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THEORETICAL FOUNDATIONS FOR ENSURING THE MECHANICAL STRENGTH OF PAPERS CONTAINING COLLAGEN HYDROLYSATES

ESHBAEVA ULBOSIN
Professor, Namangan State Technical University, Namangan, Uzbekistan
Phone.: (0890) 805-9955, E-mail.: guli-67@mail.ru
*Corresponding author

GÖKHAN ZENGIN
Professor, Ege University, Izmir, Turkey
Phone.: (90232) 311-4407, E-mail.: gokhan.zengin@ege.edu.tr

BAHRI BAŞARAN
Professor, Ege University, Izmir, Turkey
Phone.: (90533) 494-8834, E-mail.: bahri.basaran@ege.edu.tr

Abstract: The study explores the influence of fibrous raw materials, refining processes, and adhesive substances on the development of strength properties in paper and cardboard products. Special attention is given to the role of fiber morphology, cellulose refining, and interfiber bonding in improving paper quality. The paper highlights that refining significantly alters the surface structure of fibers, leading to increased fibrillation and enhanced bonding potential, which directly affects the structural and mechanical properties of the paper web. In particular, the study emphasizes the contribution of collagen hydrolysate, an organic additive, in forming fibrillar structures within cellulose and modified polyacrylonitrile (MPAN) fibers, thereby strengthening interfiber adhesion. The findings underline the dual role of structural-mechanical and chemical factors in advancing the physical and mechanical properties of paper.

Keywords: Paper strength; cellulose refining; fiber fibrillation; interfiber bonding; fibrous raw materials; pulp properties; collagen hydrolysate; adhesives; paper web structure; mechanical properties of paper.

Introduction. The development of the pulp and paper industry is closely linked to the processing of fibrous raw materials and improving the strength properties of paper and cardboard products. This process is referred to as enhancing the quality indicators of paper and cardboard. This term is considered one of the fundamental concepts in the science of paper production and utilization.

The papermaking properties of fibrous semi-finished materials are determined by the characteristics of the resulting pulp mass, the properties of the paper (or cardboard), and the adhesives used. These include various parameters of the fibrous semi-finished product (cellulose, fibrous pulp, fillers, adhesives, etc.). The quality indicators of most fibrous semi-finished materials are influenced by both internal and external factors during the papermaking process [1–3].

The strength of paper primarily depends on several factors: the morphology of the fibrous pulp (such as the average fiber length, linear density, etc.), the degree of cellulose refining (i.e., the extent to which the fiber suspension is processed and fibrillated), the strength properties of individual fibers, the interfiber bonding forces via fibrils, and the adhesive properties of the binding substances.

Fiber refining is considered one of the most important stages in the papermaking process, as the chemical and physical properties of paper largely depend on it. Refining

(or the degree of fiber treatment) leads to changes in the surface structure and integrity of plant-based or secondary fibers. As a result, fibers become shortened and undergo both internal and external fibrillation. During fiber treatment, numerous active hydroxyl and carboxyl groups are formed, which facilitate the creation of bonds between fibers. Thus, the higher the degree of fiber fibrillation, the greater the number of interfiber bonds formed, which directly affects the structure and properties of the paper [4].

2. Materials and Methods. During the refining process of fibrous pulp, the structure of the fibers undergoes significant changes, which in turn affect their interaction both with each other and with other components of the paper pulp. These changes influence the formation process of the paper web as well as the paper-making and mechanical strength properties of the fibrous semi-finished products.

In this study, the effect of external fibrillation of fibers, as influenced by cellulose, textile industry waste, and adhesive substances, on the strength properties of paper is emphasized multiple times. Various methods are used to quantitatively describe the external and internal surface characteristics of the fibers within the pulp. Determining the specific surface area of fibers and the interrelation between key papermaking properties such as tensile strength and durability allows for predicting the deformation characteristics of paper web formed under different intensities of refining.

There are two main factors that contribute to the improvement of the physical and mechanical properties of paper: structural-mechanical and chemical.

The bonding strength between fibers in the paper structure depends on the composition and microstructure of the adhesive material used. Collagen hydrolysate, an organic compound-based additive, contributes to the formation of a fibrillar structure in cellulose and MPAN (modified polyacrylonitrile) fibers and enhances adhesion.

Collagen hydrolysates may exhibit various spatial structures. As limiting structures, either a coiled macromolecular conformation or a fibrillar (i.e., linear chain-like) conformation of the macromolecule may be present. The type of spatial structure formed by the macromolecule depends on the energy of the intra- and intermolecular bonds. If the energy of intramolecular bonds is higher, the macromolecule tends to form a coiled structure. Conversely, if the energy of intermolecular bonds dominates, a fibrillar structure is more likely to be formed [5,6].

The essence of the chemical factor lies in the formation of new hydrogen and covalent bonds. A hydrogen bond with an energy of approximately 25–35 kJ/mol is considered most characteristic for cellulose molecules. This type of bond typically occurs in compounds where hydrogen is directly bonded to oxygen or nitrogen atoms. Due to the high molecular activity of macromolecules, hydrogen bonds in high-molecular-weight compounds contribute significantly to the total bonding energy and valence forces.

Under otherwise equal conditions, hydrogen bonds are most likely to form between the most polarized atoms — that is, between the strongest proton donors and acceptors.

During the drying process of paper pulp, there is also a possibility for the formation of covalent bonds through esterification between carboxyl groups of collagen hydrolysates derived from chrome leather waste and hydroxyl groups of cellulose.

$$R\text{-}COOH+[C_6H_7O_2(OH)_3]_n\text{-}[NH\text{-}CO]_n -$$
$$\rightarrow[\ C_6H_7O_2(OCOR)_x(NH)(OH)_{3-x}]_n+XH_2O$$

In papermaking, the adhesive component serves two main functions: it strengthens the bonding between fibers and enhances the water-retention capacity of the paper pulp. It should also be noted that an increase in crystallinity and uniformity improves the physical and mechanical properties of paper. Papers containing rosin exhibit lower mechanical strength, whereas replacing it with collagen hydrolysate and textile industry waste results in an increase in the breaking length of the samples.

The degree of fibrillation of the fibers, along with their bonding characteristics, affects the structural integrity, strength, and surface uniformity of the paper. The strength of individual fibers also significantly influences the overall physical and mechanical properties of the paper.

3. Methods. There are two alternative approaches to improving the mechanical strength of paper produced from various types of fibers: (1) the use of adhesive substances, and (2) pre-treatment (modification) of MPAN-based synthetic fiber waste. The theory of fiber interaction and numerous experimental studies confirm that both the incorporation of modified synthetic fiber waste and the use of collagen hydrolysate products as adhesive components in paper composition contribute to enhancing mechanical strength.

The paper and cardboard composition's suitability for papermaking is largely determined by the surface structure of the material, the size and uniformity of fibrous raw materials, and the distribution and composition of chemical components in the cell wall. The structure of paper refers not only to the arrangement and interaction of its components, but also to the geometry of the printed material's surface [7].

Assessment of the reliability of measurement results. The strength of paper material depends not only on the morphological and physicochemical factors associated with the composition of the main components of the cell wall, but also on the fibers' absorption capacity and the strength of the mass composition itself. Strongly polarized functional groups form hydrogen bonds both among themselves and with cellulose macromolecules (see Figure 1). This, in turn, increases the strength of the paper sheet.

PS (cotton cellulose), MPAN fibers, and collagen hydrolysates contain polar functional groups — such as hydroxyl, carboxyl, amino groups, and others — in their composition. The macromolecules of collagen hydrolysates and modified PAN fibers form hydrogen and covalent bonds with cellulose. Collagen protein macromolecules also form molecular interactions with modified PAN fibers [8,9].

In composite papermaking, intermolecular bonding can influence not only the bonding within layers but also between layers. This enhances the interlayer adhesion energy. Since the strength of these chemically cross-linked bonds is significantly higher than that of hydrogen bonds, the introduction of collagen molecules into or between

cellulose structures can increase the stability of the molecular architecture, forming an interlinked mesh-like network.

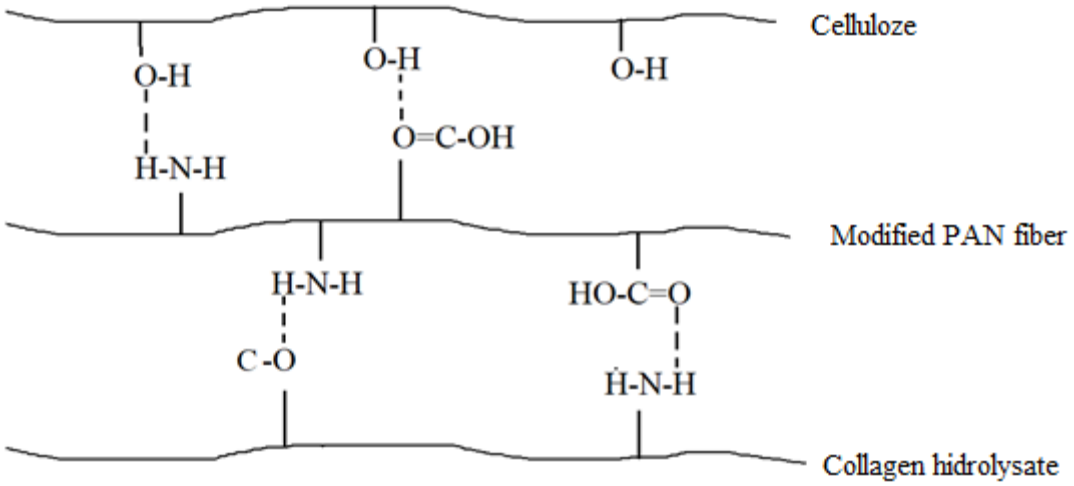

Figure 1. Interaction between Cotton Cellulose, Modified PAN Fiber, and Collagen Hydrolysate

It is important to emphasize that the optimal structure of fibrous and adhesive components within the paper pulp ensures favorable papermaking properties. As a result, a dense network is formed, promoting the generation of internal hydrogen bonds between the fibers. Along with microfibril formation, numerous fibrillar bonds develop both between and within the fibers. The adhesion mechanism of collagen hydrolysate to natural fibers is likely expressed through the formation of a dense chaotic mesh of macro- and microfibrils enriched with numerous interfiber bonds.

The introduction of textile industry waste and collagen-based adhesives into the paper composition positively influences the nanoscale structure of the interlayer surface in the paper composition. Consequently, the physical-mechanical properties and structural integrity of the paper are significantly improved.

1. Conclusions. A paper and cardboard production technology has been improved using a pulp composition consisting of 80% cotton cellulose and 20% modified PAN (MPAN) fiber waste, along with collagen hydrolysates as adhesives in various concentrations (1.5–2.5%).

2. It was established that collagen hydrolysate macromolecules and modified PAN fibers form hydrogen and covalent bonds with cellulose, and molecular interactions between collagen protein macromolecules and modified PAN fibers occur as well. It was theoretically substantiated that these intermolecular bonds can influence not only within-layer but also interlayer interactions in composite paper structures.

This study demonstrated that the use of collagen hydrolysates derived from chrome leather waste in cellulose-paper composite manufacturing, particularly for printing and

packaging enterprises, enables cross-sector integration and supports the implementation of environmentally friendly green chemistry practices.

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