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THE GLASS-LIKE GLAZED COATING MADE OF CATHODE-RAY TUBE FACEPLATES CULLET

Н.І. Завгородня, О.А. Півоваров. **Склоподібне глазуроване покриття з екранного склобою відпрацьованих кінескопів.** Тенденцією поточного часу є пошук шляхів доцільної переробки твердих побутових відходів в якості вторинної сировини із однорідними фізико-хімічними та механічними характеристиками з метою ефективного використання ресурсів та зменшення шкідливого впливу на довкілля. В зв'язку з припиненням виробництва моніторів і телевізорів з електронно-променевими трубками, значна частина їх видаляється із вжитку у вигляді габаритних відходів. Кінескопи цих електропристроїв містять цінні складові, серед яких екранне та конічне скло і катодолюмінофори. Існуючі в світовій практиці напрямки утилізації екранного склобою, свідчать за доцільність шляхів переробки цієї цінної вторинної сировини. ***Мета:*** Метою експериментальних досліджень є визначення впливу повної заміни кварцового піску склобоєм екранного скла та використання відновленого із відпрацьованих катодолюмінофорів сульфіду цинку як вторинної сировини при виготовленні склоподібного глазурованого покриття на основні властивості кольорової глазурі. ***Матеріали і методи:*** Екранне скло кінескопів підчас їх демонтажу відрізається, відмивається від бруду, висушується, роздавлюється пресом, доподрібнюється в щоковій дробарці і остаточно подрібнюється в барабанному млині. До цього порошку додаються суспензуюча домішка (глина), барвники бажаного кольору, серед яких ZnS, вода. Отриману суміш піддають мокрому помелу для виготовлення шлікеру. Шлікер наносять на склокерамічну плитку, висушують, піддають випалу при максимальній температурі 900ºC. ***Результати:*** Експериментальними дослідженнями встановлено, що із склобоєм екранного скла в глазурь вводяться склоутворюючі, модифікуючі та проміжні оксиди неорганічних речовин. Часів'ярівська глина відноситься до групи зі значним газовиділенням. Водяна пара, що виникає при дегідратацію глини відіграє роль 'носія' важких нелетких компонентів, значно прискорює газові процеси і підвищує активність газових компонентів. Сульфід цинку розчинний у розплавах силікатної глазурі і при нагріванні майже до температури випалу надає їй забарвлення. Розраховані за законом адитивності властивості отриманої глазурі підтверджують задовільну її якість. ***Висновки:*** Повна заміна кварцового піску склобоєм екранного скла при виготовленні склоподібного глазурованого покриття дозволила отримати гомогенний розплав скла з нижчою температурою плавлення, більш розм'якшеним і менш в'язким, із задовільними розрахунковими показниками властивостей глазурі. Використання відновленого сульфіду цинку як вторинної сировини в якості додатку до барвників кольорової глазурі обумовлено розчинністю його в розплавах силікатної глазурі.
Ключові слова: глазуроване покриття, екранний склобій, відновлений сульфід цинку, склоутворюючі оксиди, дегідратаційне структуроутворення.

N.I. Zavgorodnya, A.A. Pivovarov. **The glass-like glazed coating made of cathode-ray tube faceplates cullet.** The tendency of the current time is to find ways of expedient municipal solid waste recycling as a secondary raw material with similar physicochemical and mechanical characteristics for the purpose of efficient use of resources and reduction of harmful impact on the environment. Due to the termination the production of monitors and television sets with cathode-ray tubes, a significant part of them is grow out of use in the form of dimensional waste. Kinescopes of these electric devices contain valuable components including the screen and conical glass and cathode-luminophors. Existing trends in the world of CRT faceplates cullet recycling argue for reasonability of recycling ways of this valuable secondary raw materials. ***Aim:*** The aim of researches is to determine the impact of the full replacement of quartz sand by faceplates cullet and using the zinc sulfide, reconstituted of used cathode-luminophors, as a secondary raw material in the production of glasslike glaze on the basic properties of color glaze. ***Materials and Methods:*** Cathode-ray tube faceplates are cut off during removal process, washed from dirt, dried, crushed by press, milled in a cheek grinder and finally crushed in a barrel mill. The slurried impurity (clay), dyes of desired color, including ZnS, water are added to this powder. The received mix is processed of wet grinding for slip production. Slip is surfaced on glass-ceramic tile, dried up, burned at maximum temperature of 900ºC. ***Results:*** Experimental research has shown that glass-forming, modifying and intermediate oxides of inorganic substances are added to the glaze with the CRT faceplates cullet. The Chasiv Yar clay belongs to the group with significant gas emission. The water vapor arising during the clay dehydration plays role of the "carrier" of heavy non-volatile components, considerably accelerates gas processes and increases activity of gas components. Zinc sulphide, dissolved in the silicate glaze melts when heated almost to the calcining temperature, gives it color. Properties of the obtained glaze, calculated according to the additivity law, confirm its satisfactory quality. ***Conclusions:*** Complete replacement of quartz sand by cathode-ray tube faceplates cullet in the production of glasslike glaze yielded a homogeneous glaze melt, shift the melting point with less viscous and satisfactory estimate indicators of glaze. The use of reconstituted zinc sulfide as a secondary raw material in the form of additives to the dyes of colored glaze caused by its solubility in melts of silicate glaze.
Keywords: glaze, faceplates cullet, reconstituted zinc sulfide, glass-forming oxides, dehydration structurization.

Introduction. Glass-like glazed coatings, or glazes, are intended for application onto surfaces of ceramic, glass-ceramic, porcelain, earthenware and other products. Currently, the technology of glaze

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producing requires development of the production methods of the cheap raw materials of the ordinary content which would help in reducing the temperature and extending service life of glass-melting furnaces owing to decrease in refractory erosion, and also maintaining the high quality of molding machines operation. Significant role in the expansion of glass-like coatings sphere of use (made of cheap raw materials) is given to the studies and the latest developments of the various colored glaze charging components. The results, obtained in studies of the situation in the system of handling the cullet of used TV-sets and monitors with cathode-ray tubes, positive findings of experimental studies related to development of glass-ceramic tiles' manufacturing technology with the replacement of quartz sand by 50 wt % of the powder of CRT screen glass waste, obtaining of zinc sulfide as a secondary raw material of luminophores of such CRT [1] promoted the experimental investigations with regard to use of only screen glass waste in the working mixture of glass-like colored glazed coating, completely replacing the content of quartz sand (86 wt %) in the mixture with 6 wt % (suspending admixture) of Chasiv Yar clay and 8 wt % of the pigment of the desired color, among which 2 wt % of zinc sulfide is used as a secondary raw material obtained from the luminophores waste [2].

It is known, in 2008 the world famous companies reported stopping of the monitors and TV-sets production with cathode-ray tubes (CRT). At present time, the most of CRTs are removed from use as large-sized solid wastes, instead of their recycling [3]. In the opinion of the environmental specialists, all CRT should be recovered because these devices are the source of harmful heavy metals (for example, strontium, lead etc.). However, it is believed that no serious studies were conducted so far and no enough statistical data is available to make such conclusions. Nevertheless, recycling of CRT is required not only by the reason of their environmental damage, but also because they contain significant quantities of useful materials, which recovery would save the natural resources [4].

The CRT consists of 87% glass on average. Therefore, the existing lines of CRT recycling relate to recycling of screen glass and conical glass. Usage of cullet is reduced to its recycling at glass-making enterprises, although the manufacturers prefer natural resources. Other direction is the usage of construction materials in production. The serious issue is separation of barium-strontium glass and lead glass. As a whole, the companies still believe that landfilling of CRT is cheaper than their recycling. However, millions of tons of electron-tube glass are forced to seek the governmental support and to enhance the competitiveness of cathode-ray tubes glass [9]. No data about recycling of luminophores is available, in spite of the fact they comprise valuable inorganic substances [5]. Brief analysis shows that experimental studies of recycling the glass and cathode luminophores allow finding the scientific and technical solution of used TV-set CRTs and computer monitors reworking problem.

The aim of this experimental study is to determine the impact of the full replacement of quartz sand by faceplates cullet and using the zinc sulfide reduced of used cathode-luminophores, as a secondary raw material in the production of glass-like glazed coating on the basic properties of color glazed coating.

Materials and Methods. Preparation of the powder of screen glass waste. After manual dismantling of printed circuit boards, speakers, wires, protective metal casing, deflection system, and electron gun, CRT screens are cut off the cone along the line of welding using the diamond wheel; luminophore is cleaned away by the laboratory vacuum cleaner; screens are washed of process contaminations and dried to prepare for crushing and milling. Screens are crushed under press with the specific pressure of 3 t/cm^2, grinded in jaw crusher with the gap of 0.1…1.5 mm until the powder passes through the sieve No. 02 and finally are milled in the barrel mill with the porcelain grinding media. 6 wt % of Chasiv Yar clay which chemical composition (and composition of glass) is stated in Table 1 are added as a suspending admixture to 86 wt % of screen glass waste powder (0.1 mm), with 8 wt % of the colorant of required color, including 2 wt % of the reduced zinc sulfide as a secondary raw material of luminophores taken from used CRTs. The mixture is placed into porcelain drum, with 50 wt % of water added, and subject to wet grinding for 2 hours. Ready-made slurry is kept in tightly plugged containers.

Table 1

Chemical composition of Chasiv Yar clay and screen glass

Name of the substance	Chemical composition, %						
	SiO_2	Ba_2O_3	Al_2O_3	Na_2O	K_2O	CaO	Fe_2O_3
CRT screen glass	68.5...75.4	2.8...3.0	4.0...4.4	10.0...11.0	7...8.4	–	–
Chasiv Yar clay	52.3...52.6	–	30.0...33.12	0.6...0.7	2.5...2.6	0.5...0.7	0.8...1

Slurry is applied onto fired glass-ceramic tiles by pouring or spraying methods. Products are dried in the drying furnace at the temperature of 80 °C during 0.5 hours. Then they are fired in silicon carbide furnace at maximum temperature of 900 °C. Firing mode: slow increase of the temperature from the room temperature to 900 °C, holding for 0.5 hour, and cooling to the room temperature with the furnace off.

Results. During the glass-like glazed coating forming on the base (ceramic, glass ceramic tiles etc.) of slurry mass, various physical and chemical processes are running both in the base and in the boundary layer. During a number of experimental studies, it has been established that the main processes were: dehydration of mill admixtures (in our case, of Chasiv Yar clay), reaction in solid phases — beginning and end of baking, appearance of the liquid phase together with gloss, interaction of the melt with mass of the base and formation of glazed coating. Chemical composition of screen glass waste (Table 1) proves, that two glass-forming oxides of inorganic substances SiO_2 and Ba_2O_3 are added to glaze (glass) together with screen glass waste. And they are capable of filling the continuous randomly placed three-dimensional grids at cooling of glass melts. The Na_2O and K_2O oxides are modifying ones, because their action is aimed for the glass grid loosening but not the filling. The Al_2O_3 oxide is considered as an intermediate oxide, because it does not form glass itself, but it is involved in formation of glass grid (tetrahedral groups of AlO_4 can substitute SiO_4 tetrahedrons in the silicate lattice).

The Chasiv Yar clay belongs to the group with significant gas emission. In its clay fraction the fine clay particles prevail over large and medium-sized ones. These clays actively release gases up to maximum temperature (dehydration of mill admixtures occurs at the temperature of 450…650 ºC). According to [7], gas phase contained in the pores and cavities of clay contain, %: CO_2 within the limits of 0.44…89.97; CO 3.08…52.67; H_2 1.49…32.13; O_2 2.05…19.66; N_2 3.06…59.08; SO_2 0.25…7.69. Since depending on the specific conditions the same components can release gas at various temperatures, it is explained by chemical, catalytic, physical-chemical and dynamic effect of water vapor which appears during dehydration of clay. Water vapor acts as a "carrier" of the heavy nonvolatile components (Al_2O_3, SiO_2 etc.), accelerates gas processes considerably, increases the activity of gas components. Dehydration structurization is referred to one of the optimal criteria of the integrated method of resource-saving technologies intensification.

Investigation of the glass-mass interaction mechanism revealed the following factors: chemical composition of the glaze and mass of the fragment, their physical properties (wetting ability, glaze viscosity, surface tension) and firing conditions — temperature, holding period, and environment. Since the chemical composition of slurry components for the glazed coating and glass ceramics are of the same type, the role of forming oxides and clay additives is disclosed, the main physical and chemical properties of glazed coating are calculated according to the additivity rule [6], according to which any specific property of glass (glaze) is considered as a linear function of the percentage composition of its specific components. Each property is determined as a sum of product of the percentage composition of each glass-forming oxide by the relevant constant, expressing the effect of that oxide onto the given property of glass (glaze). The additivity rule can be written by general formulas as below:

$$x = a_1 \cdot k_1 + a_2 \cdot k_2 + \ldots + a_n \cdot k_n \quad (1)$$

or

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$$x = \frac{1}{100}(a_1 \cdot k_1 + a_2 \cdot k_2 + \ldots + a_n \cdot k_n), \quad (2)$$

where x — quantitative value of the specific property;

$a_1, a_2, \ldots, a_n$ — percentage of glass-forming oxide;

k — constant of the particular oxide.

The basic constants of additivity of the main glass-forming oxides are given in [6] (Table 2), averaged partial estimated constants of linear expansion coefficients a; oxides in silicate glass (glazes) according to Appen are indicated in Table 2. To calculate the partial estimated constant for SiO_2 (since the estimated constants of the linear thermal expansion for that oxide have variable values) we used the formula below:

$$a_{SiO_2} \cdot 10^{-7} = 38 - 1.0 \cdot (A_{SiO_2} - 67), \quad (3)$$

where A — molecular percentage of silica in glass or glaze,

a_{SiO_2} — averaged partial estimated constant of the linear expansion coefficient for SiO_2.

Thermal conductivity of the glaze was calculated by the formula:

$$Q = \lambda \cdot \frac{A \cdot (T_1 - T_2)}{S} \cdot t, \quad (4)$$

where Q — amount of heat, J;

t — time for which amount of heat passes through the composite bar, s;

A — bar surface area, m^2;

S — bar thickness, m;

λ — thermal conductivity coefficient, W/m·K;

T_1, T_2 — temperature on bar surface, ºC.

Coefficient of glass (glaze) thermal resistance was calculated according to Winkelman and Schott formula [6]:

$$F = \frac{p}{\alpha \cdot E} \cdot \sqrt{\frac{\lambda}{d \cdot C}}, \quad (5)$$

where F — coefficient of glass thermal resistance;

p — tensile strength;

α — linear expansion coefficient;

E — modulus of elasticity;

λ — thermal conductivity coefficient;

C — heat capacity (J/kg·K);

d — specific weight of glaze (glass).

All these parameters are determined according to the additivity rule.

It should be noted that additivity coefficients given in Table 2 according to [6] represent the estimated coefficients for the relevant physical and chemical characteristics of glass (glaze). The calculated values of glaze properties are stated in Table 2.

It is known that wetting as a kind of liquid-solid interaction, predecessor of dissolution and diffusion, is found in our study in the form of spreading the glaze drops on the base surface or, on the contrary, folding of glaze films into drops (outflow). Interfacial friction at the liquid-solid interface has a significant impact on the process of glaze spreading on the surface of the base, since the molecules of only one, i.e. liquid, phase are moving only. However, it is necessary to take into account the nature of the solid phase which can change the force of friction. For spreading of the liquid glazed coating on the surface of ceramic, glass-ceramic, and metallic solid surface the force of friction is different, and it will produce the effect on the aggregate value of internal and interfacial force of friction, on the counteraction to spreading. Besides, the possibility of forming quasi-solid film which resists spreading of viscous glaze, as a result of surface orientation of molecules, crystallization of components, and evaporation of volatile components, etc. is taken into account. The effect of factors of purely chemical nature cannot be excluded as well: the nature of solid surface of the base is changing after contacting the molten glaze, depending on duration of the contact.

Table 2

Estimated and determined values of glaze properties

No.	Property	Quantitative value	Unit of measure-ment	Note
1.	Linear thermal expansion coefficient	$83.32 \cdot 10^{-7}$	K^{-1}	
2.	Heat capacity	0.787	J/kg·K	
3.	Thermal conductivity	108	W/m·K	
4.	Specific weight	2.46	N/m^2	
5.	Tensile strength at break	8.1	MPa	
6.	Compression strength	11.85	MPa	
7.	Modulus of elasticity	6.9	MPa	
8.	Hardness	44.81	$kg \cdot s/mm^2$	
9.	Impact strength	–	–	Absence of cracks
10.	Coefficient of thermal resistance	2.924	$m^2 \cdot K/W$	
11.	Glaze surface quality class	1		Absence of blisters and pinholes

In the experimental studies the contact angle of glaze spreading on the surface of glass-ceramic tiles was equal to ~30…76 °C. Since glass serves as a source material of the glaze and glass-ceramic base, it is relatively easily "wetting" other surfaces in the liquid or plastic state.

As it is known [7], coloring of the glaze is a physical and chemical process during which oxides-colorants interact with the silica in the melt to form the relevant silicates which provide certain coloring of the glaze. Such colorants are called as molecular dyes; glaze with this type of colorants is a true solution.

Ability to absorb the light and coloring of the glaze associated with it, with the same oxide-colorant, are varying if one basic oxide in the oxide molecule is replaced by the other one. With the growing atomic weight of the basic oxide in the glaze molecule the absorption spectrum shifts from red to purple, i.e. from the spectrum of long waves to the short wave range.

Composition of the base glaze also has effect on coloring with the same oxide colorant.

For oxides with variable valence the dependence of the glaze color on the degree of its oxidation and size of the cation radius is typical (oxide with the less radius of the cation provides the higher transparence of glaze for rays with the longer wave, and vice versa). From the point of view of crystal chemistry, the higher coordination of the colorant element provides the coloring that is transparent to longer wave rays, while the closer coordination gives coloring that is transparent to short-wave light rays.

According to [6] sulfides of metals, including zinc and cadmium sulfides are soluble in the melts of silicate glass (glaze) and can be substituted at cooling in the solution, so that glass (glaze) is obtained transparent. Heating of such glass (glaze) to the temperature that is almost equal to the firing temperature provides coloring to glass (glaze). The best example is glass (glaze) containing zinc sulfide or cadmium sulfide. After melting and cooling they remain almost colorless, but upon heating they gain on thick white and yellow colors, accordingly. If oxides-colorants are added to such glazes, the glaze gains on the corresponding color of the colorant.

Since the sulfides fall out in glass (glaze) as colloid particles, these compounds may serve as the valuable catalysts of nuclei of glass (glaze) microcrystals formation. According to the literature data [6, 9], easily melted zinc glazes are studied insufficiently. However, it is known that zinc oxide formed according to reactions (6 and 7) is the necessary component of easily melted glaze, since it and promotes decreasing the thermal expansion coefficient and can act as a flux.

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$$ZnS+5H_2O \rightarrow ZnO+SO_4^{2-}+10H^+ +8e \quad (6)$$

$$ZnS+H_2O \rightarrow ZnO+S^{2-}+2H^+ \quad (7)$$

Mineralogical and radiographic studies prove that two types of crystal formations are distinguished in zinc glazes: microcrystalline zinc aluminate — halite $ZnAl_2O_4$, which provides for dullness of the glaze, and coarse-grained zinc silicate — willemite. Size of halite crystals is about 1 micron, while the length of willemite crystals is 50...80 micron. In zinc glazes with the melting point lower than 2000 °C, where magnesium oxide is replaced by sodium and potassium oxides, crystalline phase is insignificant, mainly comprising willemite. Whiteness of such glaze is reduced from 80 to 30 %. For such easily melted glaze bluish tint is typical. As in this case, where the amount of alkali metals exceeds 11 %, zinc glazes gain on intense blue color which is associated with the dispersity of crystalline formations [8…10].

Conclusions. Complete replacement of quartz sand by cathode-ray tube faceplates cullet in production of glass-like glazed coating yielded a homogeneous glazed coating melt, shift the melting point with less viscous and satisfactory estimate indicators of glaze. The use of reduced zinc sulfide as a secondary raw material in the form of additives to the dyes of colored glazed coating caused by its solubility in melts of silicate glaze.

Usage of the reduced zinc sulfide as a secondary raw material being added to the colorants of the colored glaze is conditioned by solubility of zinc sulfides in the melts of silicate glass (glaze) and its ability to gain on a distinct pure color at high temperature.

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