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Muga (*Antheraea assama*) is the famous, widely cultivated and most prestigious natural silk available in the northeastern region of India or more specifically in Assam. This lustrous natural golden-yellow silk is obtained from a semi-domesticated, multi-voltine and sericigenous silkworm called *antheraea assamensis*, which is fed on the leaves of primary host plants, namely Som (*Machilus bombycina*) and Sualu (*Litsaea polyantha*). The confined ecological distribution of these food plants and the unique climate condition restrict the cultivation of this species exclusively to Assam. Muga is well known for some of its distinctive properties such as acid resistivity, UV resistance, and highest tensile strength among all natural silk, hydrophobicity, anti-flammable etc. All such properties have made muga silk a potential candidate to draw a great deal of research interest in the fields of textile, biomedical and bioengineering research.

The work described in this thesis concerns the surface modification of muga silk by means of various cold plasma processes such as plasma surface cleaning, plasma polymerization and plasma induced surface grafting. Limited availability of this natural silk confines its utility up to textile sector and decorative purpose only. In this present work attempt has been made to explore muga silk as advanced suture biomaterial by tailoring its surface properties using capacitively coupled radiofrequency (RF) plasma. The thesis has been organised in six chapters as described below.

In chapter 1, introductions on plasma and different methods of cold plasma generation have been given in details. Besides the fundamental aspect of cold plasma processes for surface modification of materials and their various resourceful industrial, technological and biomedical applications has also been discussed in chapter 1. This chapter also discusses muga and other natural silk products available in Assam, the unique properties of muga silk and its importance in textile and biomedical sectors. Finally the prime motives that lead to the present research work are further addressed in this chapter.

A detailed description of the experimental arrangement of the RF plasma reactor that is used in the present research work is given in chapter 2. For better understanding, the plasma discharge parameters are studied using self-compensated Langmuir and
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emissive probe. Comprehensive discussion on construction and operational procedure of these two probes are included in chapter 2. Details of the experimental technique of optical emission spectroscopy (OES) for monitoring the active plasma species generated in the plasma discharge are further incorporated in this chapter. The chemical composition and surface chemistry of the Ar plasma treated muga silk and PP-grafted muga silk are investigated using Fourier transform infrared (FT-IR) spectroscopy, Raman spectroscopy and X-ray photoelectron spectroscopy (XPS). The physicochemical properties and surface morphologies of the samples are evaluated using X-ray diffraction (XRD) patterns, tensile strength tester (UTM), dynamic contact angle etc. Scanning electron microscope (SEM) and atomic force microscope (AFM) are used to study the surface morphology of Ar plasma treated and grafted muga silk samples. The details of each of the characterization technique and the corresponding experimental conditions are also included in chapter 2.

In chapter 3, the results obtained from different characterization techniques that are used to study the physico-chemical, thermal and morphological properties of virgin and plasma treated muga silk at various discharge conditions. The properties of the Ar plasma treated fibres are studied as a function of RF power and treatment time. The hydrophobicity and tensile strength of muga silk fibre are investigated using radiofrequency (RF) Ar plasma treatment at various RF powers (10-30 W) and treatment times (5-20 minutes). The Ar plasma is characterized using self-compensated Langmuir and emissive probe. The ion energy is observed to play an important role in improving the tensile strength and hydrophobicity of the plasma treated fibres. The chemical compositions of the fibres are observed to be affected by the increase in RF power rather than treatment time. XPS study reveals that the ions that are impinging on the substrates are mainly responsible for the cleavage of peptide bond and side chain of amino acid groups at the surface of the fibres. The observed properties (tensile strength and hydrophobicity) of the treated fibres are found to be dependent on their variation in atomic concentration and functional composition at the surfaces. All the plasma treated muga fibres exhibit almost similar thermal behavior as compared to the virgin one. At RF power of 10 W and treatment time range of 5–20 minutes, the treated fibres exhibit properties similar to that of the virgin one. Higher RF power (30 W) and the increase in treatment time deteriorate the properties of the fibres due to incorporation of more surface roughness caused by sufficiently high energetic ion bombardment. The
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properties of the plasma treated fibres are attempted to correlate with the XPS analysis and their surface morphologies.

In Chapter 4, the detailed studies of Ar/propylene discharge and grafting characteristics as well as the results obtained from the various characterization techniques are included. The major role of electron temperature in ionization and dissociation of propylene molecules and subsequently in plasma polymerization process is well revealed from self-compensated emissive probe analysis. Plasma assisted grafting of polypropylene on muga silk is found to be dependent on impinging ion energy. At lower RF power (20 W) and lower grafting time (5-10 minutes) the grafting yield is found to be very low. The grafting mechanism possibly occurs through the reaction of amide II with propylene to form electron-donor-acceptor (EDA) complex. The formation of EDA complex and incorporation of more hydrocarbon groups (CC, CH, CH₂ etc.) as well as the uniform interlocking among the fibres during plasma grafting of polypropylene seem to improve the hydrophobic behavior and mechanical strength of PP-grafted muga yarns as compared to the virgin yarn. However the observed properties deteriorate with further increase in RF power (60-80 W) which is ascribed to the destruction in chemical structure of the PP-grafted muga yarns by high energetic ion impinging on the substrates.

In Chapter 5, a detailed description of the process to develop an advanced muga silk suture biomaterial is described. To prepare the suture material muga silk or Antheraea assama silk fibroin (AASF) is first sterilized using Ar plasma treatment at RF power of 20 W for 10 minutes followed by grafting of plasma polymerized propylene (PP-AASF) onto its surface using Ar/propylene plasma discharge at RF power of 40 W for 10 minutes. AASF, Ar plasma treated AASF (AASFₘₐₚₖ) and PP-AASF are subjected to various characterization techniques for better comparison and the results are attempted to correlate with their observed properties. Excellent mechanical strength, hydrophobicity, antibacterial behavior and remarkable wound healing activity of PP-AASF over AASF and AASFₘₐₚₖ make it a promising candidate for application as sterilized suture biomaterial.

Chapter 6 covers the overall outcome derived from the entire research work with future prospect of the work. It is believed that plasma treated and grafted muga silk with its enhanced physico-chemical properties will be considered as a potential material for efficient utilization in not only textile but also biomedical applications.

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