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CONTRIBUTIONS TO THE DEVELOPMENT OF THE MATERIAL BALANCE OF MIGRATION PROCESS OF METAL IONS FROM THE AISI304 STAINLESS STEEL IN ACETIC ACID SOLUTIONS

***Silviu-Gabriel STROE[1], Gheorghe GUTT[1], Maria POROCH-SERIŢAN[1]**

[1] *Faculty of Food Engineering, Stefan cel Mare University of Suceava,*
13 Universitatii Street, 720229, Suceava, Romania
**silvius@fia.us.ro; g.gutt@fia.usv.ro; mariap@fia.usv.ro*
**Corresponding author*
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Abstract: *The aim of this work was the development of theoretical and real material balance by studying the diffusion phenomena of the metallic ions from AISI304 stainless steel samples in acetic acid solutions with 3%, 6% and 9% concentrations. The correlation of the quantities of substance which migrate from the metallic alloys in the food simulants through the interaction interface between the two environments can be accomplished through the materials balance developed for each component. To development the materials balance, we used the general stoichiometric equation of a chemical process. Within the comparative study of the theoretical and real mass balance, we have used the experimental data obtained after the migration tests, where the variables were represented by the working parameters: the temperature of the migration testing - T [°C], the exposure time - t [min.] and the stirring of the corrosive environment - n [rot·min^{-1}]. The value of each parameter was varied on three levels, in accordance with the real situations met in practice. In order to express the quantitative stage of the interaction between the metallic material and the corrosive environment, at a certain moment, we have used the degree of dissolution δ_M of the metallic components such as Mn, Cr, ^{56}Fe and Ni. The comparative study of the dissolution rates obtained allows to extrapolate and to elaborate in practice the optimization of the process which occurs at the interface of the two real food environments.*

Keywords: *diffusion, stainless steel, acetic acid, mass balance, dissolution rate*

1. Introduction

The knowledge and study of chemical reactions which occur at the interface between a metallic material and a food environment, considered corrosive, play an important role in the manufacturing process of food raw materials. Thus, in order to design the equipment or to optimize the processes we must know the operation conditions, such as: the nature of the corrosive environment, the temperature at which the processes occur, the duration of the contact between the two environments and the stirring of the corrosive environment.

2. Materials and methods

2.1. Metallic samples and corrosive environments

For the development of theoretical and real mass balance we have used the metallic samples of AISI304 stainless steel grade, the chemical composition being shown in the Table 1 (according to EN 10088-2:2005) [1]. The dimensions of the metallic samples used in the migration tests were of 40×40×1 mm.

Table 1.
Chemical composition of AISI304 stainless steel [wt %]

Fe	C	Mn	P	S	Si	Cr	Ni
67	0.08	2	0.045	0.03	1	18-20	8-11

As corrosive environments, we have used solutions of CH_3COOH, with the following concentrations: 3%, 6% and 9%.

2.2. Experimental design

The experimental design used for the migration tests in each of the three corrosive environments is presented in the Table 2.

Tabel 2. Experimental design used to perform the migration tests

Nr. exp.	Temperatura T-[°C]	Timp t-[min.]	Agitare n-[rot·min^{-1}]
1	22	30	0
2	22	30	125
3	22	30	250
4	28	30	0
5	28	30	125
6	28	30	250
7	34	30	0
8	34	30	125
9	34	30	250
10	22	60	0
11	22	60	125
12	22	60	250
13	28	60	0
14	28	60	125
15	28	60	250
16	34	60	0
17	34	60	125
18	34	60	250
19	22	90	0
20	22	90	125
21	22	90	250
22	28	90	0
23	28	90	125
24	28	90	250
25	34	90	0
26	34	90	125
27	34	90	250

After performing migration tests of the elements from the metallic samples in the acid solutions of the corrosive environments there have been identified and dosed, using mass spectrometry and inductively coupled plasma *ICP-MS*, the following metallic elements*: Mn, Cr,* ^{56}Fe and *Ni*, according to the Table 3.

Table 3. Concentrations of Mn, Cr, ^{56}Fe and Ni elements found in CH_3COOH solutions, used as corrosive environments

No. exp.	***Chemical element, [mg·L^{-1}]***											
	Mn			***Cr***			***^{56}Fe***			***Ni***		
	3%	***6%***	***9%***	***3%***	***6%***	***9%***	***3%***	***6%***	***9%***	***3%***	***6%***	***9%***
1	0.003	0.00157	0.00122	0.004	0.005	0.001	0.990	0.32	0.08	0.0152	0.311	0.0033
2	0.003	0.00237	0.00182	0.004	0.007	0.001	0.660	0.40	0.10	0.0072	0.341	0.0058
3	0.022	0.00287	0.00222	0.077	0.012	0.003	6.120	0.54	0.26	0.0782	0.381	0.0064
4	0.003	0.00177	0.00072	0.005	0.012	0.002	0.390	0.20	0.02	0.0132	0.391	0.0056
5	0.004	0.00227	0.00102	0.009	0.015	0.004	1.170	0.28	0.04	0.0232	0.531	0.0066
6	0.005	0.00287	0.00142	0.007	0.019	0.005	0.730	0.58	0.10	0.0352	0.551	0.0071
7	0.002	0.00197	0.00082	0.005	0.014	0.002	0.760	0.36	0.02	0.0082	0.411	0.0057
8	0.003	0.00207	0.00132	0.007	0.018	0.005	0.860	0.74	0.06	0.0132	0.511	0.0060
9	0.005	0.00247	0.00182	0.019	0.022	0.007	1.040	1.22	0.28	0.0372	0.561	0.0076
10	0.003	0.01467	0.00092	0.005	0.011	0.001	0.690	0.22	0.06	0.0132	0.281	0.0038
11	0.004	0.00607	0.00122	0.005	0.013	0.002	0.400	0.60	0.14	0.0068	0.331	0.0053
12	0.041	0.00387	0.00152	0.188	0.020	0.003	9.220	1.08	0.68	0.1372	0.381	0.0066
13	0.003	0.00207	0.00112	0.007	0.014	0.008	0.880	0.22	0.18	0.0152	0.301	0.0073

14	0.005	0.00377	0.00142	0.013	0.019	0.008	1.190	0.36	0.30	0.0192	0.321	0.0091
15	0.004	0.00357	0.00162	0.010	0.020	0.009	0.720	0.50	0.34	0.0282	0.367	0.0112
16	0.002	0.00227	0.00102	0.020	0.014	0.002	0.160	0.44	0.08	0.0072	0.231	0.0072
17	0.004	0.00287	0.00132	0.012	0.019	0.004	0.960	0.54	0.18	0.0202	0.276	0.0086
18	0.007	0.00367	0.00172	0.022	0.022	0.005	2.010	0.72	0.32	0.0322	0.306	0.0101
19	0.004	0.00257	0.00082	0.007	0.012	0.003	1.090	0.76	0.02	0.0132	0.301	0.0052
20	0.003	0.00267	0.00112	0.005	0.015	0.003	0.590	0.88	0.02	0.0082	0.331	0.0057
21	0.043	0.00297	0.00142	0.238	0.020	0.004	5.320	0.96	0.10	0.2372	0.361	0.0059
22	0.004	0.00387	0.00182	0.011	0.018	0.006	0.840	0.72	0.28	0.0172	0.121	0.0071
23	0.005	0.01167	0.00372	0.014	0.031	0.009	0.870	0.88	0.88	0.0172	0.141	0.0097
24	0.008	0.01267	0.00502	0.012	0.042	0.015	1.020	1.00	1.16	0.0302	0.172	0.0131
25	0.008	0.00147	0.00172	0.012	0.013	0.008	1.230	0.20	0.44	0.0262	0.160	0.0079
26	0.016	0.00357	0.00522	0.079	0.026	0.015	4.320	0.66	1.24	0.0752	0.211	0.0088
27	0.025	0.01567	0.00972	0.108	0.039	0.036	7.920	1.04	2.28	0.1072	0.241	0.0168

3. Results and Discussion

3.1. Theoretical material balance

Assuming that the reaction kinetics is known, this paper presents the theoretical and real material balances of the diffusion processes which occur between the AISI304 stainless steel grade samples and the food simulants (acetic acid solutions - 3%, 6% and 9%). The correlation of the substance quantities which leave the metallic material and migrate in the food environment, at the interaction interface between the two environments, can be done by the use of the materials balance elaborated by components [2]. For the development of the material balance we have started from the stoichiometric equation of a chemical process, which can be written as (1) [2]:

$$\sum_{i=1}^{n} \nu_{A_i} \cdot A_i = \sum_{i=1}^{m} \nu_{A_i'} \cdot A_i' \qquad (1)$$

where:

ν_{A_i} - the stoichiometric coefficient of the reactants;

$\nu_{A_i'}$ - the stoichiometric coefficient of the reaction products;

A_i, A_i' - reactants, reaction products, respectively.

At the interaction interface between the food environment and the metallic material, *main chemical reactions, secondary chemical reactions* and *electrochemical reactions* occur. In order to express the quantitative stage of the interaction between the aliment and the metallic material, at a certain moment, we have used a dissolution rate of a metallic contaminant or a certain component δ_{A_i} [3], [4].

This parameter is defined as *dissolution rate* of the metallic contaminant and is expressed by the relation:

$$\delta_{A_k} = \frac{n_{A_k}^0 - n_{A_k}}{n_{A_k}^0} = \frac{n_{A_i}^0 - n_{A_i}}{\frac{\nu_{A_i}}{\nu_{A_k}} \cdot n_{A_k}^0} = \frac{n_{A_i'} - n_{A_i'}^0}{\frac{\nu_{A_i'}}{\nu_{A_k}} \cdot n_{A_k}^0} \qquad (2)$$

where:

$n_{A_i}^0$, $n_{A_k}^0$, $n_{A_i'}^0$ - is the composition of the initial reaction mass of the contaminant (A_i), of a certain contaminant (A_k), and of the reaction products (A_i'), respectively;

n_{A_i}, $n_{A_i'}$ - the composition of the reaction mass at a certain moment of the contaminant (A_i), of the reaction products (A_i'), respectively. The use of the dissolution rate variable for the quantitative characterization of the process allows the development of the stoichiometric calculation in a simple form and the elaboration of the balance equation system in a form appropriate to their use for the quantitative description of the process. Starting with the composition of the reaction mass at a certain moment expressed through the sizes:

$$n_{A_1}...n_{A_n};\ n_{A'_1}...n_{A'_n} \tag{3}$$

and for each component it can be written the following balance equations [2]:

$$n_{A_i} = n^0_{A_i} - \frac{\nu_{A_i}}{\nu_{A_k}} \cdot n^0_{A_k} \cdot \delta_{A_k} \tag{4}$$

$$n_{A'_i} = n^0_{A'_i} + \frac{\nu_{A'_i}}{\nu_{A_k}} \cdot n^0_{A_k} \cdot \delta_{A_k} \tag{5}$$

$$n_{A''_i} = n^0_{A''_i} \tag{6}$$

The total number of moles in the reaction mass, at a certain moment, is expressed by the balance equation (7):

$$n_T = \sum_1^n n^0_{A_i} + \sum_1^m n^0_{A'_i} + \sum_1^s n^0_{A''_i} - \frac{\sum_1^n \nu A_i}{\nu A_k} n^0_{A_k}\delta_{A_k} + \\ + \frac{\sum_1^m \nu_{A'_i}}{\nu_{A_k}} n^0_{A_k}\delta_{A_k} = n^0_T + \frac{\sum_1^m \nu_{A'_i} - \sum_1^n \nu_{A_i}}{\nu_{A_k}} n^0_{A_k}\delta_{A_k} \tag{7}$$

The equation (7) can be written also as equation (8) [2]:

$$n_T = n^0_T \left(1 + \frac{\sum_1^m \nu_{A'_i} - \sum_1^n \nu_{A_i}}{\nu_{A_k}} \frac{n^0_{A_k}}{n^0_T} \delta_{A_k} \right) = \\ = n^0_T \left(1 + \alpha\delta_{A_k}\right) \tag{8}$$

where: $$\alpha = \frac{\sum_{i=1}^m \nu_{A'_i} - \sum_{i=1}^n \nu_{A_i}}{\nu_{A_k}} \cdot \frac{n_{A_k}}{n^0_T} \tag{9}$$

By replacing the number of moles by mass, the equation (2) can be written as equation (10):

$$\delta_{A_k} = \frac{m^0_{A_k} - m_{A_k}}{m^0_{A_k}} = \frac{m^0_{A_i} - m_{A_i}}{\frac{M_{A_i}}{M_{A_k}} \cdot \frac{\nu_{A_i}}{\nu_{A_k}} m^0_{A_k}} = \frac{m_{A'_i} - m^0_{A'_i}}{\frac{M_{A'_i}}{M_{A_k}} \cdot \frac{\nu_{A'_i}}{\nu_{A_k}} m^0_{A_k}} \tag{10}$$

3.2. Real material balance

In case of AISI304 stainless steel grade, used in the processing of food raw materials, the initial mass of reaction is considered to be formed by the metals presented in Table 1 (according to *EN 10088-2:2005*) [1], from the composition of metallic material and the acetic acid (CH_3COOH) [5].

The acetic acid has the following properties: $M = 60{,}05 g \cdot mol^{-1}$, $pK_a = 4{,}76$, at 25°C. The stoichiometric equation of the chemical process from the interaction interface between the acetic acid and AISI304 stainless steel, in case of obtaining the vinegar by acetic fermentation, is the following:

$$Mn^0 + Cr^0 + Ni^0 + 2Fe^0 + 8C_2H_4O_2 + 5/2O_2 \rightarrow (C_2H_3O_2)_2Mn + (C_2H_3O_2)_2Cr\downarrow + \\ (C_2H_3O_2)_2Ni + (C_2H_3O_2)_2Fe + Fe(OH)_2 + 3H_2O \tag{11}$$

and the secondary reaction is:

$$3Fe(OH)_2 \rightarrow Fe_3O_4 + H_2 + H_2O \quad (12)$$

In order to express the quantitative stage of the interaction reaction between the metallic material and the aliment, at a certain moment, it is used the *dissolution rate* δ_{Mn} of a metallic contaminant (*Mn*), expressed by the relation:

$$\delta_{Mn} = \frac{m_{Mn}^0 - m_{Mn}}{m_{Mn}^0} \quad (13)$$

The mass of each component, in a certain moment, is expressed by the equation (13) [3], [4].

The theoretical masses calculated for the contaminants *Mn, Cr,* ^{56}Fe and *Ni* which could result from the complete dissolution of the metallic samples are presented in Table 4.

Tabel 4. Theoretical masses of the contaminants *Mn, Cr,* ^{56}Fe and *Ni* [*mg*]

Mn	*Cr*	^{56}Fe	*Ni*
202.2	1819.8	6673.7	1011.0

Based on the relation (13) we have obtained the numeric values of the dissolution rate of the *Mn, Cr,* ^{56}Fe and *Ni* contaminants (Table 5).

Tabel 5. The dissolution rate of the contaminants *Mn, Cr,* ^{56}Fe and *Ni*

Nr. exp.	δ_{Mn}, [%]			δ_{Cr}, [%]			δ_{56Fe}, [%]			δ_{Ni}, [%]		
	3%	6%	9%	3%	6%	9%	3%	6%	9%	3%	6%	9%
1	0.0015	0.0008	0.0006	0.0002	0.0003	0,0055	0.0148	0.0048	0.0012	0.0015	0.0308	0.0003
2	0.0015	0.0012	0.0009	0.0002	0.0004	0,0055	0.0099	0.0060	0.0015	0.0007	0.0337	0.0006
3	0.0109	0.0014	0.0011	0.0042	0.0007	0.0002	0.0917	0.0081	0.0039	0.0077	0.0377	0.0006
4	0.0015	0.0009	0.0004	0.0003	0.0007	0.0001	0.0058	0.0030	0.0003	0.0013	0.0387	0.0006
5	0.0020	0.0011	0.0005	0.0005	0.0008	0.0002	0.0175	0.0042	0.0006	0.0023	0.0525	0.0007
6	0.0025	0.0014	0.0007	0.0004	0.0010	0.0003	0.0109	0.0087	0.0015	0.0035	0.0545	0.0007
7	0.0010	0.0010	0.0004	0.0003	0.0008	0.0001	0.0114	0.0054	0.0003	0.0008	0.0407	0.0006
8	0.0015	0.0010	0.0007	0.0004	0.0010	0.0003	0.0129	0.0111	0.0009	0.0013	0.0505	0.0006
9	0.0025	0.0012	0.0009	0.0010	0.0012	0.0004	0.0156	0.0183	0.0042	0.0037	0.0555	0.0008
10	0.0015	0.0073	0.0005	0.0003	0.0006	0,0055	0.0103	0.0033	0.0009	0.0013	0.0278	0.0004
11	0.0020	0.0030	0.0006	0.0003	0.0007	0.0001	0.0060	0.0090	0.0021	0.0007	0.0327	0.0005
12	0.0203	0.0019	0.0008	0.0103	0.0011	0.0002	0.1382	0.0162	0.0102	0.0136	0.0377	0.0007
13	0.0015	0.0010	0.0006	0.0004	0.0008	0.0004	0.0132	0.0033	0.0027	0.0015	0.0298	0.0007
14	0.0025	0.0019	0.0007	0.0007	0.0010	0.0004	0.0178	0.0054	0.0045	0.0019	0.0318	0.0009
15	0.0020	0.0018	0.0008	0.0006	0.0011	0.0005	0.0108	0.0075	0.0051	0.0028	0.0363	0.0011
16	0.0010	0.0011	0.0005	0.0011	0.0008	0.0001	0.0024	0.0066	0.0012	0.0007	0.0228	0.0007
17	0.0020	0.0014	0.0007	0.0007	0.0010	0.0002	0.0144	0.0081	0.0027	0.0020	0.0273	0.0009
18	0.0035	0.0018	0.0009	0.0012	0.0012	0.0003	0.0301	0.0108	0.0048	0.0032	0.0303	0.0010
19	0.0020	0.0013	0.0004	0.0004	0.0007	0.0002	0.0163	0.0114	0.0003	0.0013	0.0298	0.0005
20	0.0015	0.0013	0.0006	0.0003	0.0008	0.0002	0.0088	0.0132	0.0003	0.0008	0.0327	0.0006
21	0.0213	0.0015	0.0007	0.0131	0.0011	0.0002	0.0797	0.0144	0.0015	0.0235	0.0357	0.0006
22	0.0020	0.0019	0.0009	0.0006	0.0010	0.0003	0.0126	0.0108	0.0042	0.0017	0.0120	0.0007
23	0.0025	0.0058	0.0018	0.0008	0.0017	0.0005	0.0130	0.0132	0.0132	0.0017	0.0139	0.0010
24	0.0040	0.0063	0.0025	0.0007	0.0023	0.0008	0.0153	0.0150	0.0174	0.0030	0.0170	0.0013
25	0.0040	0.0007	0.0009	0.0007	0.0007	0.0004	0.0184	0.0030	0.0066	0.0026	0.0158	0.0008
26	0.0079	0.0018	0.0026	0.0043	0.0014	0.0008	0.0647	0.0099	0.0186	0.0074	0.0209	0.0009
27	0.0124	0.0078	0.0048	0.0059	0.0021	0.0020	0.1187	0.0156	0.0342	0.0106	0.0238	0.0017

After obtaining the numeric values (according to the Table 5), a comparative study can be done in relation to the influence of the working parameters on the dissolution rate of the contaminants.

It is known the fact that the presence of manganese in the stainless steels has the role to ensure deoxidation and to prevent the formation of iron sulphide inclusions [6].

In which concerns the behaviour of manganese (*Mn*) in acid environments, the lowest dissolution rate (*0,004%*) was obtained by the 7 and 19 experiences: *T-34°C* and *22°C, t-30 and 90 min.* and stationary environments (*n-0 rot·min*$^{-1}$).

The highest dissolution rate (*0.0124%)* was obtained in the experience no. 27, where the working parameters were: *T-34°C, t-90 min.* and *n-250 rot·min*$^{-1}$.

Concerning the behaviour of the chrome in acid environments, it is known the fact that this is not a chemical element to present an important migration phenomenon in food environments.

Due alloying chromium, the stainless steels are more resistant to corrosion [7].

The lowest dissolution rate (*0.0001*) can be noticed in case of experiences 4, 7, 11 and 16, where, as particularity, the corrosive environment was stationary, only in case of experience no. 11, the stirring of the environment being of *125 rot·min*$^{-1}$. The highest dissolution rate (*0.0131*) can be noticed in the case of experience no. 21, where the environment concentration was of 3% CH_3COOH, *T-22°C, t-90 min.* and *n-250 rot·min*$^{-1}$.

Regarding the behaviour of the iron to corrosion (^{56}Fe), the lowest dissolution rate were obtained in the case of experiences 4, 7, 19 and 20, where a stationary corrosive environment was used, the exception being the experience no. 20, where *n-125 rot·min*$^{-1}$. The highest dissolution rate of iron is observed in case of experiment no. 12, where the concentration was of 3% CH_3COOH, *T-22°C, t-60 min.* and *n-250 rot·min*$^{-1}$.

Amongst all studied metallic elements within this experiment, nickel presents the highest degree of risk on human health. The World Health Organization (OMS) recommends a maximum allowable dose of 0,005 mg/kg of body weight. OMS recommended also a value of nickel for the drinking water of 0,02 mg/l [8], and the daily intake of nickel from food products is estimated to 0,15-0,7 mg/day [8].

From studying the values of the dissolution rate for Ni, it is noticed that the minimum value of the degree of dissolution is met in case of the experience no.1, where solutions of 9% CH_3COOH, *T-22°C, t-30 min.* and *n - 0 rot·min*$^{-1}$ were used.

The maximum value of the dissolution rate is noticed in case of experience no. 9, where the concentration was of 6% CH_3COOH, *T-34°C, t-30 min.* and *n-250 rot·min*$^{-1}$.

4. Conclusions

The development of the theoretical and real mass balances of the migration processes of metallic acids in acid solutions, studied and presented in this paper, have had as main purpose the characterization of the migration processes from the interaction interface between acetic acid and AISI304 stainless steel grade samples through the calculation of the dissolution rate of the metallic contaminants *Mn, Cr,* ^{56}Fe and *Ni*. By elaborating these balances, we have obtained very important information regarding the behaviour of these stainless steels in the studied experimental conditions. One can notice that minimum values of the dissolution rate of the elements *Mn, Cr* and ^{56}Fe are obtained when solutions with 3% CH_3COOH concentrations were used, and the minimum dissolution rate of *Ni* were obtained when the environment concentration was of 9% CH_3COOH.

The maximum levels of dissolution of *Mn*, *Cr* and ^{56}Fe elements were obtained in case of the use of corrosive solutions with a concentration of 3% CH_3COOH, and for the element *Ni* when solutions with 6% CH_3COOH concentration were used.

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