Theories of adhesion and plasma surface modification of textiles for adhesion improvement : A review

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Abstract
This paper gives the review of adhesion mechanisms and use of plasma technology in textile for improving the adhesion of coated textiles. The definition of adhesion is given as the "tendency of dissimilar particles or surfaces to cling together by physical, mechanical or chemical forces". Adhesion occurs as a result of intermolecular forces acting between the adhesive and adherend. The different mechanisms of adhesion including chemical bonding, mechanical interlocking, Adsorption and wetting, Diffusion and Weak boundary layers (WBL) have been reviewed. Methods of adhesive application on textiles and testing of the adhesive joints are also discussed.

Key words:
Adhesion theory, Coating, Peel off, Plasma, Testing

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1.0 Introduction:

Bonding by the adhesive is one of the oldest techniques in textiles. The main purpose of adhesion by adhesive bonding is a partial or complete replacement of conventional mechanical bonding techniques such as riveting, soldering, bolting etc. it has certain advantages over mechanical bonding namely better stress distribution, reduced weight, and better aesthetic. However, adhesion is not only related to adhesive joints but it is concerned everywhere when solids are brought in contact to keep them together. The definition of adhesion is given as the "tendency of dissimilar particles or surfaces to cling together by physical, mechanical or chemical forces". Adhesion occurs as a result of intermolecular forces acting between the adhesive and adherend. Adhesion and interfacial strength have a direct influence on the properties of the final product (coated fabric). The degree of adhesion is dependent on cohesive forces acting within the adhesive and interactions between adhesive and adherend. Therefore, work of adhesion Wa, that is the force required to break the bond is given as

$$W_a = \gamma_1 + \gamma_2 - \gamma_{12}$$

where γ_1 and γ_2 are the surface tension of components 1 and 2, γ_{12} is interfacial tension between 1 and 2.[1].

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The above equation suggests that adhesion is a function of the interfacial energy of material present in intermolecular interactions between the substances. Usually, the work of adhesion is directly proportional to adhesion strength. Molecular affinity is the primary requirement to achieve better adhesion. Therefore, chemical compatibility (at the molecular level) with substrata is necessary for coating. It is difficult to get good adhesion when two substances are chemically incompatible with each other. In such cases, adhesion can be improved by surface modification through physical, chemical, grafting and plasma treatment[2] .

Adhesion is often misunderstood with cohesion, there is a distinct difference between the two. Cohesion is related to the intermolecular forces acting within a substance. It is important to understand the difference between adhesion and cohesion to distinguish types of failure when bond breakage occurs[3].

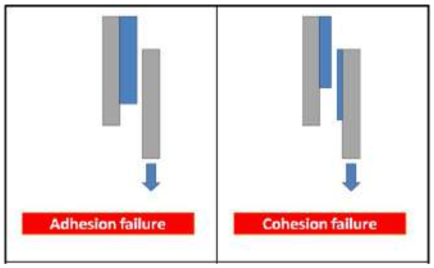

Figure 1: Types of bond failure in adhesion bonding

A bond can fail in multiple ways. Figure 1 shows the type of bond failure. Adhesive failure is at the interface between adhesive and adherend. This type of failure implies that adhesive and adherend both have greater strength than the bond between the two. Cohesive failure is within the adhesive and adhesive materials remain on both substrates. Intermolecular forces acting within the adhesive are weaker than that of the bond strength. When the substrate fails before breaking the bond and the adhesive remains intact, the failure is known as a cohesive failure of the adherend.

2.0 Mechanisms of adhesions:

There is no universal theory or mechanism of adhesion under which the reason for good bounding between adhesive and adherend can be explained. However, it is difficult to apply any single mechanism to explain adhesion because it involves several scientific fields including material science, physical chemistry and material interface, macromolecular science, rheology and mechanics and micromechanics of fracture. Following theories have been proposed based on the interactions between adhesive and adherend [4-6].

2.1 Mechanical interlocking:

It can be explained as the penetration of adhesive into the micro-cavities of the substrate. Initially, mechanical interlocking was thought as the only mechanism for adhesion to occur. To function in mechanical interlocking theory, the adhesive should flow and fill into porous, micro-void, cavities of the substrate and displace the trapped air at the interface and get solidified and lock on the substrate[4,5,7].

It is believed that increased surface roughness improves mechanical anchoring by increasing the total contact area, formation of clean surface, and highly reactive surface formation. This theory suggests that roughening of the surface is favourable for increased adhesion strength by giving peak and valley effects and increasing the contact area with adhesive. However, the substrate should be wet by adhesive otherwise increased surface area can only increase the trapped air at the interface negatively affecting the adhesion strength[5,7].

2.2 Adsorption and wetting:

Adsorption or wetting theory suggests that adhesion is the result of molecular contact between an adhesive and the adherend and the forces developed between them. The bond formed because of the adsorption of an adhesive over the surface of the adherend is generally assigned as van der Waals forces. To develop those forces the adhesive must make intimate contact with the adherend surface.

A method of creating a continuous link between an adhesive and the adherend is called "wetting". An adhesive should flow into the pores of the substrate to achieve good wetting. Poor wetting results when adhesive forms a bridge over the surface perturbations. This results in the less actual contact area between the adhesive and adherend which is the main reason for less bonding strength. Complete, spontaneous wetting to occur, either substrate should have lower surface energy or the adhesive should have lower surface tension. The surface energy of the substrate can be modified by surface modification using various techniques, one of which is plasma technology[8,9]. It is discussed in great detail in the upcoming part.

2.3 Electrostatic interactions:

This theory of adhesion is less considered a cause for adhesion strength as the contribution of electrostatic forces is negligible when compared to van der Waals forces. This theory was proposed by Deryaguin and co-workers[10] in 1948. They suggested that an electrostatic interaction (transfer of electrons) between the adhesive and substrate can occur when they use different electronic band structures to equalize the Fermi levels. Therefore, the formation of an electric double layer at the interface takes place. When peel-off tests are performed electrical discharges can be observed which supports the theory of electrostatic interactions. However, the electrical phenomena observed are the result of bond failure and not the reason for high bond strength[5,11].

2.4 Diffusion:

It works on a simple principle that if two polymers are kept in close contact above their glass transition temperature, the long chain molecules will inter-diffuse and form a strong bond by creating an interface. It is also called "autohesion" as it refers to the self-adhesion of polymers. To apply diffusion theory the polymer should fulfil two main conditions: that is both the adhesive and adherend should be chemically compatible in terms of miscibility and molecular chains of both should have mobility. Therefore, diffusion theory is less applied as in rear cases the adhesive and adherend are miscible in each other. Good examples where diffusion theory is applied are solvent or heat welding of thermoplastic materials[11,12].

2.5 Chemical bonding:

It is simply accepted that chemical bonds that form an interface of adhesive and adherend participate in the adhesion strength of both materials. Bonds that are covalent and ionic are stronger than the hydrogen and van der Waals bonds. A covalent bond has a strength in the range of 100 to 1000 kJ/mol; however, the strength of van der Waals does not exceed 50 kJ/mol. The formation of a type of chemical bond depends on the reactivity of both adhesive and adherend. For materials having polar groups such as carboxylic acid the chances of occurring covalent bonding is high as compared to that of hydrophobic materials. Covalent bonds are likely to occur in cross-linked adhesives and thermoset coating. It is the most important condition that mutually reactive groups should be present in adhesive and adherend to form covalent bonds. It is the most durable and strongest bond. Those conditions can be fulfilled by surface modification of adherend by techniques such as corona, flame and plasma[11,13,14] .

2.6 Weak boundary layers (WBL):

Weak boundary layers can arise from the adherend, adhesive, environment or by the combination of any of these three. Bond failure occurs cohesively at the weakest component of the joint. When impurities concentrate near the bonding surface and form a weak attachment with the surface then WBL occurs. In adhesion, the most important task is to remove WBL before the application of adhesive. The most common elements that form WBL are trapped air at the interface, oil on the surface of the adherend, antistatic agents etc. To avoid the formation of WBL generally, the surface of the adherend is subjected to surface cleaning, this can be achieved by plasma treatment. Plasma treatment of the material results in a cleaning effect which in terms removes the surface impurities[7,8,13].

3.0 Methods of adhesive application for coated textiles:

The main purpose of the coating is to provide specific properties to the final product which can increase its usability and application in various areas. The coating is the process in which a layer of polymeric material is directly applied over the surface of the substrate. The primary requirement is that the coating layer should adhere to the substrate. The adhesion of the polymeric material (adhesive) can be influenced by various coating methods also; besides the surface structure of the substrate. Calendaring, Flexible film laminating, Knife coating, Roller coating, Nip coating, Cast coating, Extrusion coating, Spray coating, Foam coating, UV-Cured coating and Powder Coating are the techniques used for coating depending on the type of adhesives or polymeric material[15]. Out of those methods, knife coating is commonly used in textiles.

3.1 Knife coating:

A coating chemical is applied near the knife edge and the knife spreads the coating chemical over the surface of the substrate. The thickness and the weight of the final coating are controlled by a metering knife by adjusting the pressure and distance. It is commonly used for highly viscous coating chemicals. Three types of knife coating are segregated depending on the position of the knife in the

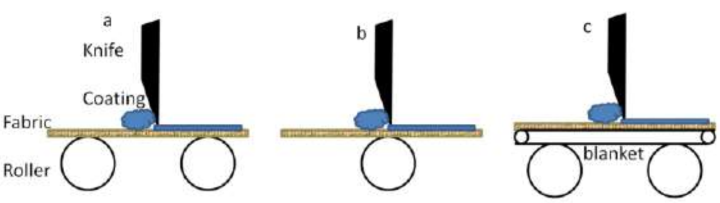

Figure 2. Knife coating, a) Floating knife, b) Knife over roller, c) Knife over the blanket

a. Floating knife- in this, the knife is positioned over the fabric. The thickness of the coating is controlled by fabric tension and the depression of the knife. It is used to apply a relatively thin coating.

b. Knife over roller- in this, the knife is suspended over the roller and does not touch the fabric. The distance between the fabric and the knife can be set to get the required thickness of the coating.

c. Knife over blanket or conveyer- this type of coating is mostly used to coat delicate fabrics.

In knife coating, penetration of the coating into the substrate and the amount of coating applied is influenced by the geometry of the knife, knife flexibility and rheology of the coating chemical.

4.0 Testing of adhesive-coated fabrics:

Testing of adhesive joints is carried out for a variety of reasons including but not limited to

1. Comparison of various properties such as tensile, shear, peel, environmental resistance, flexural, fatigue, impact, durability etc.
2. For performance and useful life prediction
3. Quality control between the batches
4. To check the effectiveness of surface treatment or surface preparation.

The major disadvantage of adhesive joints is that the interface area cannot be tested visually. It can be done in two ways by using destructive and non-destructive methods [14,16]. The list of various test methods used for the analysis of adhesive-coated fabrics are listed below. High adhesion strength is the primary requirement in all applications of coated fabric. The adhesion strength of the adhesive with the substrate can be tested using various methods. Here, only the peel test is discussed in detail as it is the most widely used for analyses of adhesive bonding strength.

4.1 Peel test:

It measures the force required to separate the adhesive layer from the adherend. This test is designed to use with flexible adhesives and it measures the highly localised stresses. The area over which the stress is applied depends on the thickness and modulus of an adhesive and the adherend. Therefore, it is difficult to evaluate exactly. Hence, it is measured as the force required per unit area for separation which is Pounds per linear inch or Newton per mm. In this type of T-peel test, all the applied load is transmitted to the bond and hence it gives the lowest peel value than any other type of peel test.[17].

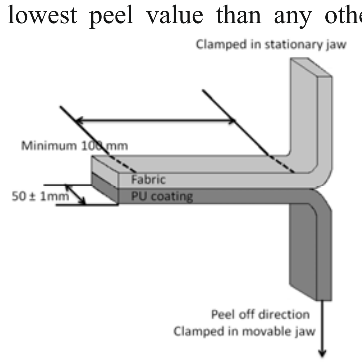

Figure 3: T-peel off test as per IS 7016-part 5

Figure 3 shows the testing of samples as per the standard test method described in IS 7016- part 5 for the determination of coating adhesion. There are other standard test methods also which include ASTM D1876-01, ASTM D903-98, ASTM-D751 etc.

- Apart from the adhesion strength of adhesive-coated fabrics other properties of the coated material are also important to measure. Those are listed below with standard test methods
- Coating mass per unit area IS 7016- part 1, BS 3424.
- Flexibility—Flat Loop Method (IS 7016 pt. 11 Based on ISO 5979-1982
- Stiffness of Fabric—Cantilever Test (IS 6490)
- Damage Due to Flexing (BS3424, IS 7016 pt. 4, ASTM D 2097
- Abrasion Resistance (ASTM D-3389, BS 3424)
- Water Vapor Permeability (ASTM E-96-80)
- Electrical Resistivity of Fabrics (AATCC 76-1995)
- Air Permeability (BS 3424)
- Tensile Strength ASTM D897-01, ASTM D5035
- Lifetime estimation and biodeterioration by thermo gravimetric analysis

5.0 Review of plasma treatments of textiles for adhesion improvement:

The term "Adhesion" is broadly used for bonding polymer-to-metal adhesion, cell adhesion in biology or the medical field, polymer composite adhesion, polymer-to-polymer adhesion etc. which practically include all types of materials and adhesives. A huge amount of research work has been done in each area therefore it is not possible to refer to all those works also not in the scope of the current review. Therefore, a review of the adhesion of textile fabrics with coating chemicals and plasma treatment is given.

Plasma interactions with the material surface result in physical and chemical modification like surface cleaning and roughness, formation of chemically reactive groups, improvement hydrophilicity etc. These modifications of the surface are mainly responsible for the improvement in adhesion bonding in the following ways

a. Cleaning of the surface removes the weak boundary lays (WBL) which are mainly responsible for lower adhesion

b. Surface roughness provides more interlocking points over the surface and improves mechanical bonding. Also, it results in an increased contact area between the fabric surface and adhesive.

c. The formation of chemically reactive groups gives rise to the formation of covalent bonds and is known to yield the strongest bond. This works on the adhesion theory of chemical bonding.

d. The adhesion theory of adsorption and wetting works well when the surface of the material is hydrophilic. The improved hydrophilicity after plasma results in better wetting and spreading of chemicals over the surface providing intimate contact between the adhesive and adherend [18,22] .

R. R. Deshmukh et al [23] have reported the effect of air plasma treatment on PET film for adhesion and printability improvement. Plasma treatment of PET was performed for 15sec to 30min and the T-peel strength of the samples was studied as a function of adhesion. An increase in adhesion strength with increased treatment time is reported. Improved adhesion after plasma exposure was attributed to surface roughness and increased surface energy. Further, it was reported that the formation of polar groups such as –CO-, -COO-, -OH is also responsible for improved adhesion. Similarly, adhesion properties of DC plasma-treated PP and PET film were studied by K. Navaneetha Pandiyaraj et al [24] who reported the enhanced adhesion with scotch tape by T-peel method and mentioned that mechanical interlocking is the key factor for improved adhesion. The same author with co-workers has also reported the adhesion improvement after DC glow, vacuum, atmospheric pressure, non- thermal plasma treatment for LDPE, PE and PP polymer films [25-30]. Improved bonding between henequen fibres and high-density polyethene (HDPE) by ethylene plasma treatment is reported. The interfacial shear strength (IFSS) was measured by a single fibre pull-out test. SEM and FTIR techniques were used to study surface morphology and chemical changes in the fibre. Increased IFSS is due to mechanical interlocking and chemical bonding is attributed to increased surface roughness and possible reaction with vinyl groups[31].

M J Shenton et. al.[32] have treated the low-density polyethylene (LDPE) and poly(ethylene terephthalate) (PET) by atmospheric and vacuum plasma using air, nitrogen and helium gases for the adhesion improvement with epoxy resin. A 180° peel test was employed to study the adhesion behaviour of the samples. It was reported that PET has better adhesion with epoxy than LDPE as PET contains oxygen in its chemical structure. Further, it was reported that atmospheric pressure plasma rapidly imparts the adhesion by a factor of two to ten; however, longer atmospheric plasma treatment does not improve the adhesion as the surface is ablated. On the contrary, LDPE and PET treated for a longer duration in vacuum plasma showed improved adhesion. Improved adhesion to LDPE for automotive applications is also reported [33] effect of helium–air atmospheric pressure plasma on adhesion between the aramid fibre and epoxy resin was studied by micro bond test. 109% increase in interfacial shear strength after the 60s of plasma exposure is reported also 16-26% increase in single fibre strength was observed[34].

Rachel M. Thurston et al [35] have treated Polyethylene and polystyrene at near-ambient temperatures using atmospheric plasma and studied the adhesion with three commercially available adhesive systems. It was reported that the improvement of adhesion is mainly due to increased surface energy and improved wetting characteristics. The effect of low and atmospheric pressure plasma on adhesion with various adhesives on almost all kinds of fibre has been reported by several researchers[35-46] .

It is well known that plasma surface modification of polymer decay with time and molecular chain re-agreement taking place with long storage time is known as ageing of the polymer. D. J. Upadhyay et al [47] have studied the plasma polymerisation of dichloromethane(CH_2Cl_2) on PP film for adhesion improvement with the commercially available adhesive tape and ageing properties were studied. They found that adhesion strength (measured by T-peel) increased with increased plasma exposure time. Further, there was a minimum change in the peel bond strength of CH_2Cl_2 plasma polymerised PP film after 2 months of ageing. N.V. Bhat et al, [48] have reported the acetone (AC) and acetone/oxygen (AC/O_2) plasma surface modification of PP film for adhesion improvement and the effect of aging on adhesion strength. They found that the surface energy of the plasma-treated film by AC and AC/O_2 has been significantly improved. Adhesion of AC-treated PP film improved but AC/O_2-treated samples showed a negative effect on adhesion initially. After aging adhesion strength moved from negative to positive for AC/O_2-treated PP film and no change in adhesion was found for AC-treated film after aging.

An interesting study was conducted on the effect of DC glow and microwave plasma treatment on adhesion of PMMA samples. Authors have found that both types of plasma treatment result in an increase in the surface energy of PMMA, however, only DC glow plasma treatment could result in improved adhesion to evaporated inorganic coatings on PMMA. Therefore, it was concluded that increased adhesion is much more determined by surface modification than only increased surface energy[49].

The adhesion of textile materials to rubber is widely studied for application in tire cords. E. L. Lawton [50] has treated the polyester multifilament 2000 denier cored with low-temperature plasma using gases such as argon, nitrogen, helium, carbon dioxide, oxygen, hydrogen, ammonia and propane. The plasma-treated PET cord was coated with resorcinol formaldehyde-latex (RFL) and adhesion strength was studied. The author found that plasma processing parameters vis type of gas, exposure time, power level, and pressure were insensitive to adhesion improvement. However, the improvement of adhesion was found with adhesive curing conditions, conditions of bonding tests and rubber composition. On the contrary, H Krump et al [51] have shown that adhesion of PET fibres with rubber matrix is positively affected by the type of plasma gas used. In another study, polyester multi-cord sewing threads were treated with barrier discharge, atmospheric pressure glow discharge and gliding arc. Adhesion properties with the rubber matrix were studied by H-test and Peel test. The best improvement was found with nitrogen gas by atmospheric pressure plasma [52]. Atmospheric pressure plasma treatment on the polyester cord and ultrahigh molecular weight polypropylene (UHMWPP) for adhesion improvement with rubber matrix have also been reported [53,54].

The effect of atmospheric air plasma treatment with respect to adhesion with silicon resin on different structures of polyester vis film, woven and non-woven fabrics have been studied and reported that the adhesion is influenced by the structure of textiles in addition to plasma treatment .

Recently, S. Palaskar et al have reported the effect of atmospheric plasma treatment on different textile structures and found the dependency of plasma processing with various GSM, Weave patterns and fabric constructions [56,57].

6.0 Summary:

Adhesion is often misunderstood with cohesion, the difference between the two is discussed in detail. A total of six theories of adhesion are reviewed. Types of coating techniques, testing of coated materials and application of plasma technology for adhesion improvement are reviewed. Plasma interactions with the material surface result in physical and chemical modification like surface cleaning and roughness, formation of chemically reactive groups, improvement hydrophilicity etc. These modifications of the surface are mainly responsible for the improvement in adhesion bonding and different mechanisms of adhesion work together.

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