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REUSABLE KITCHEN – AND TABLEWARE MATERIAL STABILITY TESTING WITH REGARD TO SPECIFIC MIGRATION OF MELAMINE AND FORMALDEHYDE

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Abstract: *Reusable melamine and bamboo kitchen and tableware containing melamine-formaldehyde resin as a filler and binding agent are popular among consumers. It has been found that melamine and formaldehyde can migrate into food and have adverse effects on human health. European legislation sets two conditions that reusable items must meet - they must meet specific migration limits and material stability requirements. This work aimed to test the compliance of reusable kitchen and tableware containing melamine-formaldehyde resin in terms of material stability. The test was carried out on 10 items made of melamine and bamboo. A Standard HPLC–UV method for melamine and spectrophotometric method for formaldehyde were used. The results for melamine migration varied. In 40% of the samples, the material did not comply with the legislation. In one item, the specific migration limit (SML) was exceeded by 8 times. In certain products, material instability has been established. In 30% of the samples, melamine migration was below the LOD, and the material demonstrated stability. Formaldehyde migration results were below the SML in all but one sample, which exceeded the limit by more than four times. In 27% of the samples, an increase in formaldehyde migration was found during the second exposure. However, according to the calculated value of* $|Z|$*, the material is considered stable under the legislation. For 40% of the items, formaldehyde specific migration levels were below the LOD. Therefore, the material stability testing of reusable kitchenware and utensils demonstrated non-compliance with the requirements of the legislation in more than half of the melamine and bamboo items tested. This can lead to unpredictable migration from bamboo and melamine products during their repeated use and raises concerns about the health of consumers.*
Keywords: *melamine, formaldehyde, food contact materials, compliance with legislation.*

1. Introduction

A wide variety of reusable plastic dishes and utensils are commonly used in modern kitchens. Among the most widely sold kitchen items are those made from melamine-formaldehyde resin, commonly referred to by consumers as melamine [1, 2]. This thermosetting plastic is produced through the catalytic polycondensation of melamine and formaldehyde (Fig. 1). It is inexpensive, durable, easily colored, and often marketed as dishwasher-safe and microwaveable. Melamine-formaldehyde resin is also used in the production of bamboo dishes and utensils as a bonding agent for bamboo flour [3, 4]. As a result of technological issues-such as incomplete polycondensation during polymer production, surface damage and scratching during molding, microwave use, heating, food acidity, and other factors-melamine and formaldehyde may migrate from such kitchenware into food. The literature reports a range of negative health effects associated with exposure to both monomers [5, 6, 7]. The IARC Monographs on the Evaluation of Carcinogenic Risks to Humans describe the harmful effects of each compound [8, 9]. Melamine primarily affects the kidneys and the male reproductive system [5, 6]. It is classified under Regulation (EC) No 1272/2008 (CLP) as a Category 2 carcinogen, toxic to reproduction, and as a substance with

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specific toxicity for certain organs and systems after repeated exposure [10]. The effects of formaldehyde primarily affect the central nervous system [8]. It is a mutagen and a proven human carcinogen, classified in Group 1 by the IARC. Formaldehyde is also an irritant to the mucous membranes, eyes, and skin [9, 11]. Both compounds-melamine and formaldehyde-are included in the list of authorized monomers in Regulation (EU) No 10/2011 on plastic materials and articles intended to come into contact with food, with specific migration limits of 2.5 mg/kg for melamine and 15 mg/kg for formaldehyde [12]. Most of the melamine and bamboo tableware and utensils available on the market are imported from China. In 2021, tests on children's bamboo utensils revealed that some items did not comply with the legal requirements regarding specific migration limits [9].

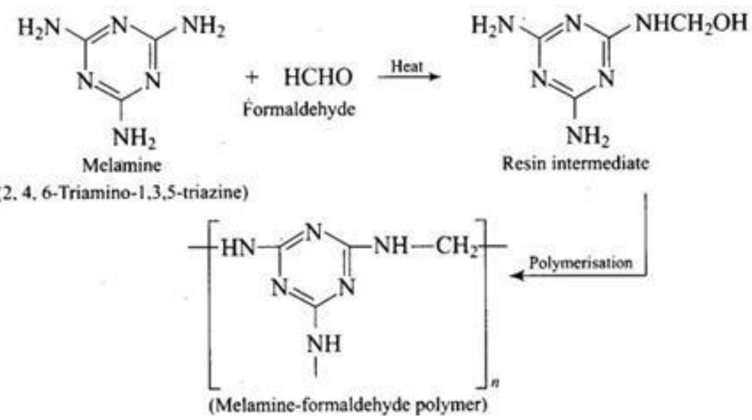

Fig.1. Preparation of melamine-formaldehyde resin [3]

Some authors [2] have reported a wide variation in the specific migration results of the two monomers across consecutive tests on the same item. As a result, a change in legislation was introduced [13], requiring reusable items to be assessed against two indicators. First, the migration test results must be below the relevant specific migration limits (SMLs); second, the material must demonstrate stability. For stability compliance, migration values from three consecutive exposures of the same item must be evaluated. These results are compared to the requirement that each subsequent migration must not exceed the previous one. To determine whether there is a significant difference between the values obtained from two consecutive migration tests, the following formula (1) is used:

$$|Z|=\frac{(m_k-m_{k-1})}{\sqrt{\left(u_{m_k}^2+u_{m_{k-1}}^2\right)}}, \quad (1)$$

where k = 1, 2, or 3 indicates the order of migration from the same item; and U_{m_k} is the standard uncertainty of the migration measurement m_k. When the value of $|Z| > 2$, the material is considered not to comply with the legislative requirements regarding material stability.

The aim of this paper was to assess the compliance of reusable tableware and cutlery containing melamine-formaldehyde resin with regard to material stability.

2. Materials and methods

2.1. Description of the samples

The study was conducted on 10 items (kitchenware sets, bamboo and melamine cups, plates, and bowls) made of melamine-formaldehyde resin and bamboo, as shown in Figures 1-10. According to the label, the items are dishwasher safe, intended for use at temperatures not exceeding 70 °C for no longer than 2 hours, and are manufactured in China.

Fig. 1 Bamboo kitchen utensils, set of 4 items, 30 cm, manufacturer 1

Fig. 2. Bamboo spoons, set of 2, manufacturer 2

Fig. 3. Bamboo bowls (450 mL)

Fig. 4. Reusable melamine plates, d=20 cm

Fig. 5. Reusable melamine plate, rectangular, 30 x 14 cm

Fig. 6. Bamboo cups with blue plastic lid, 400 mL

Fig. 7. White melamine bowl, d=10 cm

Fig. 8. Bamboo children's cups, 200 mL

Fig. 9. Plastic melamine cups, dotted, 200 mL

Fig. 10. Melamine bowls, blue, 720 mL

2.2. Analytical instrumental methods

Standardized instrumental methods, described in detail by [1], were used to test for the presence of the two monomers. A standardized reverse-phase liquid chromatography method with UV detection was used for the determination of melamine [14], while a standardized spectrophotometric method was applied for formaldehyde determination [15]. The methods were validated in accordance with ISO/IEC 17025:2017 *General requirements for the competence of testing and calibration laboratories* (Table 1).

Table 1.
Analytical Parameters of the Methods Used for Melamine and Formaldehyde Determination

Parameter	**HPLC-UV method for melamine, λ =230 nm**	**Spectrophotometric method for formaldehyde, λ=574 nm**
Limit of Detection (LOD), mg/kg	0.5	0.7
Limit of Quantification (LOQ), mg/kg	1.0	1.5
Working Range, mg/kg	(1.00 ÷ 8.00)	(1.5 ÷ 30.0)
Expanded Uncertainty, mg/kg	(0.07 ÷ 0.24)	(0.87 ÷ 2.41)
Correlation Coefficient	R^2 = 0.9997	R^2 = 0.9987

Sample pre-treatment was performed according to the procedures described in the Guidelines of the European Reference Laboratory for Food Contact Materials and Articles (EURL-FCM) [16, 17]. The applied test conditions for repeated exposure were as follows: simulant B (3% acetic acid), for 2 hours at 70 °C.

3. Results and discussion

Table 2 presents the results obtained for the specific migration of melamine from reusable melamine and bamboo tableware and kitchenware over three consecutive exposures using a food simulant—3% aqueous acetic acid. For three of the items (Item 3 – bamboo bowls, 450 mL; Item 5 – rectangular melamine plates; and Item 9 – dotted melamine cups), the results were below the detection limit of the method used (LOD = 0.5 mg/kg), indicating compliance with the legislation regarding specific melamine migration.

By contrast, the melamine migration limit was exceeded in all three exposures for Items 1 and 2—bamboo kitchen utensils and bamboo spoons, respectively. In the first test, melamine migration from the bamboo kitchen utensils exceeded the specific migration limit (SML) by approximately 8.7 times, while the bamboo spoons exceeded it by nearly three times. Although the specific migration values for both items decreased progressively from the first (m_1) to the third (m_3) exposure (i.e., $m_1 > m_2 > m_3$), the items are not considered compliant with the legislative requirements-neither in terms of the specific migration limit nor with respect to material stability [10].

For the bamboo cups with a blue plastic lid (Item 6), melamine migration during the second exposure (m_2) was higher than during the first (m_1), i.e., $m_2 > m_1$. Since $|Z| = 2.86 > 2$, the material was deemed unstable, and the article does not comply with the legislative requirements. For the melamine plates (Item 4), melamine migration during the first exposure exceeded the specific migration limit (SML) by nearly twice the permitted amount, although subsequent exposures were below the limit of detection (LOD). Since $|Z|_{1\text{-}2} > 2$, the item is considered non-compliant.

In contrast, the results for the reusable bamboo cups (Item 8) and melamine bowls (Item 10) showed a decrease in migration from the first (m_1) to the third (m_3) exposure, i.e., $m_1 > m_2 > m_3$. The migration values were below the SML, indicating that these products comply with the legislative requirements for both stability and specific migration of melamine.

For the white melamine bowls (Item 7), a migration value of (2.46 ± 0.45) mg/kg was recorded at the first exposure, which, including the expanded uncertainty, exceeds the specific migration limit (SML). Despite a decrease in migration during the subsequent two exposures, $|Z| = 3.00 > 2$, indicating that the material is unstable and the article does not comply with the legislation in this regard.

Table 2.

Melamine migration in reusable melamine and bamboo table and kitchenware

Item No.	Melamine-formaldehyde resin and bamboo articles, SML[1] = 2.5 mg/kg	Exposure 1, m_1	Exposure 2, m_2	Exposure 3, m_3	$\|Z\|_{1-2}$, m_1 and m_2	$\|Z\|_{2-3}$, m_2 and m_3
1	Bamboo kitchen utensils, 30 cm (Fig. 1)	21.73 ± 1.92	9.30 ± 0.82	7.47 ± 0.66	5.95	1.74
2	Bamboo spoons, couple (Fig. 2)	7.85 ± 0.69	4.58 ± 0.40	4.43 ± 0.39	4.10	0.27
3	Bamboo bowls (450 mL) (Fig. 3)	< LOD[2]	< LOD[2]	< LOD[2]	N/A[3]	N/A[3]
4	Reusable melamine plates, d=20 cm (Fig. 4)	4.60 ± 0.41	< LOD[2]	< LOD[2]	11.22	N/A[3]
5	Reusable melamine plate, rectangular, 30 x 14 cm (Fig. 5)	< LOD[2]	< LOD[2]	< LOD[2]	N/A[3]	N/A[3]
6	Bamboo cups with blue plastic lids, 400 mL (Fig. 6)	1.98 ± 0.31	2.08 ± 0.34	0.84 ± 0.27	0.22	2.86
7	White melamine bowl, d=10 cm (Fig. 7)	2.46 ± 0.45	0.99 ± 0.18	1.00 ± 0.18	3.00	0.04
8	Bamboo children's cups, 200 mL (Fig. 8)	2.33 ± 0.42	1.99 ± 0.36	1.95 ± 0.35	0.61	0.08
9	Plastic melamine cups, dotted, 200 mL (Fig. 9)	< LOD[2]	< LOD[2]	< LOD[2]	N/A[3]	N/A[3]
10	Melamine bowls, blue, 720 mL (Fig. 10)	2.22 ± 0.41	2.02 ± 0.36	2.00 ± 0.35	0.37	0.04

[1]SML - The specific migration limit; [2]LOD - Limit of detection of the method;
[3]N/A - "Not Applicable" for |Z|, when the results are <LOD.

Table 3 presents the results for the specific migration of formaldehyde. Except for one item, all analyzed articles showed formaldehyde migration levels below the specific migration limit (SML) of 15 mg/kg. In the case of the melamine plastic cups with dots (Item 9), the migration value recorded at the first exposure exceeded the SML by more than nine times, rendering the item non-compliant with the legislation regarding formaldehyde migration and material stability. The results for kitchen utensils (Item 1), bamboo spoons (Item 2), reusable melamine plates (Item 5), and melamine bowls (Item 10) were below the detection limit of the method used (LOD = 0.7 mg/kg), thus complying with the legislative requirements for formaldehyde migration and material stability. Although the results for the bamboo bowls (Item 3) showed a higher value during the second exposure compared to the first (i.e., $m_2 > m_1$), the calculated $|Z| = 1.46 < 2$ indicates that the material is stable, and the item complies with the legislative requirements. For bamboo cups with blue plastic lids (Item 6), migration during the second exposure (m_2) was higher than during the first (m_1), i.e., $m_2 > m_1$; however, since $|Z| = 0.81 < 2$, the material is considered stable, and the item complies with legislative requirements. For melamine plates (Item 4), an increase in migration values was observed from the first to the third exposure, i.e., $m_1 < m_2 < m_3$. Although the specific migration limit (SML) of 15 mg/kg was not exceeded, the material is considered unstable since $|Z| = 2.41 > 2$, and therefore the article does not comply with legislative requirements. For white melamine bowls (Item 7), migration decreased from the first to the third exposure ($m_1 > m_2 > m_3$), with $|Z| = 0.34$, indicating compliance with legislative requirements. Similarly, for children's bamboo cups (Item 8), migration also decreased across exposures ($m_1 > m_2 > m_3$), with $|Z| = 0.34$; therefore, the material is stable and complies with legislation regarding formaldehyde migration.

Table 3.

Formaldehyde migration in reusable melamine and bamboo tableware and kitchenware

Item No.	Melamine-formaldehyde resin and bamboo articles, SML[1] = 15 mg/kg	Exposure 1, m_1	Exposure 2, m_2	Exposure 3, m_3	$\|Z\|$, m_1 and m_2	$\|Z\|$, m_2 and m_3
1	Bamboo kitchen utensils, 30 cm (Fig. 1)	< LOD[2]	< LOD[2]	< LOD[2]	N/A[3]	N/A[3]
2	Bamboo spoons, couple (Fig. 2)	< LOD[2]	< LOD[2]	< LOD[2]	N/A[3]	N/A[3]
3	Bamboo bowls (450 mL) (Fig. 3)	1.88 ± 0.26	2.54 ± 0.37	2.23 ± 0.33	1.46	0.42
4	Reusable melamine plates, d=20 cm (Fig. 4)	4.40 ± 0.64	3.00 ± 0.44	5.10 ± 0.75	1.80	2.41
5	Reusable melamine plate, rectangular, 30 x 14 cm (Fig. 5)	< LOD[2]	< LOD[2]	< LOD[2]	N/A[3]	N/A[3]
6	Bamboo cups with blue plastic lids, 400 mL (Fig. 6)	2.57 ± 0.37	3.04 ± 0.45	2.50 ± 0.37	0.81	0.93
7	White melamine bowl, d=10 cm (Fig. 7)	3.76 ±0.56	2.18 ± 0.32	2.03 ± 0.30	2.45	0.34
8	Bamboo children's cups, 200 mL (Fig. 8)	2.48 ± 0.36	2.31 ± 0.35	1.51 ± 0.23	0.34	1.91
9	Plastic melamine cups, dotted, 200 mL (Fig. 9)	66.49 ± 9.67	N/A[3]	N/A[3]	6.88	N/A[3]
10	Melamine bowls, blue, 720 mL (Fig. 10)	< LOD[2]	< LOD[2]	< LOD[2]	N/A[3]	N/A[3]

[1]SML - The specific migration limit;
[2]LOD - Limit of detection of the method;
[3]N/A - "Not Applicable" for |Z|, when the results are <LOD.

Our results on melamine and formaldehyde migration from three consecutive tests, along with material stability assessments, are consistent with findings reported by other authors. Previous studies have confirmed that monomer migration from melamine kitchenware persists throughout the product's lifespan, particularly under conditions of heating or exposure to acidic environments. Testing of melamine and formaldehyde migration over 10 consecutive exposures was performed by Lund and Petersen [2] and Bradley et al. [18]. The authors noted differences in migration behavior between individual items of the same type, as well as variations in the extent of migration during successive exposures. Bradley et al. [18] observed a general trend regarding migration in melamine products. With continued use, there is a gradual decrease in the molar ratio of formaldehyde (CH_2O) to melamine (MEL), shifting towards lower CH_2O/MEL values. According to their hypothesis, newly produced products contain a relatively large amount of unreacted formaldehyde. However, monomer migration continues throughout the product's lifespan and, due to gradual polymer degradation, eventually stabilizes at a steady CH_2O/MEL ratio.

4. Participation in an interlaboratory comparison test organized by the European Reference Laboratory for Food Contact Materials and Articles

The European Reference Laboratory for Food Contact Materials (EURL-FCM) organised a comparative test to determine the specific migration of melamine and formaldehyde from bamboo/melamine tableware (FCM PT-23/01) in accordance with points 2.1.6 and 3.3.2. of Annex 5 to Commission Regulation (EU) 2020/1245 on the repeated use of materials and articles [19]. Twenty-four (24) national reference laboratories and 18 official control laboratories participated in the test.

The National Center of Public Health and Analyses (NCPHA) was assigned the code N03. The European Reference Laboratory for Food Contact Materials (EURL-FCM) provided two test products for the analysis: product 1 (T1), a 20 ml solution of melamine and formaldehyde prepared in food simulant B, and product 2 (T2), consisting of five cups designated for migration testing with food simulant B. The test procedure was performed on product 2 (T2), which is illustrated in Figure 11.

Fig. 11. EURL-FCM MUGS (T2)

The purpose of the test was twofold: to determine the specific migration levels of the two monomers-melamine and formaldehyde-and to assess the material stability of the analyzed items in accordance with the legislative requirements. The results obtained by the National Centre for Public Health and Analysis (NCPHA) for the analysis of the reference solution (T1) showed a z-score of 1.77 for melamine and -0.55 for formaldehyde (Table 4).

To assess the stability of the materials, additional analyses were conducted on three consecutive exposures of three test items (MUG1, MUG2, and MUG3), as shown in Table 5.

Table 4.

Results from the solution provided (T1)

	Melamine DD CEN/TS 13130-27:2005	Formaldehyde СД CEN/TS 13130-23:2005
T1 – 20 mL of a standard solution of melamine and formaldehyde in a model solution of 3% acetic acid	(3.20 ± 0.54) mg/kg *z-score* = 1.77	(11.45 ± 0.99) mg/kg *z-score* = (-0.55)

The laboratory N03 values obtained for melamine and formaldehyde from the interlaboratory comparison test conducted by EURL-FCM show z-scores up to "+2" for melamine and up to "(-1)" for formaldehyde, confirming the good technical competence and reliability of the results.

Table 5.

Interlaboratory comparison testing of EURL FCM

Melamine						Formaldehyde					
	T2 – bamboo cups						T2 – bamboo cups				
	Expo sure 1	Expo sure 2	Expo sure 3	u	RSu (k=1) *		Expo sure 1	Expo sure 2	Expo sure 3	u	RSu* (k=1)
MUG 1	1.09	1.00	0.85	0.17	7.8%	MUG 1	3.22	2.48	2.40	0.39	6.1%
MUG 2	1.16	1.53	1.18	0.21	9.1%	MUG 2	2.62	4.21	2.58	0.46	8.8%
MUG 3	1.05	1.07	0.91	0.17	8.1%	MUG 3	2.60	3.47	1.79	0.38	7.3%
Average	1.10	1.24	1.02	0.18		Average	2.81	3.39	2.26	0.41	

* RSu (k=1) - the calculated relative standard uncertainty

All results for melamine and formaldehyde were below their respective specific migration limits (SMLs), confirming compliance with regulatory requirements. The average melamine migration value during the second exposure (m_2) was higher than that of the first (m_1), i.e., $m_2 > m_1$; however, the calculated stability index $|Z| = 0.5 < 2$, indicating that the material is considered stable. Similarly, the average values for formaldehyde migration showed an increase from the first to the second exposure ($m_2 > m_1$), yet with a $|Z|$ value of $1.37 < 2$, the material again meets the stability criteria. Consequently, the tested item complies with the legislative requirements in terms of both migration limits and material stability. The items submitted for interlaboratory comparison testing demonstrated a wide variability in results, both regarding the specific migration of the two monomers and the material's stability. This variability highlights the importance of material homogeneity as a critical factor in determining whether a given article complies with legislative requirements.

5. Conclusions

The test results for melamine-formaldehyde resin and bamboo products reveal significant variability in both melamine and formaldehyde migration, as well as in material stability. More than half of the tested items (specifically items 1, 2, 4, 6, 7, and 9) failed to comply with the legislative requirements for either or both parameters. These findings raise serious concerns regarding consumer safety. Therefore, melamine and bamboo products should not be widely marketed for repeated use in households. Clear labeling should be mandated to inform consumers that such items are not safe for use in dishwashers or under elevated temperatures, as supported by the evidence obtained in this study.

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