

UDC: 621.798.144:667.637.2

RESEARCH OF FRUIT CONSERVES' CORROSIVE AGGRESSIVENESS

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Abstract. Corrosion of metal canning containers is one of the obstacles in spreading its application for packing of food. Particularly aggressive to the metal container is fruit canned medium, containing organic acids. The basic material for the production of metal canning container is white tinplate. The main advantage of white tinplate is the tin compounds are harmless to human organism. For this reason, a white badge is used widely, usually used for production of canning containers, packaging beverages. Despite the fact that recently often used containers made of aluminum badge (foil), the basic material for manufacturing metal canning containers is steel white tinplate.

Now applied for coating paints and varnishes do not provide anti-corrosion protection of inner surface of metal containers during storage. Preserving of canned fruit quality in metal containers is largely defined corrosion resistance of the containers. This is due to the fact that the metal transition to canned fruit in due courses of corrosion processes is lowering the nutritional value and deterioration taste of the product, and while allocation of hydrogen is accompanied by swelling and destruction of metal containers. We have investigated a number of anti-corrosion coatings based on Fe-Cr and Fe-Sn-Ti of their behavior in aggressive mediums canned fruit. For the purpose of modeling such mediums the solutions of most widespread organic acids were used. The research allowed conclude, that in surface solid solutions Fe-Sn-Ti increase the corrosion resistance of carbon steel in aqueous solutions of malic, citric and tartaric acids. This implies that the surface solid solutions' formation can significantly improve corrosion resistance in aggressive canning mediums.

Keywords: metal packaging canning, aggressive mediums, an organic acid, corrosion, anticorrosive coatings.

ДОСЛІДЖЕННЯ КОРОЗІЙНОЇ АГРЕСИВНОСТІ ФРУКТОВИХ КОНСЕРВІВ

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Анотація. Корозія металевої консервної тари є однією з перешкод у поширенні її застосування для пакування харчових продуктів. Особливо агресивними до металевої консервної тари є середовища фруктових консервів, що містять органічні кислоти. Попри те, що останнім часом часто використовується тара з алюмінієвої бляхи (фольги), основним матеріалом для виготовлення металевої консервної тари є сталеві білі бляхи.

Нами досліджено ряд антикорозійних покриттів на основі Fe-Cr та Fe-Sn-Ti щодо їхньої поведінки в агресивних середовищах фруктових консервів. Із метою моделювання таких середовищ використовувалися розчини найбільш поширених органічних кислот. Проведені дослідження дозволили прийти до висновку, що поверхневі тверді розчини Fe-Sn-Ti підвищують корозійну стійкість вуглецевої сталі у водних розчинах яблучної, лимонної та винної кислот. Звідси випливає, що формування поверхневих твердих розчинів дозволяє істотно підвищити корозійну стійкість бляхи в агресивних консервних середовищах.

Ключові слова: металева консервна тара, агресивні середовища, органічні кислоти, корозія, антикорозійні покриття.

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This work is licensed under the Creative Commons Attribution International License (CC BY). <http://creativecommons.org/licenses/by/4.0><http://dx.doi.org/10.15673/fst.v11i3.611>**Introduction. Formulation of the problem**

The main material for the production of metal cans is a white badge, that is, a black badge, covered on both sides with a thin, protective layer of tin. Corrosion of metal cans is one of the obstacles to its application to the packaging of food products, in particular of vegetable origin ones, since the latter often contain free organic acids, which causes the mentioned process.

Despite the fact that aluminum foil packaging is often used recently, the main material for the manufacture of metal cans is a steel white plaque for hot lamination and electrolytic disintegration. Acute shortage of tin has led to a reduction in the world production of white hot-dip tinning and a significant increase in the produc-

tion of electrolytic lamination patches, for which requires 2-3 times less tin.

The so-called white tinplate is a sheet steel, coated on both sides with a thin protective layer of tin. Its main advantage is the harmlessness of tin compounds for human organism. For this reason, the white plate is widely used for the manufacture of cans, packaging of beverages. 99.9 % pure tin is safe because the lead content in it usually does not reach 0.1 % and is actually 0.05 %.

Along with the electrolytic lamination plate, new materials for the production of cans are being introduced recently. Among them, the most important is the chromium plated, which is a steel base with an electrolytic coating of chromium with the superfine intermediate

layer of Fe-Cr solid solution. To increase the corrosion resistance of the tinplate, surface solid solutions can be used.

The currently used coating serial enamel EP-5147 and lacquer PL-559 do not provide effective protection against corrosion of the inner surface of metal containers during long storage of various canned fruits and vegetables, especially compotes of dark-colored fruits. Even when, after a certain storage time, the paint coating has unchanged appearance, hydrogen bombing cans with cherry and plum compotes are in place. This defect can be eliminated by applying compositional electrochemical coatings. In developing such coatings of the inner surface of metal containers and setting the terms for preserving preserves, it is necessary before all to take into account the corrosive aggressiveness of the canning mediums, which plays a decisive role in the running products, such as fruit juices, beverages, compotes, etc.

Analysis of Literature

Familiarization with literary sources showed that, despite the existence of a number of fundamental works on the corrosion of domestic and foreign authors (G.V. Akimov, N.D. Tomashov, Ya.N. Kolotyркиn, Yu.N. Mikhaylovsky, V.S. Grzhivo, V. Ande, G. Bataler, D. Dickinson, H. Uhlig, etc.), which are related to the investigation of the process of metal cans' corrosion with fruit canned food, is relatively small. In addition, they contain some controversial data that do not allow the quantification of medium corrosive aggressiveness, what is necessary for the proper selection of protective coatings [1].

In works by E.A. Boytsova, E.A. Andriushchenko, T.N. Ulyanova [2] authors examined the titrated acidity in the calculation of malic acid, pH of juice, and the general corrosion losses of the unfrozen white tinplate ENK-III – the main container material for fruit canned food. The rate of corrosion and corrosion losses are high enough. The paper considers the use of anti-corrosion coatings. The change in the corrosion speed when giving pectin was studied by M.S. Stechyshyn [3]. The formation of composite electrolytic coatings on a nickel base was studied to increase the corrosion resistance of steel. Coverage is subject to different requirements. The general requirements that all coverings must meet are their strong grip with the base. Coatings should have a maximally uniform thickness at different sites, because thickness is the most important characteristic of the coating, because it determines the protective action time of the coating. Nickel-based coatings are much lower than those covered by the coatings developed by us.

In works of G.I. Robsman, A.N. Gorenkova, T.F. Platonova, N.S. Tovstokora [4,5] a new method for assessing the continuity of paint and varnish coatings by measuring the magnitude of corrosion current, which appears in a thermostatically controlled cell, was

developed: the inhibitory ability of phytic acid to corrosion aggressiveness of canned fruits and vegetables was studied.

I.E. Rosenblatt studied the corrosive aggressiveness of canned food for various types of packaging materials [6]. It was found that there is an elevated transition of iron to the syrup and the hydrogen content in the gas phase due to the absence of a continuous coating of enamel EP-5147 on the interior surface of tinplate cans. In aggressive canning mediums, corrosion processes are accelerated due to the shift of electrical potential to more positive meanings, and these processes may cause the release of hydrogen white tinplate to be replaced with aluminum packaging, but the latter has a drawback – a high cost compared to the white tinplate. In works Yu.A. Huslienko [7] the study of the structure and properties of composite electrochemical coatings nickel – chromium diboride is presented. The feasibility of using different compounds containing boron to obtain crisis has no doubt: these substances have a significant reduction potential. Coatings formed with the use of borohydride compounds contain boron in their composition, and this fact opens the prospects for the creation of alloys and with new properties.

Coatings containing compounds with metal-boron bonds are characterized by high hardness, high corrosion and wear resistance, high melting point [6]. The borohydride method allows to obtain coatings at relatively low temperatures (about 40 °C). This makes it possible to apply boron-based coatings with relatively low energy expenditure. Some warnings are caused by a relatively insufficient study of the influence of boron on human organism.

Preserving the quality of canned fruit in a metal container is largely determined by the corrosion resistance of the container itself. This is due to the fact that the transition of metals in fruit canned foods as a result of the course of corrosive processes causes a decrease in nutritional value and a deterioration of the products' taste properties and the allocation with hydrogen is accompanied by bloating and destruction of metal containers.

The corrosion resistance of metal cans depends on the corrosion resistance of metal tare materials, the quality of paint and varnish coatings and the corrosive aggressiveness of canned mediums. Due to the specifics of the technology of tinplate containers manufacture in the production of lacquered metal containers, in most cases it is impossible to exclude the formation of pores and the appearance of mechanical damage to paint and varnish coatings. In this regard, the corrosion resistance of lacquered metal containers in general is determined by the development of corrosion processes in pores and micro-clefts of coatings, that is, the corrosion resistance of metal tare materials.

Currently, the corrosive aggressiveness of canned mediums is usually evaluated by the change of the metal containers' state in the process of canned food storage and the activity of heavy metal transitions from the

packaging to the product. The gravimetric method for determining the corrosion resistance of metal tar materials due to low sensitivity does not always allow a reliable and quick study of the corrosion processes' kinetics, especially for materials with high collagen resistance, such as white tinplate and anodized aluminum tape.

The purpose of the work is to investigate the corrosive aggressiveness of fruit canned food for the choice of the most resistant coating in order to protect metal packaging from corrosion by increasing the stability of the tinplate in the mentioned mediums.

To achieve this goal, it was necessary to solve the following tasks:

- to study the kinetics of metal containers' corrosion in organic acids' solutions;
- to study the degree of influence of various organic acids, which are part of fruit canned food, on the kinetics of metal tar materials' corrosion;
- to develop composite electrochemical coatings to increase the corrosion resistance of tinplate and to investigate their corrosion resistance;

Research Materials and Methods

As a research object, the steel of 08KP brand (the initial base of white tinplate) was taken as well as a white tinplate of electrolytic tinning of the ENK-III brand. The so-called white tinplate is a sheet steel, coated on both sides with a thin protective layer of tin. Its main advantage is the harmlessness of tin compounds for the human organism. Along with the white tinplate of electrolytic tinning recently introduced new materials for the production of cans. Among them, the most important is the chromium plated steel sheet, which is a steel base electrolytically coated by chromium with the finest intermediate layer of Fe-Cr solid solution. For comparison, samples of plaques were used, one of which was covered with nickel with a surface density

of 30-300 mg/m², and another with chromium (5 – 50 mg/m²).

A new type of plaque was developed, on the surface of which an electrolytically alloyed tin with titanium (titanium content was 0.025 – 0.5 %) with a surface density of 100 – 2500 mg/m². The electrodeposition of composite electrolytic coatings (CEC) was carried out at the electrolyte constant mixing. Qualitative coatings with good adhesion can be obtained by electrolysis mode: $I_k = 2 \text{ A/dm}^2$, $t^p = 40 - 50 \text{ }^\circ\text{C}$, the concentration of salts was 0.02 – 0.2 mol/dm³ [8,10]. The main corrosion-active components of fruit canned are organic acids: citric, malic and tartaric. Their concentration can fluctuate within very wide limits. As a model medium, basing on the above-mentioned, aqueous solutions of the most common organic acids that are part of fruit canned: citric (0.01 %), malic (0.5 %) and tartaric (0.02 %) were applied.

The rate of corrosion was investigated by the method of polarization resistance in the cells after the polarization currents, as well as by mass losses, established gravimetrically [9].

The stationary potentials of tin coatings and steel base were measured on the P-5827 potentiostat (an electrode of comparison – saturated calomel). In the model mediums, the transition of iron and tin into the solution was determined by the atomic adsorption spectrophotometry method according to the rate of stationary corrosion, the aggressiveness of fruit canned products was determined, and the corrosion kinetics of container materials in solutions of organic acids, the components of canned fruit [10], was studied.

The results of the research and their discussion

Corrosive processes were controlled by the transition of metals into a solution of aggressive medium. The results of general corrosion losses ($\Delta\rho$, g/m²) and losses of iron and tin ($\Delta\rho_{\text{Fe}}$, g/m² and $\Delta\rho_{\text{Sn}}$, g/m²) are given in tables 1-3.

Table 1. Results of determination of total corrosion losses ($\Delta\rho$, g/m²) and weight losses of iron and tin ($\Delta\rho_{\text{Fe}}$, g/m² and $\Delta\rho_{\text{Sn}}$, g/m²) in citric acid solution ($\omega = 0.01\%$ by weight)

Type of coating	General corrosion losses, $\Delta\rho$, g/m ²	Corrosion losses of iron $\Delta\rho_{\text{Fe}}$, g/m ²	Corrosive losses of tin $\Delta\rho_{\text{Sn}}$, g/m ²
Fe-Ni	6.63	0.06	4.50
Fe-Cr	7.19	0.06	6.28
Fe-Sn-Ti	1.94	0.04	4.92

Table 2 – Results of determination of total corrosion losses ($\Delta\rho$, g/m²) and weight losses of iron and tin ($\Delta\rho_{\text{Fe}}$, g/m² and $\Delta\rho_{\text{Sn}}$, g/m²) in a solution of tartaric acid ($\omega = 0.02\%$ by weight)

Type of coating	General corrosion losses $\Delta\rho$, g/m ²	Corrosion losses of iron $\Delta\rho_{\text{Fe}}$, g/m ²	Corrosive losses of tin $\Delta\rho_{\text{Sn}}$, g/m ²
Fe-Ni	2.37	0.04	4.50
Fe-Cr	2.50	0.04	4.50
Fe-Sn-Ti	1.70	0.02	2.18

Table 3 – Results of determination of total corrosion losses ($\Delta\rho$, g/m²) and weight losses of iron and tin tin ($\Delta\rho_{Fe}$, g/m² and $\Delta\rho_{Sn}$, g/m²) in malic acid solution ($\omega = 0.5\%$ by mass)

Type of coating	General corrosion losses $\Delta\rho$, г/м ²	Corrosion losses of iron $\Delta\rho_{Fe}$, г/м ²	Corrosive losses of tin $\Delta\rho_{Sn}$, г/м ²
Fe-Ni	3.35	0.10	6.50
Fe-Cr	3.45	0.18	6.92
Fe-Sn-Ti	2.42	0.09	6.12

The resulting coatings were tested for corrosion resistance. Data from these studies are presented in Table 4.

Table 4 – Corrosion resistance of steel samples (item 3) with protective coatings in the solution of malic acid ($\omega = 0.5\%$ by mass)

Composition of the coating	Deposition current density D_i , A/dm ²	Depositional temperature $t^\circ C$	Corrosion resistance (Group / score)
Fe-Ni	6	50	high / 2
Fe-Cr	6	50	increased / 3
Fe-Sn-Ti	6	50	very high / 2

Corrosive behavior of the coatings was studied in laboratory conditions. The duration of the research was 3850 hours. Data are presented in Table 5.

Table 5 – Results of laboratory corrosion tests of corrosion-resistant coatings in organic acids' solutions

Coating	Solution of malic acid (0.5 %)		Citric acid solution (0.4 %)	
	40°C, mm/year	80°C, mm/year	40°C, mm/year	80°C, mm/year
Fe-Ni	0.09	0.1	0.15	0.21
Fe-Cr	0.05	0,075	0.06	0.03
Fe-Sn-Ti	0.000	0.001	0.002	0.001
Steel without protection	0.75	0.927	0.753	1.05

Determination of iron losses by a white plate in acid solutions, carried out by atomic adsorption spectroscopy, shows that the largest corrosion losses were in samples of Fe-Cr coatings, that is, the corrosion resistance of Fe-Cr coatings was the smallest.

In order to study the effect of organic acids on metal containers with the proposed protective coatings, a larger range of concentrations of said acids was used.

The largest corrosion losses of metal took place at the malic acid concentration of 0.5 %. The average corrosion rate was 0.113 g/(m²·h). Solutions with a concentration interval of 0.1÷1 % acquire a brown color. At concentrations from 1.5 to 3 %, the average corrosion rate decreases by an order of magnitude and is about 0, 01 g/(m²·h). Solutions remain clear and colorless. High rates of corrosion indicate intensive processes of complex formation of Fe ions with malic acid in the range of dilute solutions. Investigation of stationary potentials of Fe-Cr and Fe-Sn-Ti coatings, the original ones and after corrosion tests ones, allowed to establish that in the concentration area below 1 % Fe-Cr coating has a more electronegative potential than that of the Fe-Sn-Ti coating. This promotes the transition of iron to the solution. At the concentration range of the malic acid solution, more than 1 % the Fe-Sn-Ti coating becomes more electronegative than the Fe-Cr coating and the iron transition into the solution decreases by two orders of magnitude. Thus, for solutions of malic acid,

the most corrosive-active is the concentration of about 0.5 %.

Analysis of data on the coatings' corrosion speed, obtained for solutions with different levels of tartaric acid revealed a not monotonous extreme dependence of the corrosion speed on the solutions' concentration. With an increase in the concentration of tartaric acid from 0.01 to 0.5 %, the corrosion speed and corrosion losses are growing; at the concentration of 0.5 %, the rate of corrosion is maximal, and in further, the corrosion speed growth is slowing down: in the area of concentrations of 2 – 4 %, the growth rate of corrosion and corrosion losses remains unchanged. The analysis of iron content, investigated by atomic adsorption spectroscopy, confirms this regularity. Investigation of stationary potential of Fe-Cr coatings and Fe-Sn-Ti coatings showed that they are also from the concentration of tartaric acid in solutions. At concentrations of this acid below 0.5 %, the Fe-Cr coating has a more negative stationary potential compared to the Fe-Sn-Ti coating. At the concentration range 0.5 – 0.75 %, the stationary potentials of both coatings are fairly close. At higher concentrations of tartaric acid solutions, the Fe-Sn-Ti coating potential becomes more electronegative than of Fe-Cr coating one and the potential difference becomes positive. This is confirmed by a significant decrease in the iron concentration in the tartaric acid solutions. After contact with a solution

of tartaric acid in the pores of the Fe-Sn-Ti coating, the formation of white color corrosion products, having a crystalline structure, and a shift of the Fe-Sn-Ti potential to more negative area is observed. Thus, it is clear from the data obtained that the solutions of tartaric acid are the most corrosive active ones, having a concentration from 0.29 to 1 %.

As well, an analysis of the iron content in solutions of citric acid in the range from 0.1 to 3 %, which was used to investigate the corrosion of the tin packaging with a protective coating. In the course of studies it turned out that the greatest iron transition from the surface of Fe-Sn-Ti coating to the solution occurs at a concentration of citric acid of 0.25 %. The intensity of the iron transition to the solution depends on the magnitude of the potentials for Fe-Sn-Ti and Fe-Cr coatings in various solutions of citric acid. Thus, in dilute solutions of the citric acid with concentrations below 0.75 %, the Fe-Cr coating has a slightly negative initial potential compared to the Fe-Sn-Ti coating that is why the dissolution of iron through the pores in the tin coating took place. With a concentration of citric acid over 1 %, the tin potential becomes more electronegative. In this case, the solubility of tin is due to the reaction of its oxidation and the weakening of the iron dissolution process. After contact with the solution of citric acid in the pores of the Fe-Sn-Ti coating, corrosion products are formed. The formation of corrosion products leads to a shift in the potentials of the Fe-Sn-Ti coating to a more negative area than of the Fe-Cr coating.

From the data obtained, it follows that solutions of citric acid at concentrations below 0.75 % are significantly more corrosive to metal tare materials than solutions with higher concentrations of citric acid. The corrosion speed of Fe-Cr and Fe-Sn-Ti coatings in the solutions of the most common organic acids that are incorporated into fruit canned foods such as malic, tartar and citric, in all cases coincides with the data on the transition of iron into solutions.

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Conclusions

- 1) The kinetics of corrosion processes for Fe-Cr and Fe-Sn-Ti coatings has been investigated.
- 2) The solutions of malic acid are most corrosively active at concentrations of 0.5 – 1.0 % (maximal 0.75 %).
- 3) Tartaric acid solutions are most corrosively active at concentrations of 0.25 – 1.0 % (maximal 0.5 %).
- 4) Citric acid solutions are most corrosively active at concentrations of 0.1 – 0.75 % (maximal 0.29 %).
- 5) It has been established that the choice of coatings for metal containers can be made after corrosion aggressiveness.

Since most of the canned fruits and vegetables are characterized by high acidity, the bulk of these canned foods are packed in containers, the disadvantages of which are fragility and liquefaction in a neutral medium. In the acidic medium, washing out alkali metal ions (sodium, potassium) from the glass as ions and saturation of any solutions of canned mediums in the form of organic acid salts, which are strongly hydrolyzed, and therefore do not conduce to the preservation of various canned products.

Samples with surface solid solutions of Fe-Sn-Ti, like Fe-Cr, had no damage of the coatings' inviolability; the corrosion speed was 0.001 – 0.002 mm/year.

The conducted studies allowed arrive at the following conclusions. Surface solid solutions of Fe-Sn-Ti increase the corrosion resistance of carbon steel in aqueous solutions of malic, citric and tartaric acids. It follows that the formation of superficial solid solutions can significantly increase the corrosion resistance of patches in aggressive canning mediums, to increase the term of products' storage in metal containers.

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ИССЛЕДОВАНИЕ КОРРОЗИОННОЙ АГРЕССИВНОСТИ ФРУКТОВЫХ КОНСЕРВОВ

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Аннотация. Коррозия металлической консервной тары является одним из препятствий в распространении ее применения для упаковки пищевых продуктов. Особенно агрессивными к металлической консервной таре являются среды фруктовых консервов, содержащие органические кислоты. Несмотря на то, что в последнее время часто используется тара из алюминиевой жести (фольги), основным материалом для изготовления металлической консервной тары может быть стальная белая жесьть

Исследован ряд антикоррозионных покрытий на основе Fe-Cr и Fe-Sn-Ti относительно их поведения в агрессивных средах фруктовых консервов. С целью моделирования таких сред использовались растворы наиболее распространенных органических кислот. Проведенные исследования позволили сделать вывод, что поверхностные твердые растворы Fe-Sn-Ti повышают коррозионную стойкость углеродистой стали в водных растворах яблочной, лимонной и винной кислот. Отсюда следует, что формирование поверхностных твердых растворов позволяет существенно повысить коррозионную стойкость жести в агрессивных консервных средах.

Ключевые слова: металлическая консервная тара, агрессивные среды, органические кислоты, коррозия, антикоррозионные покрытия.

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Отримано в редакцію 17.05.2017
Прийнято до друку 14.07. 2017

Received 17.05.2017
Approved 14.07. 2017