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# DEVELOPMENT OF A COMPOSITION FOR FOOTWEAR USING SECONDARY RECYCLED MATERIALS

*The object of this study is the process of forming a coating on a chromium-tanned split leather semi-finished product. The study is aimed at developing an optimal formulation of a finishing composition for the production of footwear upper leather.*

*A technology has been developed for forming a decorative coating on chromium-tanned split hides from pigs and heavy cattle. The composition of the finishing formulation was determined through computer modeling and multiparameter optimization using Harrington's desirability function. Based on the analysis of the physico-mechanical properties of the resulting monolithic films, the qualitative composition of the film-forming finishing composition was established.*

*Computer-aided modeling of the "composition – property" system for a three-component formulation, using the Scheffé mathematical model, enabled the derivation of analytical relationships between the physico-mechanical properties of the finishing composition and its constituents. The optimal composition was determined at the maximum values of the desirability function and physico-mechanical parameters through multiparameter optimization.*

*The leather produced using the optimal composition was tested under industrial conditions and complies with DSTU 2726-94 and DSTU 3115-95, for upper footwear leather and leather for garment production, respectively. The use of a highly porous chromium-tanned split leather semi-product derived from pig hides ensures the production of high-quality, elastic upper leather.*

*The developed finishing technology for split hides of pigs and heavy cattle demonstrates significant potential for use in the manufacture of everyday footwear.*

***Keywords:*** *chromium-tanned split leather, polymer films, multiparameter optimization, physico-mechanical properties of leather.*

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## 1. Introduction

The finishing and final property formation of leather and fur materials represents one of the key stages in leather and fur production technology. At this stage, the active interaction between the collagen of the semi-finished chrome-tanned leather and functional chemical reagents leads to the development not only of physico-chemical, but also consumer-relevant properties. Addressing this issue requires computer-based modeling and multiparameter optimization of the finishing composition.

Particular attention is given to formulations incorporating secondary resources from leather and fur processing. Due to the significant volume of such waste generated during production, the effective utilization of secondary resources is a pressing concern.

A wide range of polymer-based finishing compositions with diverse chemical structures is employed in the final stage of forming the physico-chemical properties of leather materials [1]. Among them, polymer systems based on polyacrylates and polyurethanes have demonstrated the greatest effectiveness. Acrylic polymers enhance the hardness, flexibility, and resistance of coatings to solvents and environmental factors, while polyurethanes provide high elasticity and a set of essential physico-mechanical properties, such as abrasion resistance, tear strength, and frost resistance of the coatings.

The study by [2] investigated the effects of an exopolysaccharide and a structuring agent – basic chromium sulfate – on the physico-mechanical properties of polymer films, establishing that modified films exhibited enhanced mechanical strength, elasticity modulus, and reduced elongation at break. Research into polyurethane-based coatings, particularly the SZPU-7 ATZ variant, revealed a lower swelling degree compared to the acrylic-based MBM-3 films, attributable to a higher degree of chemical cross-linking in the polyurethane systems. Self-healing coatings with polyurethane containing disulfide bonds in the main chain have also been developed [3]; surface defect correction was recommended at 60°C over 12 hours.

Other studies have reported on the use of multifunctional organic-inorganic polymer-based nanocomposites for leather finishing [4]. Effective application of polymer composite materials requires mathematical modeling and multiparameter optimization of their chemical formulations. For example, [5] explored the effect of a composition based on exopolysaccharide, polyacrylate, and polyurethane on the physico-mechanical and protective properties of finished leather. Optimization was carried out using a D-optimal simplex-lattice Kiefer design, resulting in a formulation that provided high performance.

In [6], optimization of a polymer composition comprising polyvinyl alcohol, urethane prepolymer, and 2,4-toluene diisocyanate was performed using a modified Scheffé simplex-lattice design, yielding a fourth-order polynomial model. The optimized formulation demonstrated strong resistance to extreme service conditions, including excellent water resistance. Study [7] applied an orthogonal central composite experimental design to optimize the composition of oligomer ED-20 and polyethylene polyamine, resulting in a formulation with high operational performance.

Further, study [8] examined the effects of the acrylic composition Syntan RS-540 in combination with modified emulsions Synthol LC and Sulphirol EG 60 on Wet Blue semi-finished leather. The optimized formulation facilitated effective final treatment conditions, enabling the production of elastic leather with a desirable combination of physico-chemical properties and efficient raw material use.

Efficient use of technological reagents in leather and fur manufacturing requires the rational selection and optimization of their composition and processing conditions. The choice of optimization strategies – especially for finishing composition formulations – is driven by the need to significantly reduce the number of experimental trials.

Of particular significance is the integration of secondary production resources [9] into finishing compositions, such as leather powder in pigment concentrates. It is noteworthy that the volume of secondary resources generated globally in the leather industry may reach up to 50 billion tons annually [10]. This fact underlines the relevance of improving existing and developing innovative technologies for leather and fur production.

*The aim of this research* is to develop an optimized formulation of a finishing composition for the production of upper footwear leather. The following objectives were set:

– to assess the effect of pigment concentrate on the properties of polymer films;
– to perform computer modeling and optimization of the finishing composition;
– to determine the physico-mechanical properties of the finished leather produced under industrial conditions.

## 2. Materials and Methods

*The object of this research* is the process of forming a coating on a chromium-tanned split leather semi-finished product. *The subject of the study* is the effect of the components of the finishing composition on the physico-mechanical properties of the finished leather.

The experiments utilized chromium-tanned split semi-finished leather derived from pig hides and heavy cowhides (heifers), manufactured in accordance with TU-U 00302302391-03-98, with a thickness of 1.2–1.4 mm. The pig hide split was characterized by the presence of through-holes from removed bristles, while the heavy heifer split exhibited structural non-uniformity.

For final finishing, an optimized pigment concentrate (PC) composition [11] was used, consisting of the following components (wt. %): leather powder – 14.6%, alkylcarboxyethanolamines – 3.2%, polyvinyl alcohol (10% solution) – 6.9%, and water – 75.3%.

The film-forming agents of the finishing composition included a mixture of MBM-3 emulsion and polyurethane dispersion Melio Promul 66A (Clariant, Germany). MBM-3 is a copolymer of methacrylate and butyl acrylate containing 3% methacrylic acid, with a solids content of 38.5% and pH 4.35. The anionic aqueous dispersion of polyurethane Melio Promul 66A had a solids content of 32.5% and pH 8.0.

Prior to topcoat finishing, the pig and heifer splits were primed with a polyurethane latex (PUL) composition with the following chemical formulation (wt. %): pigment concentrate (40%) – 20.0%; polyurethane latex (20%) – 80.0%; and water to a density of 1.07 g/cm$^3$. This PUL composition contained 39.0% polyurethane, exhibited a viscosity of 0.022–0.03 Pa·s, surface tension of 340–370 N/m, particle size of 10 $\mu$m, and pH 3.0–3.5.

Multiparameter optimization of the finishing composition for coating the split leather semi-finished product included acquisition of a priori data, selection of the Scheffé "composition-property" model, and design and execution of an experimental plan. Analytical relationships were established between the properties of filled films and the composition of the finishing system, and the formulation was optimized using the Harrington desirability function.

The physico-mechanical properties of the finished semi-finished leather and model films were evaluated according to standardized procedures [12]. Specifically, the mechanical properties of the films and leather were determined using tensile testing machines RMU-0.05-1 and RM-250M at deformation rates of 50 mm/min and 90 mm/min, respectively.

Meridional strength, elongation, and plasticity of the finished leather were measured using a rapid leather testing device. Abrasion and flex resistance of the coatings were assessed using devices IPK-1 (without additional load) and IPK-2 (at a flexing frequency of 100 cycles/min), respectively. All film and leather samples were conditioned for 24 hours under standard laboratory conditions prior to testing.

## 3. Results and Discussion

To optimize the formulation of the finishing composition, monolithic films were prepared using the film-forming agent MBM-3, the polyurethane Melio Promul 66A, and a pigment concentrate. The films were cast in Teflon molds at a temperature of 20°C and subsequently heat-treated at 60°C for 3 hours. The results of physico-mechanical testing of the films, with a thickness of 100 ± 10 $\mu$m, are presented in Fig. 1, 2.

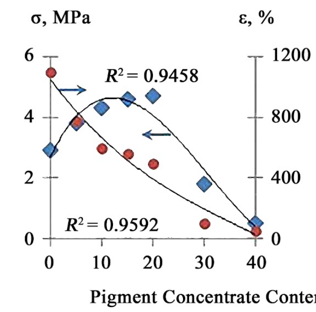


**Fig. 1.** Effect of pigment concentrate on the physico-mechanical properties of MBM-3 film

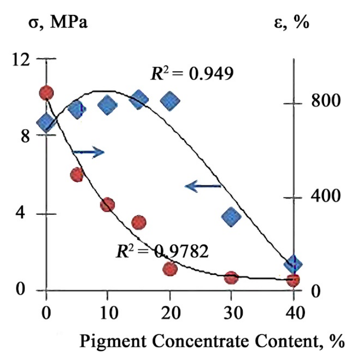


**Fig. 2.** Effect of pigment concentrate on the physico-mechanical properties of Melio Promul 66A film

As shown in Fig. 1, the tensile strength of MBM-3 films increases with increasing pigment concentrate (PC) content, reaching a maximum at a 20% concentration. Beyond this level, strength sharply decreases. Simultaneously, the elongation at break drops to 10% and remains low beyond the 20% pigment concentration, which includes leather powder as a component.

A similar trend was observed in the tensile strength of polyurethane films based on Melio Promul 66A (Fig. 2), although the absolute values were approximately twice as high as compared to MBM-3 films. The same effect was noted for elongation at break in the polyurethane films.

These findings indicate strong interaction between the film components and a pronounced structuring effect of the fillers on the properties of the film-forming agents. This supports the feasibility of using a mixture of MBM-3 and Melio Promul 66A dispersion in the multiparameter optimization of the finishing composition.

In the computer modeling of the finishing composition, the formulation variables (factors) were as follows: MBM-3 emulsion – $x_1$, polyurethane dispersion Melio Promul 66A – $x_2$, and pigment concentrate – $x_3$. The effectiveness of the finishing composition was evaluated using the following physico-mechanical properties of the composite films and coatings:

– Modulus of elasticity at 100% elongation – $y_1$ (MPa);
– Tensile strength – $y_2$ (MPa);
– Elongation at break – $y_3$ (%);
– Wet rub resistance – $y_4$ (cycles).

Based on prior studies, constraints for the formulation factors of the finishing composition were established (Table 1)

$$0 = g_i \le x_i \le h_i = 1, \qquad (1)$$

where $g_i$ and $h_i$ represent the lower and upper experimental bounds for factor $x_i$, respectively, with $i=1$–3.

**Table 1**

Constraints for the formulation factors

| Factor | $g_i$ | $h_i$ |
|---|---|---|
| $x_1$ | 0.7 | 1.0 |
| $x_2$ | 0 | 0.3 |
| $x_3$ | 0 | 0.3 |

To model the properties of the three-factor finishing composition, a third-order incomplete Scheffé polynomial model was selected

$$\hat{y} = b_1x_1 + b_2x_2 + b_3x_3 + b_4x_1x_2 + b_5x_1x_3 + b_6x_2x_3 + b_7x_1x_2x_3, \qquad (2)$$

where $b_i$ are the unknown coefficients of the model, and $x_i$ are the formulation factors.

To execute the experimental design within the domain defined in equation (1) using $x$-coordinates, the region was transformed into a $z$-coordinate system, where the vertices of the simplex correspond to the formulation factors, interpreted as pseudo-components. In the transformed coordinate system, the domain is defined as $0 \le z_i \le 1$ for $i=1, 2, ..., k$, $z_1^{(u)} + z_2^{(u)} + \ldots + z_k^{(u)} = 1$, where $u$ is any point within the experimental region. The relationship between the coordinates of the original ($x$) and transformed ($z$) systems $(x_1, x_2, \ldots, x_k)$ and $(z_1, z_2, \ldots, z_k)$, which satisfy these constraints, is defined by a matrix equation $X = AZ$

$$\begin{Vmatrix} x_1^{(u)} \\ x_2^{(u)} \\ \vdots \\ x_k^{(u)} \end{Vmatrix} = \begin{Vmatrix} x_1^{(1)} & x_1^{(2)} & \cdots & x_1^{(k)} \\ x_2^{(1)} & x_2^{(2)} & \cdots & x_2^{(k)} \\ \vdots & \vdots & \cdots & \vdots \\ x_k^{(1)} & x_k^{(2)} & \cdots & x_k^{(k)} \end{Vmatrix} \times \begin{Vmatrix} z_1^{(u)} \\ z_2^{(u)} \\ \vdots \\ z_k^{(u)} \end{Vmatrix}. \qquad (3)$$

In equation (3), the elements of matrix **A** represent the coordinates of the vertices of the simplex, $x_i^{(u)}$ and $z_i^{(u)}$ $(i=1, 2, ..., k)$ – correspond to the original and transformed coordinates of the $u$-th point. The component structure of matrix **A**, based on the data from Table 1, is presented in Table 2.

To design the experiment, the working $x$-plan (Table 3) was calculated based on the standard $z$-plan using the matrix transformation equation (3). The experimental results corresponding to this plan are presented in Table 4.

**Table 2**

Vertices of the constrained simplex domain

| Simplex Vertex | $x_1^{(i)}$ | $x_2^{(i)}$ | $x_3^{(i)}$ |
|---|---|---|---|
| 1 | 1.0 | 0 | 0 |
| 2 | 0.7 | 0 | 0.3 |
| 3 | 0.7 | 0.3 | 0 |

**Table 3**

Working experimental plan

| Experi-ment | Plan | | | | | |
|---|---|---|---|---|---|---|
| | Pseudo-components | | | Working | | |
| | $z_1$ | $z_2$ | $z_3$ | $x_1$ | $x_2$ | $x_3$ |
| 1 | 1.0 | 0 | 0 | 1.0 | 0 | 0 |
| 2 | 0 | 1.0 | 0 | 0.70 | 0 | 0.30 |
| 3 | 0 | 0 | 1.0 | 0.70 | 0.30 | 0 |
| 4 | 0.5 | 0.5 | 0 | 0.85 | 0 | 0.15 |
| 5 | 0.5 | 0 | 0.5 | 0.85 | 0.15 | 0 |
| 6 | 0 | 0.5 | 0.5 | 0.70 | 0.15 | 0.15 |
| 7 | 0.333 | 0.333 | 0.333 | 0.80 | 0.10 | 0.10 |

**Table 4**

Experimental results

| Result | Experiment | | | | | | |
|---|---|---|---|---|---|---|---|
| | 1 | 2 | 3 | 4 | 5 | 6 | 7 |
| $y_1$ | 0.3 | 1.8 | 0.8 | 2.8 | 0.59 | 3.2 | 2.7 |
| $y_2$ | 2.9 | 1.8 | 5.7 | 4.6 | 4.5 | 6.2 | 5.8 |
| $y_3$ | 1100 | 100 | 806 | 560 | 890 | 550 | 656 |
| $y_4$ | 250 | 200 | 428 | 450 | 350 | 470 | 448 |

To determine the coefficients of the mathematical model (2), the least squares method was applied in matrix form [13]

$$B = \left(F^T F\right)^{-1} F^T Y = DF^T Y,$$

where $B$ is the vector of unknown coefficients; $Y$ – the column vector of experimental values of the dependent variable; $D = \left(F^T F\right)^{-1}$ – dispersion matrix of the design; $F_{nl}$ – experimental design matrix, generalized according to the model type; $\tilde{f}(\bar{x})$; $T$ – matrix transposition operation; $l$ – number of coefficients in mathematical model (2).

Following the processing of experimental data using a custom-developed software application, the following regression models were obtained:

$$\hat{y}_1 = 0.2999999z_1 + 1.8z_2 + 0.8000001z_3 + 7.0z_1z_2 + \\ + 0.1599995z_1z_3 + 7.599999z_2z_3 + 2.642542z_1z_2z_3;$$

$$\hat{y}_2 = 2.9z_1 + 1.8z_2 + 5.7z_3 + 8.999999z_1z_2 + \\ + 0.8000011z_1z_3 + 9.799998z_2z_3 + 4.424406z_1z_2z_3;$$

$$\hat{y}_3 = 1100z_1 + 99.99994z_2 + 806z_3 - 159.9998z_1z_2 - \\ - 251.9998z_1z_3 + 388.0001z_2z_3 - 252.8486z_1z_2z_3;$$

$$\hat{y}_4 = 250z_1 + 200z_2 + 428z_3 + 899.9999z_1z_2 + \\ + 43.99998z_1z_3 + 623.9999z_2z_3 - 494.1759z_1z_2z_3.$$

To verify the adequacy of the obtained models, additional control experiments were conducted according to Student's $t$-test [11], as presented in Tables 5, 6.

**Table 5**

Coordinates of control experiments

| Experiment | $x$-system | | | $z$-system | | | $\xi$ |
|---|---|---|---|---|---|---|---|
| | $x_1$ | $x_2$ | $x_3$ | $z_1$ | $z_2$ | $z_3$ | |
| 1 | 0.900 | 0.050 | 0.050 | 0.667 | 0.167 | 0.167 | 0.4480 |
| 2 | 0.800 | 0.150 | 0.050 | 0.333 | 0.167 | 0.500 | 0.6982 |
| 3 | 0.700 | 0.250 | 0.050 | 0.000 | 0.167 | 0.833 | 0.6298 |
| 4 | 0.800 | 0.050 | 0.150 | 0.333 | 0.500 | 0.167 | 0.6982 |

**Table 6**

Results of control experiments

| Result | Control experiment | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | 1 | | 2 | | 3 | | 4 | |
| $y_1$ | 1.73 | 1.69 | 1.93 | 1.97 | 2.02 | 2.05 | 3.09 | 3.06 |
| $y_2$ | 4.68 | 4.62 | 5.75 | 5.67 | 6.41 | 6.46 | 5.36 | 5.30 |
| $y_3$ | 848 | 851 | 757 | 760 | 742 | 736 | 527 | 533 |
| $y_4$ | 380 | 384 | 417 | 423 | 469 | 477 | 453 | 449 |

The obtained mathematical relationships $\hat{y}_1 - \hat{y}_4$ for the composition of the finishing system were found to be adequate at the 5% significance level based on validation at the control points. This served as the basis for their use in identifying the optimal region using the Harrington desirability function [11]

$$D = \sqrt[v]{d_1 d_2 \dots d_v},$$

where $d_i$ ($I = 1, 2, \dots, v$) are the individual desirability functions corresponding to the target responses $y_i$, taking values in the interval [0; 1].

A multiparameter search was carried out to determine the most effective formulation of the finishing composition corresponding to the maximum value of the desirability function. The solution to the multiparameter optimization problem based on Harrington's generalized desirability function was performed with consideration of the "worst" and "best" a priori values of the physico-mechanical properties of the composite films and coatings. In particular, the optimization took into account the following reference values: modulus of elasticity – 2.0 and 3.0 MPa; tensile strength – 3.0 and 6.0 MPa; elongation at break – 500 and 700%; wet rub resistance – 400 and 500 cycles. The resulting optimal formulation of the finishing composition in $z$-coordinates corresponded to the mass fractions: $z_1 = 4.172325 \cdot 10^{-7}$, $z_2 = 0.3799999$, $z_3 = 0.6199997$. In real coordinates, the composition was as follows: $x_1 = 0.700$, $x_2 = 0.186$, $x_3 = 0.114$.

The effect of the composition on the physico-mechanical properties of the polyacrylic-urethane films filled with pigment concentrate is shown in Fig. 3, where the optimal region is bounded by the physico-mechanical characteristics of the compromise zone.

At a desirability function value of 0.54, the optimal formulation of the finishing composition corresponded to the following performance values:
– Modulus of elasticity: 3.0 MPa;
– Tensile strength: 6.5 MPa;
– Elongation at break: 630.0%;
– Wet rub resistance: 490.0 cycles.

Thus, through computer modeling and multiparameter optimization, the composition of the pigmented polyacrylic-urethane finishing system was determined. The optimized composition included, in mass fractions:
– MBM-3 acrylic emulsion – 70.0%;
– Melio Promul 66A polyurethane dispersion – 19.0%;
– Pigment concentrate based on leather powder – 11.0%.

The optimized finishing composition was tested under industrial conditions in the production of elastic upper leather from chromium-tanned split semi-finished pig and cattle hides. The coating process for pig hide semi-finished leather began with double sanding using a sanding-dedusting machine, employing abrasives No. 6 and No. 3, respectively.

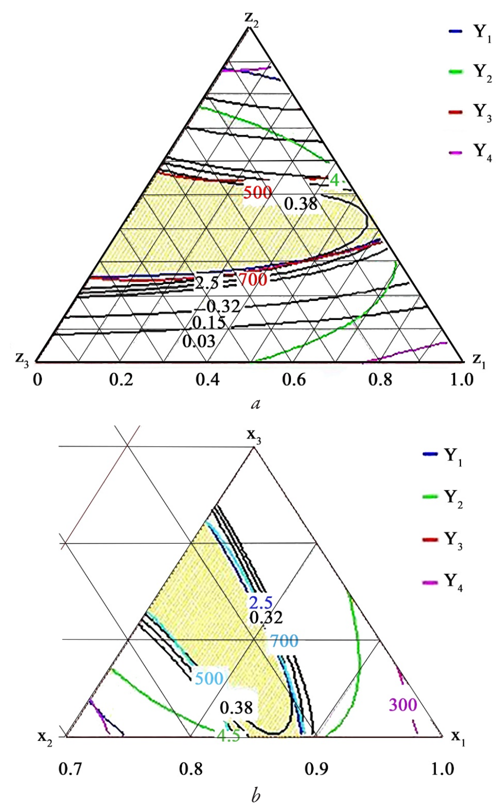


**Fig. 3.** Optimal region of physico-mechanical properties for the polyacrylic-urethane composition: *a* – *z*-coordinate system; *b* – *x*-coordinate system (main figure)

A non-pigmented polyurethane latex (PUL) impregnation composition was then applied to the prepared split surface in two passes using a spraying unit, with an application rate of 70.0 g/m$^2$ per pass.

After conditioning the impregnated semi-finished leather for 8–10 hours at 18–20°C and partial drying to a moisture content of 14–16%, it was pressed using a hydraulic press at 15 ± 1 MPa and 75 ± 5°C. A pigmented PUL priming composition was then applied in the same manner, followed by pressing at 11 ± 1 MPa and 65 ± 5°C.

The topcoat was applied in three passes: 140 g/m$^2$ for the first layer and 40 g/m$^2$ for each subsequent layer. The finishing process concluded with nitrocellulose sealing and pressing.

In the case of heifer hide semi-finished leather, impregnation was applied in a single pass, followed by pressing at 5 MPa lower pressure and 5°C lower temperature than for pig hides. The pigmented primer was applied in two passes without intermediate drying.

After sorting and measuring the area of the resulting leather, samples were selected for physico-mechanical testing. Control samples were produced using the standard technology of PJSC Baryshivka Leather Plant (Baryshivka, Ukraine).

The test results are presented in Table 7. The coatings on leather from heifer hides demonstrated slightly superior physico-mechanical properties compared to the control samples, with the exception of color fastness to wet rubbing.

A similar trend in coating properties was observed in leather derived from pig hides. Virtually identical physico-mechanical properties were recorded for leather produced from cattle hides and pig hides, both in the experimental and control processes. Notably, the overall physico-mechanical performance of the finished leather obtained using the developed technology meets the requirements for elastic upper footwear leather as specified by relevant standards.

The results of industrial trials involving the finishing of chromium-tanned split semi-finished leather from pig and heifer hides confirm the feasibility of using the optimized acrylic-urethane composition for the manufacture of elastic upper leathers. The porous and structurally heterogeneous nature of the split semi-finished material required preliminary priming prior to application of the finishing system.

**Table 7**

Physico-mechanical properties of finished leather

| Property | Leather | |
|---|---|---|
| | cattle hides | pig hides |
| Adhesion to leather semi-finished product, N/m | 490/450 | 540/500 |
| Meridional strength, N | 400/380 | 430/400 |
| Wet rub resistance, cycles<br>– Flex resistance, thousand cycles | 450/400<br>79/64 | 430/390<br>77/63 |
| Color fastness to rubbing, grade:<br>– dry<br>– wet | <br>5/4<br>4/4 | <br>5/4<br>4/4 |
| Tensile strength under uniaxial deformation, MPa | 14.6/14.0 | 14.2/14.0 |
| Elongation at 10 MPa stress, % | 32.0/30.0 | 34.0/32.0 |
| Meridional strength, N | 480/470 | 470/450 |
| Meridional elongation at rupture, % | 36.0/34.0 | 37.0/36.0 |
| Plasticity, % | 30.0/31.0 | 35.0/36.0 |

**Note:** the numerator represents data for the experimental sample, and the denominator corresponds to the control sample

To further expand the scope of the present study, additional investigations should be conducted to evaluate the influence of differently colored leather powders in the pigment concentrate on the full spectrum of physico-chemical properties of leathers produced from other raw materials. It should be noted that the optimized formulation may be effectively applied to finish semi-finished leathers with non-uniform, high-porosity structures.

## 4. Conclusions

A finishing composition has been developed for coating chromium-tanned split semi-finished leather derived from pig and heavy heifer hides. The formulation of the finishing system was determined using computer modeling and multiparameter optimization based on Harrington's desirability function.

Monolithic films were formed using film-forming agents, including polyacrylate MBM-3, polyurethane, and a pigment concentrate containing dyed leather powder. Analysis of the physico-mechanical properties of these films enabled the identification of an effective qualitative composition for the finishing system.

Computer-based modeling of the "composition-property" relationship in a three-component system, utilizing the Scheffé mathematical model, yielded analytical relationships between the formulation components and the resulting physico-mechanical properties. At maximum values of the desirability function and performance parameters, the optimal composition was determined via multiparameter optimization.

Leather produced using the optimized finishing composition and tested under industrial conditions meets the requirements of DSTU 2726-94 and DSTU 3115-95 for upper footwear leather and leather goods, respectively.

The results of the conducted research demonstrate the feasibility of effectively utilizing secondary resources from leather production in the manufacture of leather and fur materials. Furthermore, the use of a high-porosity chromium-tanned split semi-product from pig hides allows for the production of high-quality, elastic upper leather.

The developed finishing technology for split pig and heifer hides shows strong potential for use in the manufacture of everyday footwear.

## Conflict of interest

The authors declare that they have no conflict of interest related to this study, including financial, personal, authorship, or other aspects that could have influenced the research or its outcomes as presented in this article.

## Financing

This research was conducted without financial support.

## Data availability

This manuscript is associated with data stored in a repository, as it is a continuation of previous research in which one of the ingredients – the pigment concentrate – was previously optimized [11].

## Use of artificial intelligence

The authors confirm that no artificial intelligence technologies were used in the preparation of this work.

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