEDI$, the equation for MCL can be obtained as follows:

$$MCL=(CEDI, \text{ ng/kg BW/d})(\text{d}/3000 \text{ g})\ (1/CF)(60 \text{ kg BW})(155 \text{ g/dm}^2)(1/f_T) \quad (4)$$

When letting $CEDI$=6 ng/kg BW/d and assuming f_T=1, equation 4 can be simplified as "MCL=18.6/CF". Considering the dietary style in Taiwan involving both cooking pans and coated papers, CF is assumed to be 0.8 for high exposure consumers and 0.4 for general exposure consumers. This analysis suggests a regulatory MCL of 25 and 50 ng/dm^2 for high and general exposure consumers, respectively. Substituting ML to Equation 2, a regulatory dietary concentration limit (DCL) of 0.15 and 0.30 ng/g (ppb) can be determined for high and general consumers, respectively. However, the DCL should be 0.12 ppb or less as suggested by USFDA (2007).

IV. *ECS Regulatory Analysis*

The European Union migration calculation is well described in the European Standard of EN 13130-1[4], "Materials and articles in contact with foodstuffs – Plastics Substances subject to limitation – Part 1: Guide to test methods for specific migration of substances from plastics to foods and food simulants and the determination of substances in plastics and the selection of conditions of exposure to food simulants". China fully adopted the EU standard and translated it into the Chinese Standard of B/T 23296.1-2009. As shown in Figure 2, MC in ng/dm^2 is first calculated from EC (in ng/mL) for containers when the surface to volume ratio (S/V) is known using Equation 1. When S/V is not known such as for plates, a default food contact factor of 100 mL/0.6 dm^2 (or 167 mL/dm^2) is used to determine the volume of food simulant to be used. It is of interest to note that a higher default value of 2 mL/cm^2 (or 200 mL/dm^2) is used in Taiwan[14,15]. In the second step, the unit of MC is converted from ng/dm^2 to ng/kg, which is equivalent to the DC in Equation 2, using the following equation:

$$MC, \text{ ng/kg}=(MC, \text{ ng/dm}^2)(\text{dm}^2/0.167 \text{ kg}) \quad (5)$$

In the third step, a EUROPA FCM internet database[7] is searched for the value of specific migration limit (SML), which is estimated with a tolerable daily intake (TDI) and an assumed exposure scenario of 1 kg/6 dm^2 (=167 g/dm^2), 1 kg/head/d, 60 kg BW/head as follows:

$$\text{SML, mg/kg}=(\text{TDI, mg/kg BW/d})\ (60 \text{ kg BW})(\text{d}/1 \text{ kg}) \quad (6)$$

According to EU Directive 2002/72/EC, an overall migration limit (OML) of 10 mg/dm^2 is required for plastic articles. This limit may be relieved to 60 mg/dm^2 in the

following cases: (1) container articles with a capacity of 0.5-10 L, (2) articles impractical to estimation of surface area, (3) caps, gaskets, stoppers or similar devices for sealing. To date, there are 994 chemicals being listed for regulation in the EUROPA database. PFOA is listed as Substance No. 00468 under FCS Category of 9.1. However, there is no recommended SML value for PFOA in the EU database.

CONCLUSIONS AND RECOMMENDATIONS

In comparison with test results using oil alone, Chinese cooking involving ionic seasonings may enhance the migration of PFOA up to a level of 1.2 mg/dm^2 from cooking pans and up to 9.2 mg/dm^2 from coated papers. Considering the significance of dietary exposure on top of many other pathways of PFOA and the extensive uses of coated papers in various food markets in Taiwan, it is of public health importance that the safety of these food papers be examined and regulated.

Using a value of CEDI of 6 ng/kg-day for PFOA and the exposure scenario recommended by the USFDA[22], a deterministic estimate in this study suggests a regulatory limit of 25 ng/dm^2 for PFOA for high consumers and 50 ng/dm^2 for general consumers, respectively, for both cooking pans and food papers.

The migration test with methanol at room temperature proposed by USEPA[20] is a relatively conservative yet simple method, compared with the migration method recommended by either the USFDA[22] or ECS[4] using conventional food simulants. Given the advantages of cleaner matrix and less interference, it is suggested that the USEPA protocol be adopted as a screening method for routine regulatory surveillance of FCAs.

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