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Possibility of application of green inhibitor for the protection of copper

The paper presents the assessment of copper corrosion protection by treatment of the corrosion surrounding with inhibitors. Copper samples measuring (50x50x1) mm after chemical preparation were subjected to corrosion in uninhibited and inhibited solutions during time of 2, 4, 6 and 24 hours. Two basic solutions were used: 3% NaCl and 4% HCl 0.1% solutions of thiocarbamide, furfural and hydrazine, and 1g/dm³, 2g/dm³ and 3g/dm³ of vitamine C and caffeine solutions were used as inhibitors.

Thus the highest protection effect was shown by furfural, but also vitamine C for 2 h. No toxicity and good protective properties recommend vitamine C as an inhibitor of the future.

Key words: *copper, inhibitor, corrosion, corrosion indicators, protective factor z.*

INTRODUCTION

Apart from the great technological progress in the manufacture and protection of modern construction materials and the wide range of possibilities in chosing the materials in designing, corrosion damages may be large and difficult to assess even nowadays. Chosing a construction material which is most resistant to corrosion in a medium is a very complex task which requires not only a wide knowledge of materials but also of chemical and electrochemical processes [1-15].

Inhibitors are nowadays applied in the protection of materials from abrasion (corrosion), oil production and refining, in heat exchangers, energy facilities, machinery conservation, rocket technics, mechanical engineering and other industrial branches. Inhibitors are substances that, after being introduced into a corrosion medium usually in very small quantities (0.01% -5%), are able to reduce the rate of electrochemical corrosion of metals and alloys (construction materials) to a great extent. Corrosion inhibitors are chemical compounds which are adsorbed on the surface of metals, and they are broadly classified into inorganic and organic groups of inhibitors based on their chemical composition [1-15].

A large number of potential inhibitors have been investigated so far. Although inorganic inhibitors are among them, organic compounds and their derivatives are much more numerous, such as azole compounds, amines, amino acids and many other. It has been observed that the presence of heteroatoms such as nitrogen, sulphur, phosphorous in a molecule of an organic compound improves its corrosion inhibitive action in certain construction materials.

This is explained by the presence of incomplete *d* orbitals in the atom which forms coordinative links wth atoms that are able to donate electrons. There is also an interaction with the rings containing conjugated links, i.e. π electrons. Based on these results, there is recently a growing tendency to synthetize compounds that possess more heteroataoms and functional groups, because it has been observed that these very factors in compounds are responsible for good characteristics during corrosion inhibition, due to the fact that they enable chemisorption. Greater molecular masses of compounds also have a favourable effect on the increase of physical adsorption.

Basic disadvantage of most commercial inhibitors used today is their toxicity. As environmental issues and harmful effects of the application of chemicals are receiving ever greater attention throughout the world, the traditional approach to corrosion inhibitors is beginning to change gradually. It is necessary to replace the toxic inhibitors, which are widely used in industrial processes, with the new "green inhibitors" that are safe to the environment. Research efforts are therefore beginning to be focused on non-toxic "green corrosion inhibitors" [7-21].

The aim of this paper is to investigate corrosion stability of copper in the 3% solution of NaCl and 4% solution of HCl depending on time. The aim is also to treat the corrosion medium with several commercial inhibitors, as well as with vitamine C as a non-toxic "green inhibitor", and to investigate the effect of inhibitors on the corrosion rate and to determine the most efficient inhibitor ion the given medium for the conditions applied.

EXPERIMENTALS

(50x50x1) mm copper samples were subjected to chemical preparation, and then the rate of corrosion in inhibited solutions was investigated. All the experiments were performed at room temperature, in

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two basic solutions (3% solution of *NaCl* and 4% solution of *HCl*), as well as in the solutions inhibited with: hydrazine, thiocarbamide, furfural, vitamine C and caffeine. Hydrazine, thiocarbamide and furfural are added in concentrations of 0,1 mas %, whereas vitamine C and caffeine were added in the concentrations of $1g/dm^3$, $2g/dm^3$ and $3g/dm^3$.

Copper samples were prepared in the following way:

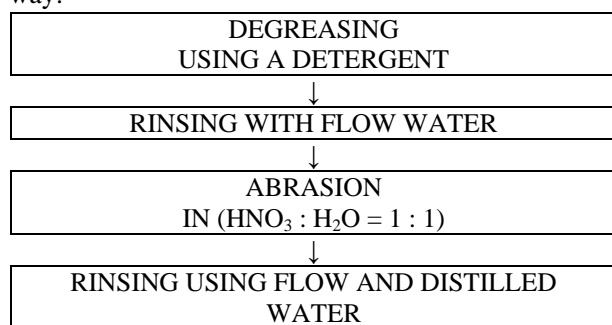| DEGREASING USING A DETERGENT |
| :---: |
| ↓ |
| RINSING WITH FLOW WATER |
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| ABRASION IN ($HNO_3 : H_2O = 1 : 1$) |
| ↓ |
| RINSING USING FLOW AND DISTILLED WATER |

Figure 1 - A flow chart of the preparation of the sample

Investigation of the rate of corrosion was performed in the following solutions:

Solution 1. *4% HCl*

Solution 2. *3% NaCl*

Solution 3. *4% HCl+0,1%(mas) of hydrazine*

Solution 4. *4% HCl+0,1%(mas)of furfural*

Solution 5. *4% HCl+0,1%(mas)of thiocarbamide*

Solution 6. *4% HCl + 1g/dm³ of vitamine C*

Solution 7. *4% HCl + 2g/dm³ of vitamine C*

Solution 8. *4% HCl + 3g/dm³ of vitamine C*

Solution 9. *3% NaCl + 1g/dm³ of vitamine C*

Solution 10. *3% NaCl + 2g/dm³ of vitamine C*

Solution 11. *3% NaCl + 3g/dm³ of vitamine C*

Solution 12. *3% NaCl + 1g/dm³ of caffeine*

Solution 13. *3% NaCl + 2g/dm³ of caffeine*

Solution 14. *3% NaCl + 3g/dm³ of caffeine*

Solution 15. *4% HCl + 1g/dm³ of caffeine*

Solution 16. *4% HCl + 2g/dm³ of caffeine*

Solution 17. *4% HCl + 3g/dm³ of caffeine*

Based on the loss in the mass of the sample after a certain period of time of exposition to corrosion medium, the negative mass corrosion index was calculated K_m^-, deep corrosion index π, degree of efficiency (degree of protection of the inhibitor) z. Moreover, the average values $\overline{K_m^-}, \overline{\pi}, \overline{z}$ for every sample were calculated.

RESULTS AND DISCUSSION

Copper samples of (50x50x1)mm after chemical preparation were subjected to corrosion in the solutionso 1 and 3–5.

Table 1 - Copper corrision indices in the solution 1 and solutions 3–5

Corrosion indices	Solution 1 4% HCl without the inhibitor			Solution 3 4% HCl + 0,1% hydrazine			Solution 4 4% HCl +0,1% furfural			Solution 5 4% HCl + 0,1% thiocarbamide		
τ (h)	2	4	6	2	4	6	2	4	6	2	4	6
K_m^- (g/m²h)	0,6442	0,6298	0,6186	0,2885	0,2885	0,2692	0,2115	0,1298	0,0993	0,2019	0,2163	0,2596
$\overline{K_m^-}$ (g/m²h)	0,6309			0,2821			0,1469			0,2259		
Z (%)	0	0	0	55,22	54,19	56,48	67,17	79,39	83,95	68,66	65,66	58,03
$\overline{z}$ (%)	0			55,29			76,84			64,12		
π (mm/god)	0,6341	0,6199	0,6089	0,2840	0,2840	0,2650	0,2082	0,1278	0,0977	0,1987	0,2129	0,2555
$\overline{\pi}$ (mm/god)	0,6209			0,2777			0,1446			0,2224		

Figure 2 shows the representation of deep corrosion index $K_{\bar{m}}$ in solution 1 and solutions 3–5.
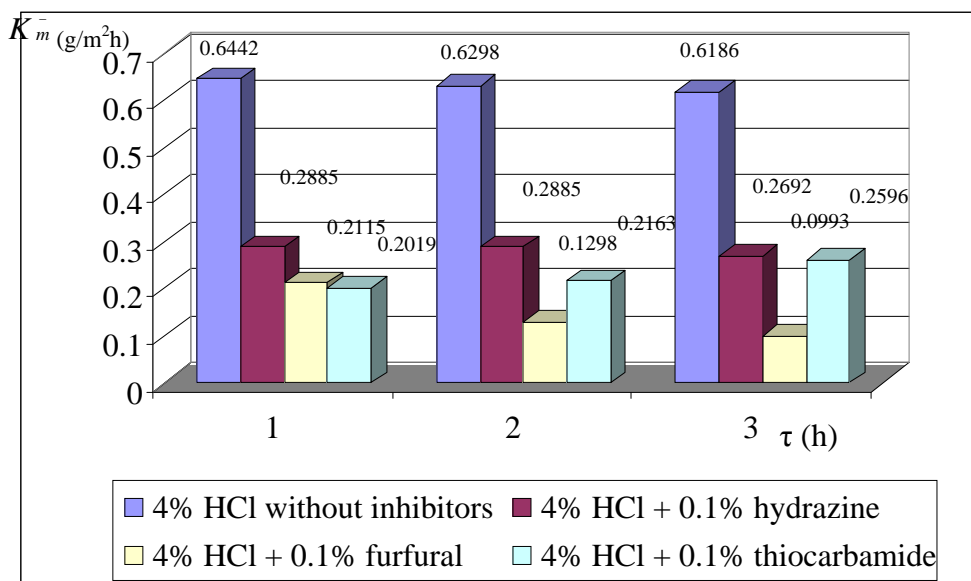

Figure 2 - Diagram of the deep corrosion index $K_{\bar{m}}$ in solution 1 and solutions 3–5

From Figure 2 it can be seen that the largest value of $K_{\bar{m}}$ is in the hydrazine solution $K_{\bar{m}}$ =0,2692–0,2885 g/m²h, and the lowest value is in the furfural solution $K_{\bar{m}}$ =0,0993–0,2115 g/m²h.

Figure 3 gives the representation of deep copper corrosion index in the solution 1 and solutions 3–5.
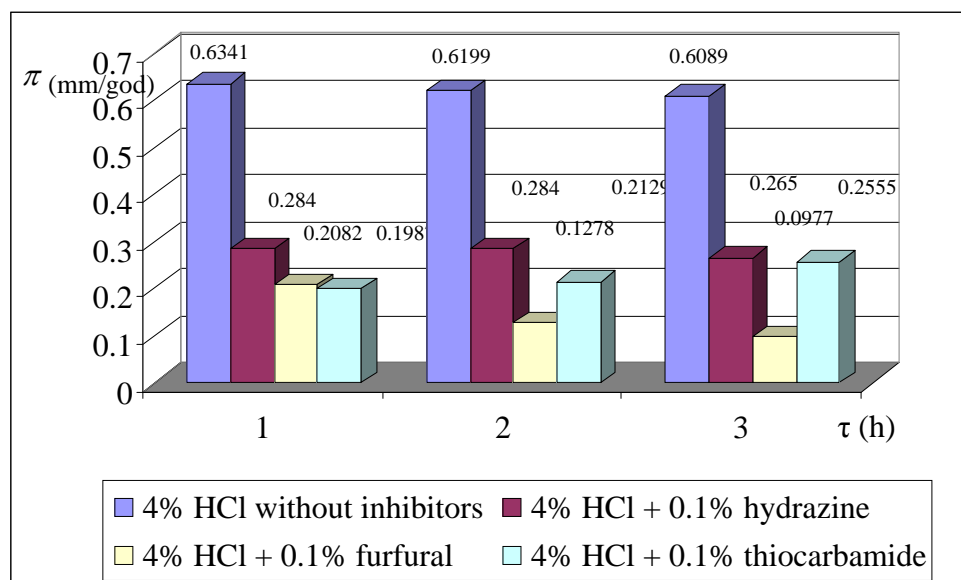

Figure 3. Representation of deep copper corrosion index in the solution 1 and solutions 3–5 depending on time

From Figure 3 it can be seen that the highest value of π is in the hydrazine solution π = 0,2650–0,2840 mm/year, whereas the lowest one is in the furfural solution π = 0,0977–0,2082.

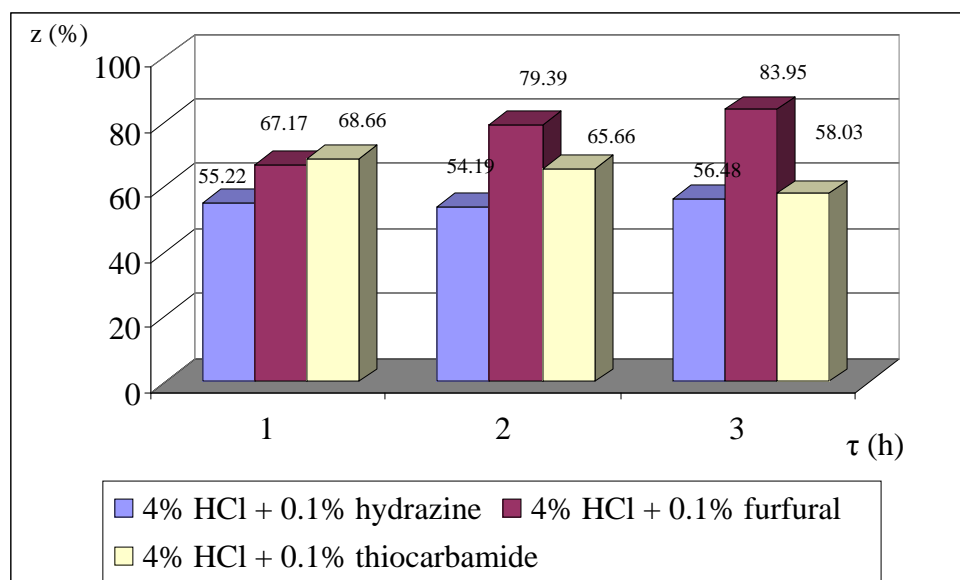Figure 4 shows the protection factor z (%) depending on time in the solutions 3–5.

Figure 4 - Diagram of protection factor z (%) depending on time in the solutions 3–5.

Figure 4 shows that the highest protection degree is with furfural z = 67,17–83,95 and that value z increases with time. the lowest value is for hydrazine z = 54,19–56,48 and there are no larger deviations of the value z with time.

Table 2 shows indices of copper corrosion in solution 1 and solutions 6–8.

Table 2 - Results for copper corrosion in Solution 1 and Solutions 6–8

Corrosion indices	Solution 1 4% HCl without inhibitors				Solution 6 4% HCl + 1g/dm³ of vitamine C				Solution 7 4% HCl + 2g/dm³ of vitamine C				Solution 8 4% HCl + 3g/dm³ of vitamine C			
τ (h)	2	4	6	24	2	4	6	24	2	4	6	24	2	4	6	24
$\bar{K_m}$ (g/m²h)	0,6442	0,6298	0,6186	0,6563	0,3712	0,3605	0,3598	0,3631	0,2885	0,2743	0,3142	0,2913	0,2115	0,2342	0,2973	0,2244
$\bar{\bar{K_m}}$ (g/m²h)	0,6372				0,3636				0,2921				0,2419			
z (%)	0	0	0	0	42,38	42,76	41,84	44,67	55,22	56,45	49,21	55,61	67,17	62,81	51,93	65,81
$\bar{z}$ (%)	0				42,91				54,12				61,93			
π (mm/year)	0,6341	0,6199	0,6089	0,6460	0,3654	0,3548	0,3541	0,3574	0,2840	0,2699	0,3093	0,2867	0,2082	0,2305	0,2973	0,2209
$\bar{\pi}$ (mm/year)	0,6272				0,3579				0,2875				0,2392			

Figure 5 shows the dependency between the negative mass index of copper corrosion and time in solution 1 and solutions 6–8.
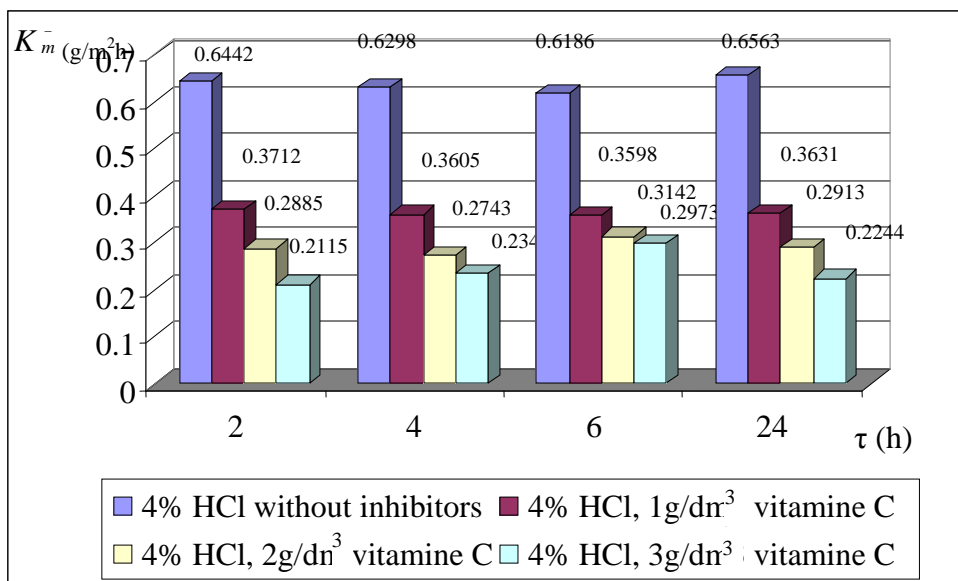
Figure 5 - Representation of the negative mass index of copper corrosion in solution 1 and solutions 6–8 depending on time

It can be seen from Figure 5 that value K_m^- decreases with the increase of the concentration of the inhibitor. It can also be observed that within one concentration of the solution there is no considerable change of the value K_m^- with the passing of time. For example, for the inhibitor concentration of $1g/dm^3$ the value of K_m^- ranges from $0,3598 – 0,3712$ g/m^{2}h.

Figure 6 gives a diagram of the protection factor z (%) depending on time in solution 1 and solutions 6–8.
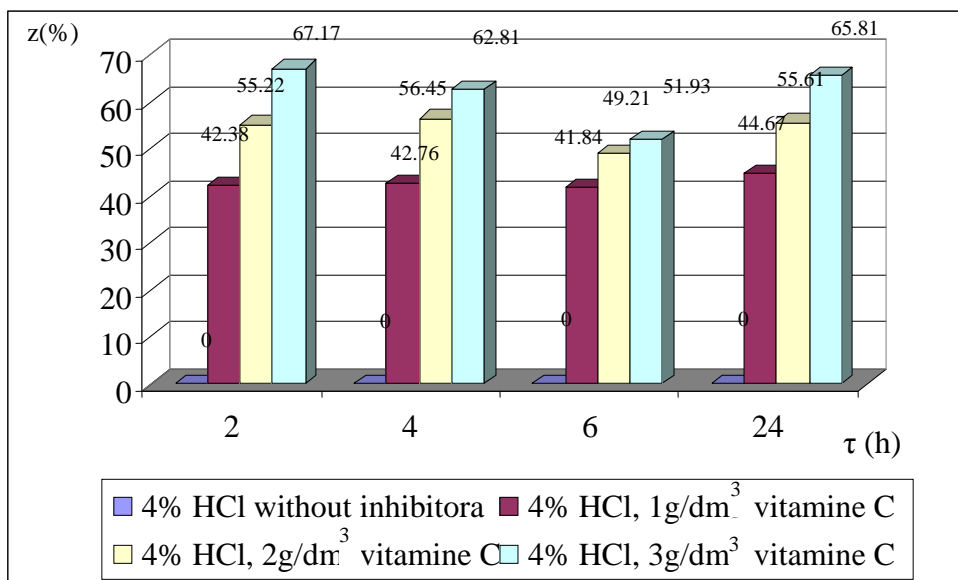

Figure 6 - Protection factor z (%) depending on time in solution 1 and solutions 6–8

Similar conclusion can be drawn as from the previous Figure 5, that with the increase of the concentration of the inhibitor results in the increase of the protection factor of the inhibitor, i.e. smaller loss of mass of copper. It can also be observed that for one concentration of the inhibitor there is no significant change of the value z with the passing of time, i.e. for all four time intervals z , it has approximately the same value.

Figure 7 gives a representation of the dependency between deep copper corrosion index and time in solutions 6–8 and solution 1.

It can be seen from Figure 7 that the value of deep corrosion index π decreases with the increase of the concentration of the inhibitor, as well as that these values do not change significantly with the passing of time.
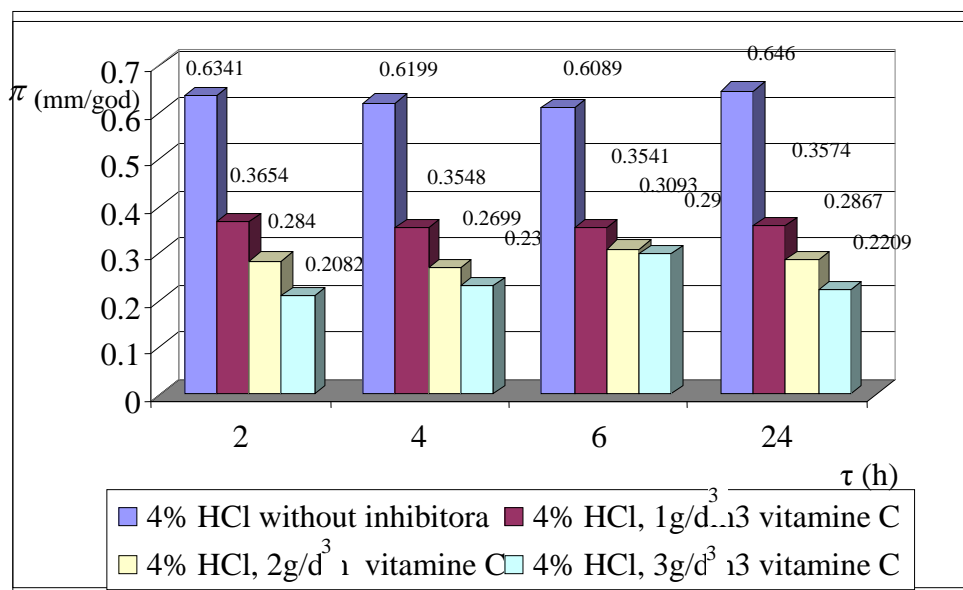

Figure 7 - Representation of the dependency between deep copper corrosion index and time in solutions 6–8 and solution 1

Figure 8 gives a representation of negative mass index of copper corrosion in solution 2 and solutions 9–11 depending on corrosion time.
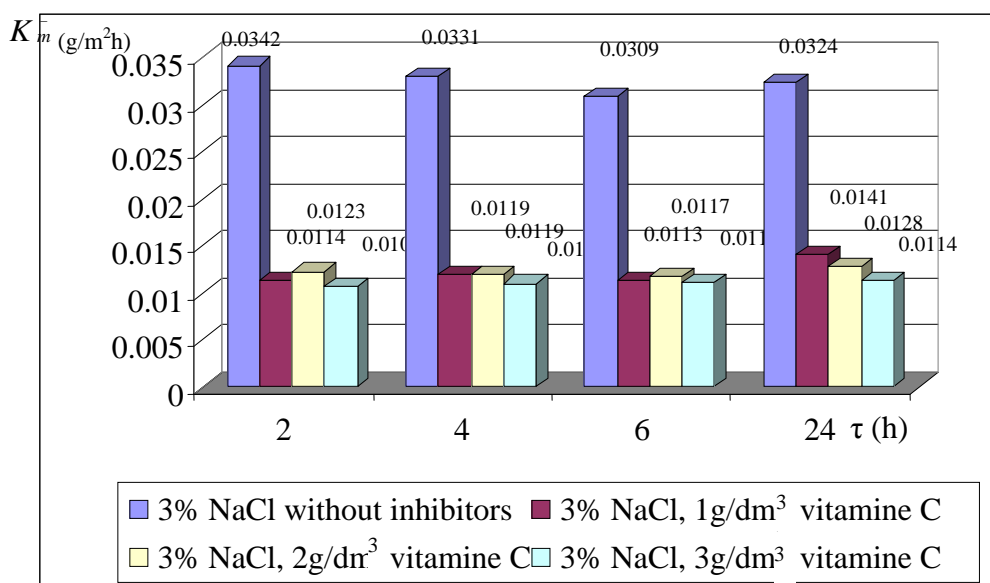

Figure 8 - Representation of negative mass index of copper corrosion in solution 2 and solutions 9–11 depending on corrosion time

Table 3 gives a representation of copper corrosion indices at room temperature in solution 2 and solutions 9–11.

Table 3 - Copper corrosion results solution 2 and solutions 9–11

Corrosion indices	Solution 2 3% NaCl without inhibitors				Solution 9 3% NaCl + 1g/dm³ of vitamine C				Solution 10 3% NaCl + 2g/dm³ of vitamine C				Solution 11 3% NaCl + 3g/dm³ of vitamine C			
τ (h)	2	4	6	24	2	4	6	24	2	4	6	24	2	4	6	24
$\overline{K}_m$ (g/m²h)	0,0342	0,0331	0,0309	0,0324	0,0114	0,0119	0,0113	0,0141	0,0123	0,0119	0,0117	0,0128	0,0107	0,0110	0,0112	0,0114
$\overline{\overline{K}}_m$ (g/m²h)	0,0327				0,0122				0,0122				0,0111			
z (%)	0	0	0	0	66,67	64,05	63,43	56,48	64,04	64,05	62,14	60,49	68,71	66,77	63,75	64,81
$\overline{z}$ (%)	0				62,66				62,68				66,01			
π(mm/year)	0,0336	0,0326	0,0304	0,0319	0,0112	0,0117	0,0111	0,0139	0,0121	0,0117	0,0115	0,0126	0,0105	0,0108	0,0110	0,0112
$\overline{\pi}$ (mm/year)	0,0321				0,0120				0,0120				0,0109			

It can be seen from figure 8 that there are no significant deviations from the value $\overline{K}_m$ with the pasing of time as well as with the change of the concentration of the inhibitor. The values of $\overline{K}_m$ are in the interval from 0,0107 to 0,0141 mm/year.

Figure 9 shows the dependency between protection factor z (%) of copper corrosion and the time of exposition of samples to corrosion medium in solution 2 and solutions 9–11.
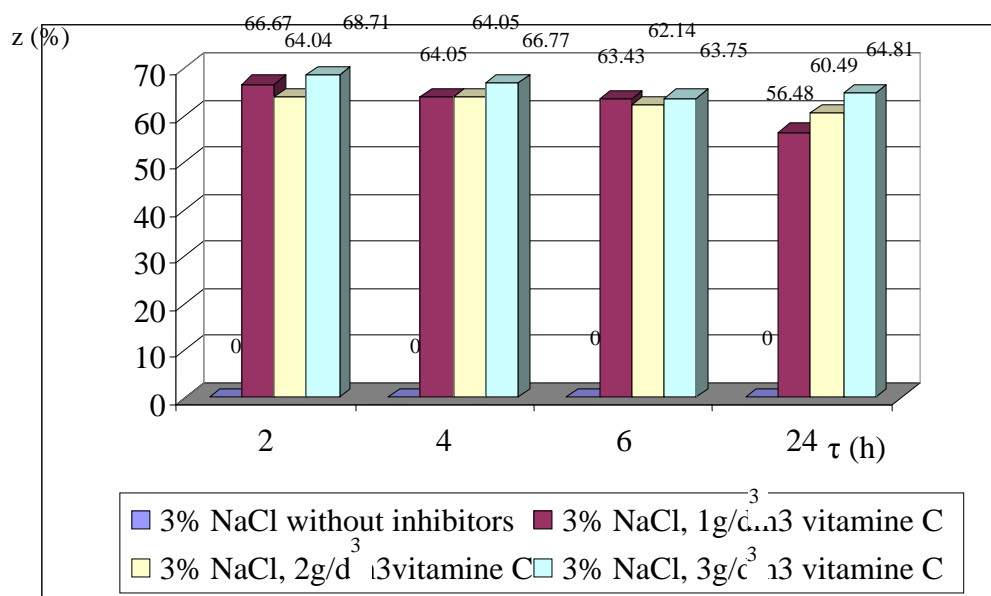

Figure 9 - Dependency between protection factor z (%) of copper corrosion and time

It can be seen form Figure 9 that the value of protection factor z increases with the increase of the concentration of the inhibitor in the solution, but decreases with the passing of time. It can be concluded that vitamin *C* is used up more quickly than other inhibitors used in this paper, so it is necessary to add it more

frequently in order to perform concentration correction. However, this property is less pronounced in NaCl than in the HCl solution.

Figure 10 gives a diagram of deep index for copper corrosion in solution 2 and solutions 9–11 depending on time.
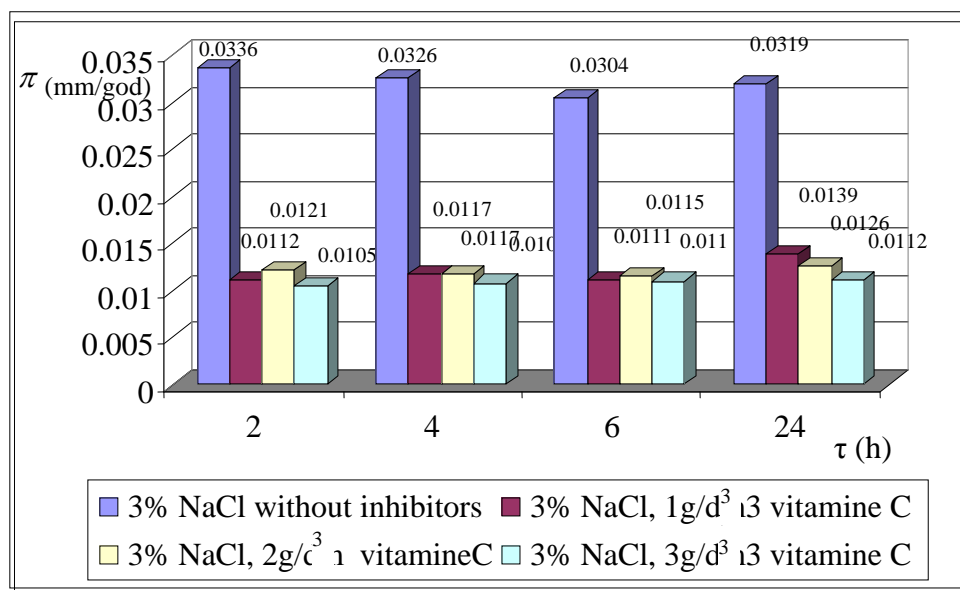

Figure 10. Diagram of deep index for copper corrosion in solution 2 and solutions 9–11 depending on time.

Table 4 - Representation of copper corrosion in solution 2 and solutions 12–14

Corrosion indices	Solution 2 3% NaCl without inhibitors				Solution 12 3%NaCl +1g/dm³ of caffeine				Solution 13 3% NaCl +2g/dm³ of caffeine				Solution 14 3%NaCl +3g/dm³ of caffeine			
τ (h)	2	4	6	24	2	4	6	24	2	4	6	24	2	4	6	24
$\bar{K_m}$ (g/m²h)	0,0342	0,0331	0,0309	0,0324	0,0200	0,0214	0,0206	0,0211	0,0198	0,0192	0,0201	0,0200	0,0190	0,0188	0,0184	0,0179
$\bar{\bar{K_m}}$ (g/m²h)	0,0327				0,0208				0,0198				0,0185			
z (%)	0	0	0	0	41,52	35,35	33,33	34,88	42,11	41,99	34,49	38,27	44,44	43,32	40,05	44,75
$\bar{z}$ (%)	0				36,27				39,22				43,14			
π (mm/year)	0,0336	0,0326	0,0304	0,0319	0,0197	0,0211	0,0203	0,0208	0,0195	0,0189	0,0198	0,0197	0,0187	0,0185	0,0195	0,0176
$\bar{\pi}$ (mm/year)	0,0321				0,0205				0,0195				0,0182			

In this case there is no significant change in the value π with the increase of the concentration of the inhibitor even with the time of exposition of the sample to corrosion medium. The values of π range from 0,0105–0,0126 mm/year.

Table 4 gives a representation of copper corrosion indices in solution 2 and solutions 12–14, in which caffeine was used as an inhibitor in the concentration of 1, 2 and 3g/dm^3.

Figure 11 shows the dependency between the negative mass index of copper corrosion and time in solution 2 and solutions 12–14, Figure 11.
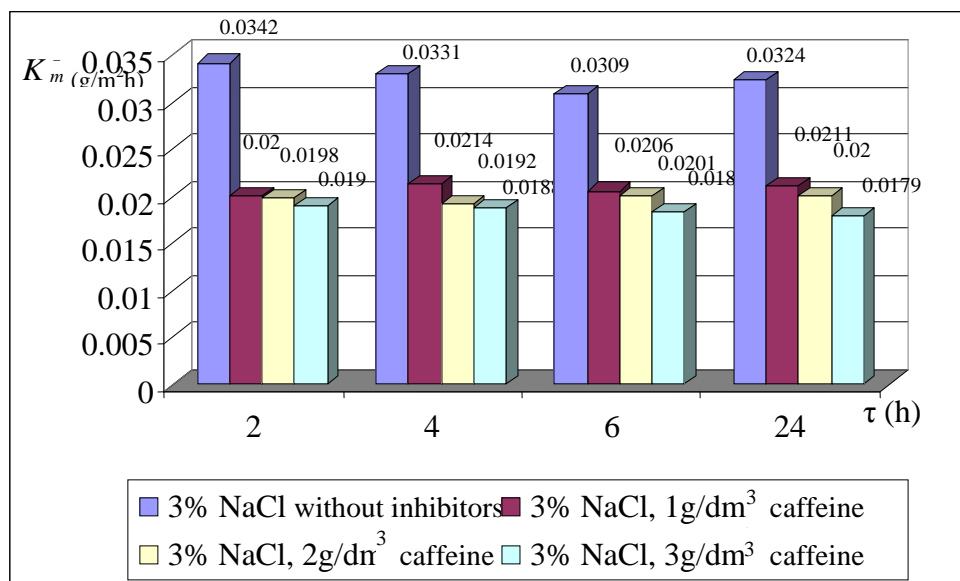

Figure 11 - Dependency between the negative mass index of copper corrosion and time in solution 2 and solutions 12–14

It can be seen from Figure 11 that in the application of caffeine as the inhibitor in copper corrosion, the value K_m^- does not exhibit any significant change with the increase of the concentration of the inhibitor, as well as with the passing of time. This proves that caffeine is stable as an inhibitor with the passing of time in 3% solution of NaCl.
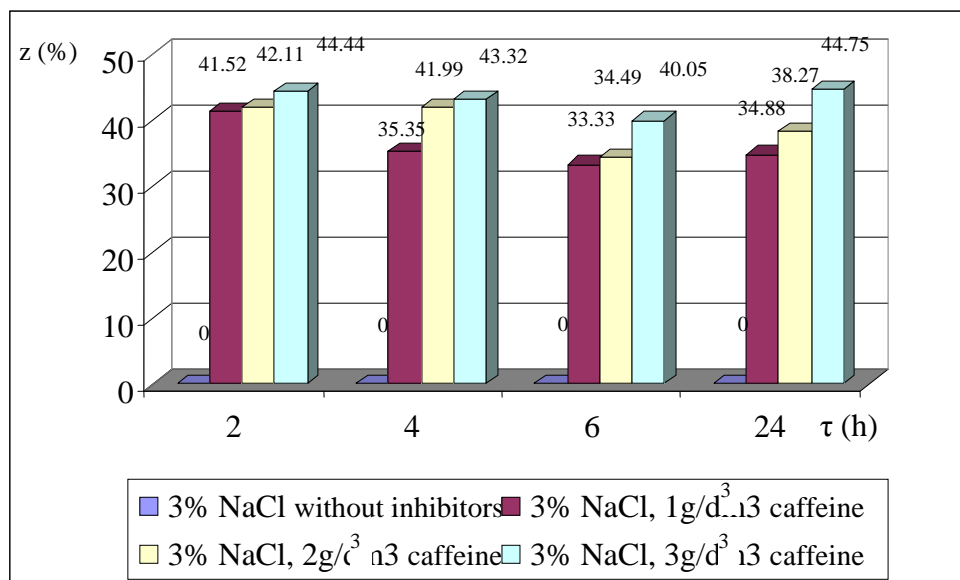

Figure 12 - Representation of protection factor z (%) depending on time in solution 2 and solutions 12–14

Figure 12 gives a diagram of the protection factor z (%) in solution 2 and solutions 12–14 depending on the time of exposition to corrosion medium.

Figure 13 gives a diagram of deep index for copper corrosion in solution 2 and solutions 12–14 depending on time.
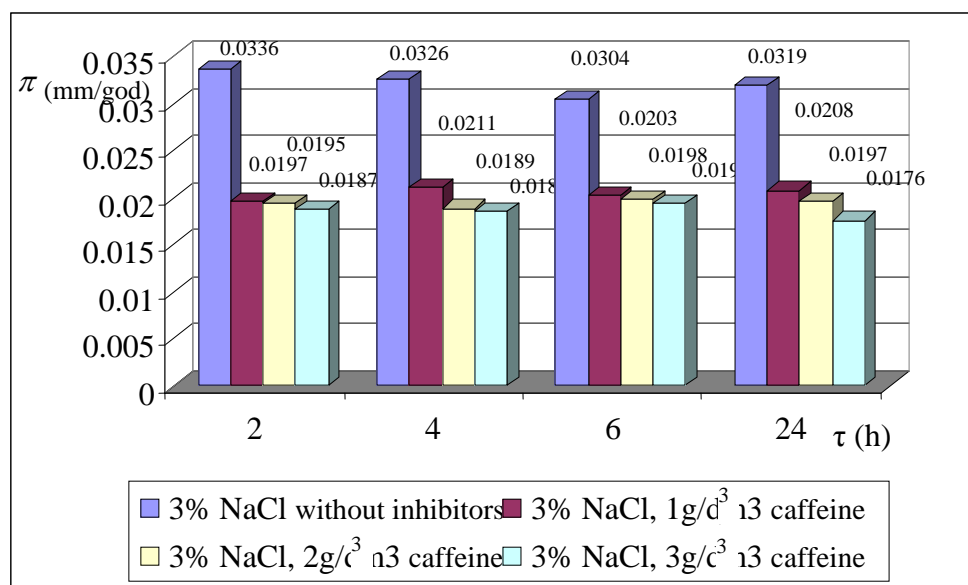

Figure 13 - Diagram of deep index for copper corrosion in solution of NaCl with caffeine as an inhibitor depending on time

It can be seen from figure 13 that the values of deep index of corrosion π do not change significantly with time as well as with the increase in the concentration of the inhibitor and they are in the interval between 0,0176 and 0,0211 mm/year. This confirms the stability of caffeine as an inhibitor in the solution of *NaCl*, as well as that the increase of the concentration of the inhibitor does not affect significantly the rate of copper corrosion in 3% NaCl.

Figure 14 gives a representation of negative mass index of corrosion depending on time in solutions 1 and 15-17.
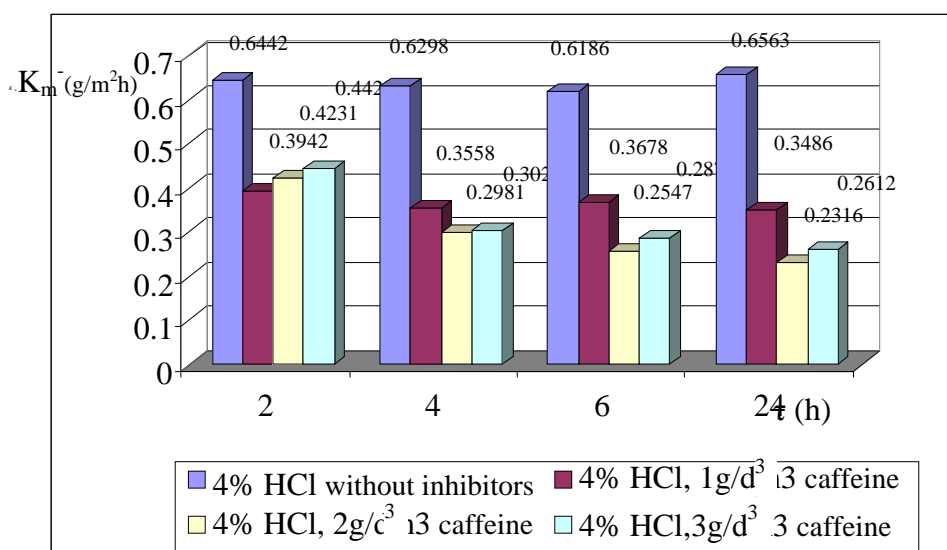

Figure 14 - Representation of negative mass index of copper corrosion in solutions 1 and 15–17 depending on time

Table 5 gives a representation of copper corrosion indices in solution 1 and solutions 15–17.

Table 5 - Copper corrosion indices in solution 1 and solutions 15–17

Corrosion indices	Solution 1 4% HCl without inhibitors				Solution 15 4% HCl + 1g/dm³ of caffeine				Solution 16 4% HCl + 2g/dm³ of caffeine				Solution 17 4% HCl + 3g/dm³ of caffeine			
τ (h)	2	4	6	24	2	4	6	24	2	4	6	24	2	4	6	24
$\bar{K_m}$ (g/m²h)	0,6442	0,6298	0,6186	0,6563	0,3942	0,3558	0,3678	0,3486	0,4231	0,2981	0,2547	0,2316	0,4423	0,3029	0,2874	0,2612
$\bar{K_m}$ (g/m²h)	0,6372				0,3666				0,3019				0,3235			
z (%)	0	0	0	0	38,81	43,51	40,54	46,88	34,32	52,67	58,83	64,71	31,34	51,91	53,54	60,20
$\bar{z}$ (%)	0				42,44				52,63				49,25			
π (mm/year)	0,6341	0,6199	0,6089	0,6460	0,3880	0,3502	0,3620	0,3431	0,4164	0,2934	0,2507	0,2280	0,4353	0,2981	0,2829	0,2571
$\bar{\pi}$ (mm/year)	0,6272				0,3608				0,2971				0,3184			

Results from figure 14 show that $\bar{K_m}$ has different values. For example, after 2 h the values of $\bar{K_m}$ increase with the increase in the concentrtaion of the inhibitor, whereas, in 4, 6 and 24h intervals the lowest value of $\bar{K_m}$ is in the solution with the concentration of caffeine of 2g/dm³. It is also observed for a particular concentration of the inhibitor the values of $\bar{K_m}$ decrease with time which proves that caffeine is a better corrosion inhibitor with the passing of time.

Figure 15 gives a diagram of protection factor z (%) in solution 1 and solutions 15–17.

Figure 15 shows that values of z decrease with the increase of the concentration above 2g/dm³ of caffeine. It can aslo be observed that the degree of protection at these concentrations increases with time.
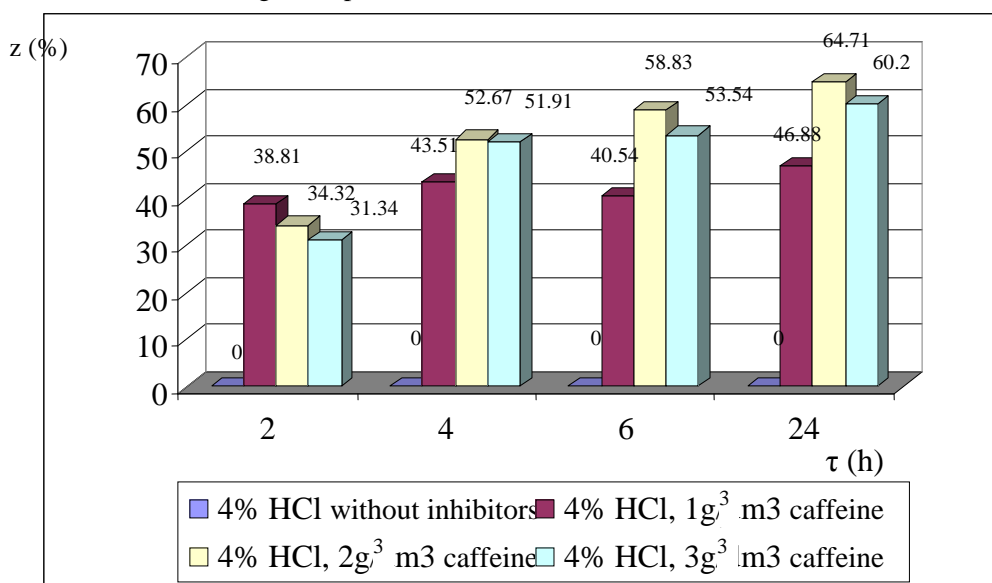

Figure 15 - Representation of protection factor z (%) in solutions 15–17 and solution 1 depending on time

Figure 16 gives a representation of deep copper corrosion index depending on time of exposition of the sample to corrosion medium in solution 1 and solutions 15–17.
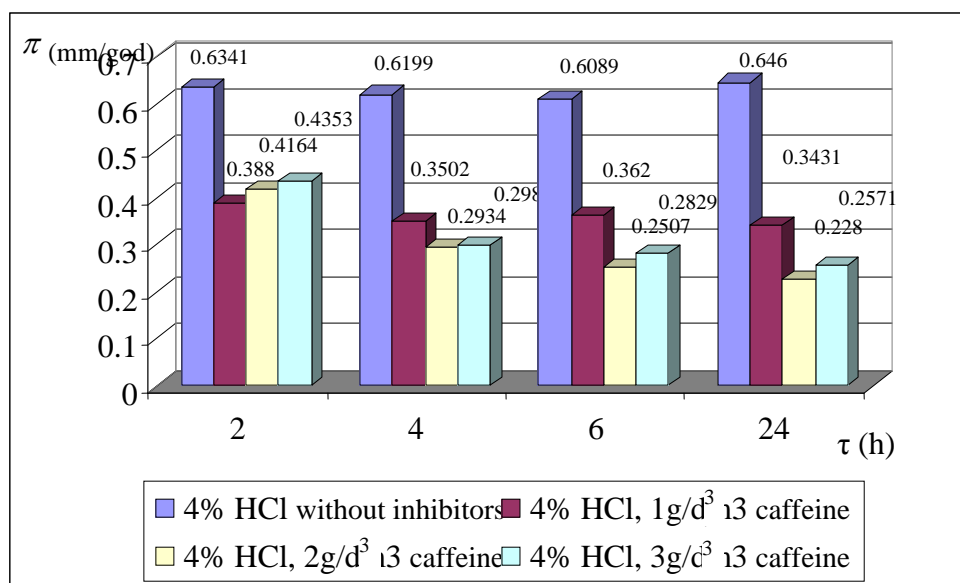

Figure 16 -Representation of deep copper corrosion index in solutions 15–17 and solution 1 depending on time

It can be concluded from Figure 16 that as well as for the prevoius Figures (Figure 14 and Figure 15), the value of deep copper corrosion index does not depend significantly on time and the lowest value is for the concentration of caffeine of $2g/dm^3$ and amounts to 0,228 mm/year.

Figure 17 gives a diagram of the average protection factor $\bar{z}$ (%) in solutions 9-14.

It can be seen from Figure 17 that in solutions 9-14 there is no significant deviations of the value of $\bar{z}$ regardless the concentration of vitamine C as an inhibitor. The average protection factor ranges from $\bar{z}$ =62,66–66,01%. In solutions where caffeine is used as an inhibitor, it can be seen that the value of the average protection factor $\bar{z}$ increases with the increase in the concentration of caffeine, so that it amounts to $\bar{z}$ = 43,14% in solution 14.
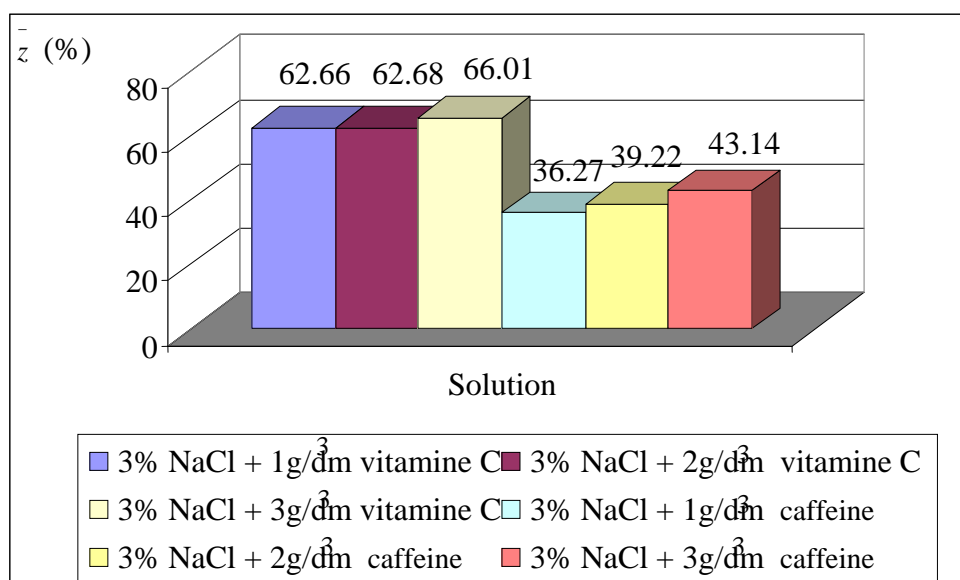

Figure 17 - Diagram of the average protection factor z (%) in solutions 9-14.

Figure 1 gives a diagram of the average protection factor $\bar{z}$ in solutions 3-8 and 15-17.
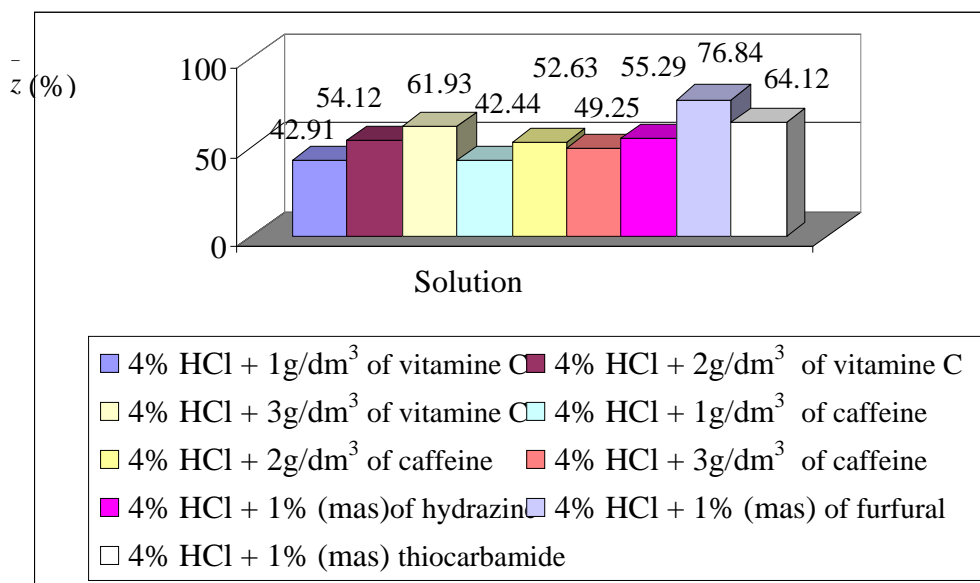

Figure 18 - Diagram of the average protection factor $\bar{z}$ in Solutions 3-8 and 15-17.

It can be seen from Figure 18 that in the case of caffeine and vitamin C the value of protection factor $\bar{z}$ increases with the increase of the concentration of the inhibitor so that it amounts ($\bar{z}$ = 61,93%) in solution 8 and in solution17 it is ($\bar{z}$ = 49,25%).

It can also be seen that in solutions 3-5 the highest value of the average protection factor is for furfural $\bar{z}$ =76,48%, then thiocarbamide $\bar{z}$ =64,12%, the lowest being for hydrazine $\bar{z}$ =55,29%.

CONCLUSION

Caffeine used as an inhibitor in copper corrosion does not exhibit good results. The degree of protection in both solutions is in the interval of 36–53%. If we consider the market value of caffeine and the achieved degree of protection, caffeine does not prove to be an adequate inhibitor for copper protection.

Vitamin C used as an inhibitor in copper corrosion shows better results than caffeine. The degree of protection is in the interval of 40–67%. Vitamin C is cheaper than caffeine, it is available on the market and it is environmetally acceptable. The degree of protection increases with the increase of the concentration of vitamin C both in HCl and NaCl solutions. Vitamin C is non-toxic and therefore may be termed "green inhibitor". All the above said makes it a recommendable corrosion inhibitor for the appli-cation in the near future. The only disadvantage is a short protection time because it decomposes; therefore, it is necessary to perform correction of concentration after two hours in order to keep the concentration in the solution unchanged, which keeps the degree of protection on the same level as well.

Of all the inhibitors used, furfural is the one with the greatest protective action in copper corrosion in 4% HCl. Its average protection factor is $\bar{z}$ =76,48%, however, it is environmentally unacceptable.

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IZVOD

MOGUCNOST PRIMENE ZELENIH INHIBITORA ZA ZASTITU BAKRA

U ovom radu izvršeno je istraživanje zaštite bakra obradom korozione sredine inhibitorima. Uzorci bakra dimenzija (50x50x1) mm nakon hemijske pripreme podvrgnuti su koroziji u neinhibiranim i inhibiranim rastvorima u vremenu od 2, 4, 6 i 24 sata. Korišćena su dva osnovna rastvoraa: 3% NaCl i 4% HCl. Kao inhibitori korišćeni su tiokarbamid, furfural i hidrazin u koncentracijama 0,1%(mas), kao i vitamin C i kofein u koncentracijama $1g/dm^3$, $2g/dm^3$ i $3g/dm^3$.

Od korišćenih inhibitora najbolje zaštitno dejstvo ima furfural, ali i vitamin C u vremenu od 2h. Netoksičnost i dobra zaštitna svojstva vitamin C preporučuju kao inhibitor budućnosti.

Ključne reči: *bakar, inhibitor, korozija, pokazatelji korozije, zaštitni faktor z.*