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Mulberry Silk, In Its Natural Color, Employed A Range of Artificial Dyes on The Silk Thread

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Abstract:

Due to its environmentally friendly characteristics, colored mulberry silk thread has grown in popularity in recent years. Organically grown colored mulberry silk, which eliminates the use of hazardous chemicals in dyeing and processing, is the most sustainable method to satisfy the rising demand for environmentally friendly silk. Colored mulberry silk yarn has not yet been successfully created by advancements in genetic engineering or biotechnology. However, many species in the mulberry silk genus naturally produce cocoons in a variety of hues, including brown, black, mahogany red, red, khaki, pink, blue, green, dirty white, and white. In the fashion industry, silk is still one of the most desirable and opulent fabrics. The conventional method of silk manufacture entails collecting the fiber before the silkworm exits the cocoon, which ultimately kills the insect. Although silk is naturally white or yellow, the market demands a wide range of colors. Manufacturers frequently employ synthetic colors that include harmful chemicals in order to accomplish this, which has a major impact on water contamination. The pressing need for a more sustainable alternative is made clear by this environmental impact. One encouraging strategy is to change the Bombyx mori silkworm larvae's diet by giving them mulberry leaves that have been treated with synthetic dye solutions. This approach may significantly lessen the need for post-production dyeing and the accompanying environmental harm, even if not all dyes produce colored silk. Studies on the chemical properties of synthetic colors have revealed that a balance of hydrophilic and hydrophobic traits is essential for the dye to travel from the larva's digestive system into the hemolymph and eventually into the silk glands. This knowledge is essential for creating novel dye molecules that may be used to feed larvae in order to create naturally colored silk. In reality, fifth-instar silkworms are given leaves treated with synthetic colors like colomill red, colocid blue, colocid violet, colomill maroon, colocid green, and colomill yellow. The study also includes two food-grade colorants: kesar yellow and lemon yellow. Diets fortified with colomill red, colomill maroon, and colomill yellow were used to successfully make colored cocoons in three trial groups.

Keywords: Silkworm, Modified diet, synthetic dyes, colored cocoon, silk glands

Introduction:

Silk has been essential to the development of human civilizations and economies for more than 4,000 years. Traditionally utilized in important cultural occasions like weddings and festivals, it is considered the queen of fabrics **(Kang, Pl-Don et al., 2020)**. Because of its exceptional qualities—high tensile strength, brilliant sheen, comfort, soft feel, versatility to varied climates, and superb dye absorption—natural silk is a standout among fabrics. Of particular note is the fact that silk fiber can be stronger than steel of comparable thickness, which increases its worth **(Kant, Rita et al., 2011)**. The tale claims that the fiber was discovered when a silkworm cocoon fell into Xilingji's tea, causing the silk to break apart. She continued by creating sericulture, which served as the cornerstone for China's silk industry a closely kept secret for over two thousand years. The historic Silk Road later made it easier for silk to spread throughout the world. With India coming in fifth place globally, the top silk-producing nations nowadays are Italy, Brazil, India, Japan, Korea, and the former Soviet Union. Sericulture satisfies the significant global demand for natural silk while also providing a useful alternative source of income, particularly for farmers during the off-season. Colored mulberry silk thread, which is different from the more popular white kind, is frequently used to describe mulberry silk with naturally colored lint. The Indus Valley Civilization's evidence suggests that India practiced silk farming in the distant past. Naturally pigmented mulberry silk has ancient roots. According to historical records, it was used in Mexico between 3400 and 2300 BC, in Peru in 3100 BC (fibers 12 to 43 mm long), in Egypt between 2250 BC (19 to 22 mm), and at random times in China prior to 1200 AD. Drawing upon her knowledge of textile technology, Sally Fox started conducting research on naturally colored mulberry silk in 1982. She established Natural Mulberry Silk Thread Colors, Inc. , after successfully creating the first naturally colored, long-staple fiber. Her product had gained commercial success by 1988. Although the majority of mulberry silk used in commerce is made from white lint, some wild species have naturally occurring colored fibers, and certain genotypes also create pigmented silk. Before 2500 BC, colored silk with colors ranging from red to tan was utilized and produced at Huaca Prieta on Peru's northern coast, according to archaeological data. Nonetheless, some species seem to have vanished without a trace from botanical publications. Because white mulberry silk can absorb a wide variety of colors, it is used in a variety of applications and dominates the market. Conversely, the color of naturally colored silk is achieved without the use of chemical treatments. The former Soviet Union, China, Israel, India, Brazil, Peru, Greece, Turkey, Russia, and several countries in Central and South America are among the current producers of colored mulberry silk. Prior to dyeing, mulberry silk, which is white, yellow, or golden-green, must be bleached and chemically treated. These procedures are not necessary for naturally pigmented fibers. Several chemicals utilized in traditional silk processing, such as bleaching agents, chlorinated compounds, phenols, and formaldehydes, have been associated with skin conditions. Traces of heavy metals such as arsenic, lead, cadmium, cobalt, zinc, and chromium in dyes can also be irritating, especially to children. Some azo dyes have been shown to be cancer-causing. Furthermore, the dyeing procedure uses a lot of water. The two main origins of naturally colored lint in mulberry silk are genetic diversity and wild species. Crop development depends heavily on genetic resources. The National Gene Bank for Mulberry Silk in India, which is housed at the Central Institute for Research in Nagpur, maintains approximately 40 color genotypes of Morus alba, the majority of which are brown and green. These include native and exotic accessions from the United States, the former Soviet Union, Israel, Peru, Mexico, Egypt, and other places. About ten germplasm lines of Asiatic diploid mulberry silk (Morus alba and Morus nigra) display light brown lint. The fiber quality and commercial viability of most of these colored lines have been examined **(Karizova, Hana et al., 2014).** Wild mulberry species, some of which gave rise to contemporary tetraploid cultivars, are the second source of color. The most prevalent natural hue observed in these wild kinds is brown in its many shades.

Methodology:

The color of lint is determined by genetics. Prior to the boll opening, pigment begins to collect in the lint lumen. Color formation in upland cotton starts about 32 days after fertilization and takes about six days to become apparent. Pigmentation in Asian mulberry silk occurs 46–47 days following fertilization, and it takes 5–6 days for the full color to develop. But the full expression of lint color only occurs after the boll splits open and the fibers are exposed to sunlight. The lint usually takes about a week to acquire its complete natural color, but the length and strength of color development vary depending on the genotype's genetic makeup. The most prevalent natural lint colors are brown and green. Acid dyes include acidic functional groups such as -COOH and -SO3H that react with the slightly basic -NH groups found in the amide bonds of protein fibers such wool, silk, and nylon. Groups like -SO3Na or $-SO3^{-}$, which also promote ionic connections with protein fibers, affect the water solubility of these dyes. Sulfonated azo chemicals, which range from mono- to bis-azo structures, are the most prevalent acid dyes, yielding colors that range from yellow to red, violet, and brown. The navy blue bis-azo dyes have the potential to cluster and produce black hues. In general, azo dyes bind well to protein and polyamide fibers. Sulfonate groups are less common in dyes with higher molecular weight. Acid dyes based on anthraquinone, which are renowned for their high light fastness, complement azo dyes by producing colors ranging from violet to blue and green. This research uses examples such as Colomill red, Colocid blue, Colomill maroon, Colocid green, Colocid violet, and Colomill yellow. The classification of acid dyes is based on their dyeing properties, with a focus on wash fastness, migration behavior during application, and ideal pH levels. The molecular weight and degree of sulfonation of the dye have an impact on its dyeing ability **(Anto Animol, S. R. et al., 2014)**. Functional groups like SO3Na or SO3− have two functions: they act as solubilizing groups to guarantee water solubility and create ionic interactions with protein fibers like silk. Below is a description of how acid colors interact with silk:

Fibre-NH2(S) + H+ (aq) + HSO4- (aq) → Fibre-NH3+HSO4-Fibre-NH3HSO4(S) + Dye-SO3-(aq) → Fibre-NH3+DyeSO3-(S) + HSO4-(aq).

India's colored mulberry silk thread: Despite substantial progress made by Indian scientists in mulberry silk production over the past century and continuing into the present decade, the white kind has remained dominant, outperforming advancements in colored varieties. The textile industry has suffered as a result of the lack of widespread promotion of the creation and use of naturally colored mulberry silk for a variety of reasons. The fiber's length, maturity, and strength are immediately impacted by environmental variables including soil makeup, nutrient availability, sunshine exposure, and post-boll opening circumstances. Institutions such as the research center in Khandwa (Madhya Pradesh) and the University of Agricultural Sciences (UAS) in Dharwad, Karnataka, have conducted encouraging studies on colored mulberry silk, covering organic silk thread, yarn, and fabric manufacturing, as well as uses like shirting. These initiatives, which involve organic farmers and different aspects of the textile industry, such as ginning, spinning, weaving, and garment manufacturing, steer clear of synthetic colors and chemicals. This strategy has aesthetic appeal and environmental advantages, but its future depends on its financial viability and ongoing profitability. The commercial manufacturing of naturally colored mulberry silk is now facing little interest from both seed producers and the larger textile business. Increased demand, though, might pique the curiosity of biotechnology businesses. The end product is a cloth that is used for casual apparel, home textiles, and upholstery.

Silk Gland: The silk gland, which is a specialized skin gland, is formed from the labium's ectodermal tissue. These glands have a tubular, cylindrical shape, elongated structure, and branched nucleus; they are located beneath the midgut. The sericulture textbook by (**Madan Mohan Rao 1990)** states that the gland is made up of three distinct layers: the outer tunica propria, the middle layer of glandular cells, and the inner tunica intimacy, which lines the lumen. With every larval moult, a fresh tunica intima is created **(Ma, Mingbo, 2014)**. The silk gland, as **Nisal, Anuya, et al. (2014)** have demonstrated, may be split into three functional sections: the anterior, middle, and posterior.

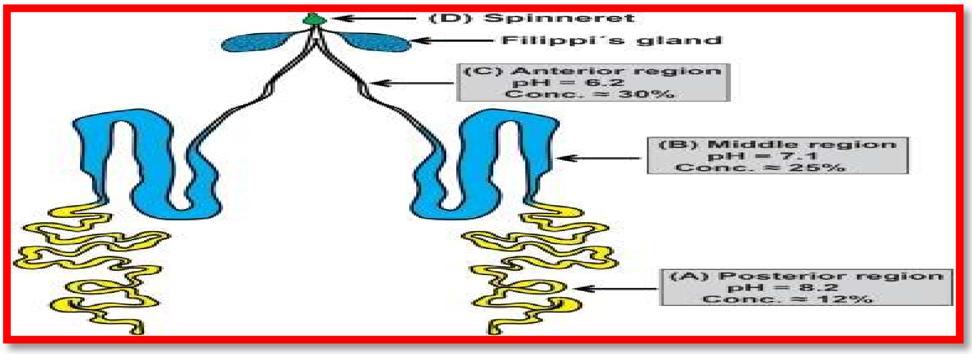

Fig: Silk Gland (Source: Science Direct.com Viewed from Google site)

The silk glands in the head come together in the front and link to the labium's spinneret. The front of these glands serves only as a conduit for silk material and produces no secretions. The gland's biggest, middle portion is bent into an S-shape and may be broken down into anterior, middle, and posterior regions. During the resting phase, fibroin develops in this core area, which functions as a storage location **(Periyasamy et al., 2012)**. Sericin is also secreted around the fibroin core; it is divided into three categories: Sericin-I, Sericin-II, and Sericin-III. The bulk of fibroin itself is produced in the posterior portion of the gland. Filipi's glands release into the silk duct at two locations close to the point where the left and right anterior sections converge; these are believed to create a waxy coating that surrounds the silk fiber. The genes that regulate the creation of silk proteins are most active during the fifth larval instar stage of development **(Shaklshimma et al. , 2013;**

Shamsheer B et al. , 2012). Silk is primarily made up of sericin and fibroin, which are produced by the posterior and central gland regions, respectively. Each pair of silk glands produces two main filaments naturally, called brin, which are then joined by sericin to create a single, continuous strand. Silk's amino acid makeup creates β-pleated sheets with side chains and inter-chain hydrogen bonds that are located above and below the bonding plane. These fibers are robust and flexible due to a high glycine concentration (about 50%), which facilitates a tight molecular packing **(Srivastava R. et al., 2014).** The fibroin, which is made in the posterior gland of Bombyx mori, is made up of three different proteins: glycoprotein, low molecular weight chain, and high molecular weight chain **(Tansil, Natalia C. et al., 2012)**.

Characteristics of Silk and Silk Protein: Unlike many synthetic fibers, silk has a soft, smooth texture that is not slick. The triangular cross-section with rounded edges, usually measuring 5 to 10 micrometers in width, gives B. mori silk its distinct characteristics. A repeating 59-amino acid sequence with only slight variations results in the bulk of the fibroin's large chain being made up of β-sheets. Light is reflected at different angles by the flat surfaces of the fibers, giving silk its inherent shine. Sericin proteins link the two primary brin filaments into a single composite fiber as they exit their respective glands (Tekmedhin T. B. , 2013). The amino acids in silk are arranged in β-pleated sheets that are held together by hydrogen bonds between chains, with side groups protruding above and below this plane. The high glycine content promotes a dense molecular structure (Trivedy Kanika et al. , 2014), which increases the fiber's strength and resilience against stretching. Silk is extremely stable and insoluble in most solvents because of its high crystallinity, numerous hydrophobic areas, and significant hydrogen bonding (Xiao Hang et al. , 2014). Despite being one of the strongest natural fibers, silk can lose as much as 20% of its strength when wet and can deteriorate when exposed to sunlight for extended periods. It adheres to surfaces due to its poor electrical conductivity. Sulfuric acid breaks down silk, whereas most mineral acids do not **(Hanafi Muhammad, Farahan et al., 2018)**.

Protein from Sericin: Sericin is divided into three fractions according to solubility: A, B, and C. The outer layer is composed of sericin A, which has amino acids and 17. 2% nitrogen but is not soluble in hot water. The components of sericin A, tryptophan, and 16. 8% nitrogen are produced by acid hydrolysis of the middle layer, sericin B. Sericin C, the innermost layer, is the closest to the fibroin. Sericin's water solubility declines as it changes from a random coil to a β-sheet conformation **(Joseph B and S. J. Ray, 2019).** It displays sol-gel behavior, easily dissolving in water at 50–60°C and then reverting to a gel when cooled.

Protein (Fibroin/Sericin): A disulfide bond connects the two identical protein subunits that make up fibroin, a glycoprotein. The crystalline and amorphous regions are both present in the fibroin filament. The amino acids alanine, glycine, and serine predominate in crystalline regions, while amorphous regions have a wider variety of them. These crystalline regions' anti-parallel β-sheet structure produces microfibrils, which combine to make bigger bundles that make up a single silk fiber. The majority of a silk strand is made up of around 75% fibroin and 22.5% sericin, with the remaining portion consisting of wax and fat (1.5%), fibroin ash (0.5%), and mineral salts (0.5%). Scouring is used to process raw silk in order to get rid of sericin and other contaminants before it is wound and made ready to be woven into cloth. Although most cocoons are white, some silkworm breeds produce naturally colored ones, like pink, yellow, brown, or green, but these varieties are rare and challenging to domesticate. Since the color is frequently lost during degumming, comprehending it necessitates determining the genes responsible for bringing pigments such as carotenoids, carotenes, and xanthophylls from mulberry leaves into the cocoon. These pigments are only found in the sericin layer that surrounds the fibroin core.

Fig: Multicolor Raw silk

Result and Discussions:

With the exception of one test group, which was able to create colored cocoons, the other test groups produced white cocoons akin to the control. The experimental group produced yellow cocoons while eating a changed diet that included Colomill Red. The weight and length of the cocoon were measured and recorded. Three groups 7, 8, and 10 created colored cocoons when they were given mulberry leaves treated with dyes. In particular, these organizations created cocoons with yellow, musky, and somewhat musky hues. The cocoons produced by Group 10, which was given the Colomill Red-enhanced diet, were yellow in color and similar to those from earlier batches. The other groups kept making white cocoons that were similar to the control. The weight and dimensions of every cocoon were documented. The green technique is the most effective way to create naturally colored silk since it is environmentally friendly and somewhat successful in allowing the silkworm's silk glands to absorb dye. The Colomill Red Yellow, Colomill Maroon Light Musk, and Colomill Yellow Musk cocoons are among the notable outcomes. The experiment was conducted once more in a second trial, this time employing a changed diet and Colomill Red, which once more produced yellow cocoons validating the reliability of the previous result. Previous issues, such as poor color retention during silk degumming, may be addressed by fine-tuning dye parameters that are closely related to silk fibroin. Higher dye concentrations also increase the intensity of the color. In modified diets, azo dyes such as acid orange, moderate black, and direct acid-fast red resulted in inherently colored silk in hues of light violet, pink, and bright orange. But, the results were different from those described by **Baburaj et al. (2014)**, who had managed to produce colored cocoons under similar circumstances, when food colorants took the place of synthetic colors in the diet mixture. Prior to feeding, mulberry leaves were treated with rhodamine B, resulting in the formation of pink cocoons without harming the silkworms. This indicates that it may be used as a dependable dye for sericulture. To make colored silk more commercially viable, more research using different environmentally friendly dyes is necessary. Notably, none of the colors employed in this experiment damaged the silkworms. Only three of the eight dyes tested over two batches—Colomill Red Yellow, Colomill Maroon Light Musk, and Colomill Yellow Musk were found to have enough penetration into the silk glands to produce visually colored cocoons **(Chung et al., 2007)**.

Conclusion:

Depending on whether acid dyes, natural materials, or particular silk lint are utilized, silk threads can be dyed in a variety of colors, creating vibrant, realistic, or multicolored effects. Mordanting to enhance color retention, heat-sensitive dyes, and occasionally resist techniques such as tying are used to produce multicolored motifs on individual threads. Using chemicals that pose environmental hazards is the conventional method for coloring silk. Using pre-dyed mulberry leaves to feed silkworms is a more environmentally friendly option since it enables the colour to be integrated during cocoon development. However, the goals of this research were only partially realized since it only examined a small number of dye hues and concentrations. Additional study employing a wider variety of dyes at higher intensities may shed more light on how to get reliable outcomes. In addition to recent technical developments brought about by increasing demand for environmentally friendly textiles, the use of natural dyes for silk is also examined. The book looks at several printing methods and strategies that are utilized on silk fabric because printing is a kind of localized dyeing. The various techniques for using many colors on silk are only mentioned briefly, and they are not covered in detail. Then, the chapter highlights current advancements in printing technology and points out important trends and driving forces that are likely to influence future study in this field.

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