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INVESTIGATION OF THE EFFECT OF ZINC OXIDE-MODIFIED GUM ARABIC ON POLAR SUBSTRATES

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ABSRACT

Gum Arabic solution, a water-based adhesive, was modified with zinc oxide filler and the formulation was applied on wood, ceramic, glass and textile substrates. A strip of paper was used as a common adherent to all the substrates. Zinc oxide increased the viscosity of 30wt% gum Arabic solution and increased bond strength on all the substrates except on glass where low bond strength was observed. The low bond strength on glass was perhaps due to localized stresses in adhesive film. Wood, ceramic and glass had cohesive failure on the paper adherent while adhesive failure on the paper adherent was observed on the textile substrate.

KEYWORDS: Gum Arabic, Zinc oxide, Substrate, Bond, Strength.

INTRODUCTION

Gum Arabic, also known as acacia Senegal, is a dried exudates from the species of the acacia tree found in various tropical and semi-tropical regions of the world. The gum consists of a mixture of calcium, magnesium and potassium salts of Arabic acid which is a complex branched polysaccharide that contains galactose, glycuronic acid, arabinose and rhamnose residues. It also contains some glycoproteins and has a broad molecular weight of 260,000 to 1,160,000 (Anderson, 1991).

Primary or secondary chemical bonds are known to exist between an adhesive and an adherent. Primary bonds include electrovalent, covalent and metallic bonds. By far the most important of the adhesive bonds are the secondary or Van der Waals bonds that give rise to attraction between molecules. Most significant of these are the London or Dispersion forces which are responsible for virtually all the molar cohesion of non-polar polymers such as polyethylene, natural rubber and butyl rubber (Skeistand and Miran, 1990). In polar compounds, interaction of permanent dipoles results in strong bonds especially from hydrogen bonding. This explains why such diverse adhesives as starch and dextrin, polyvinyl alcohol, cellulose nitrate, phenolics and epoxies form excellent bonds with polar substrates or adherents such as wood, paper, leather, glass and metals, which have features for hydrogen bonding.

The labelling, sealing and carton-closure market, once served mainly by water-remoistening gummed tapes and labels, is shifting towards pressure sensitive adhesives, hot-melt sealable and delayed-action heat sealable adhesives due to the new water-resistant plastic packaging materials in use today. However, water-based adhesives made from natural polymers such as gum Arabic, starch and dextrin are still used to produce labels for paper, glass, textiles and unfinished wood where quality performance is provided at very low cost. Water-based adhesives are also made from emulsions or dispersions of polyvinyl acetate, poly acrylate or natural rubber as well as from other synthetic water-soluble polymers such as polyvinyl pyrrolidone, polyethylene oxide or polyvinyl alcohol (National Adhesives, 2006). Recent developments in water-based adhesives for dry bond laminating are providing compelling options for packaging converters to choose between traditional solvent-based adhesive systems to economical and environmentally friendly water-based alternatives (Rohm and Haas, 2004).

Gum Arabic, a highly branched polymer, forms weak and brittle adhesive films. The aim of this work is to improve the adhesive strength of gum Arabic by incorporating zinc oxide filler and to determine the effect of the modified gum Arabic on polar substrates.

MATERIALS AND METHODS

MATERIALS

Gum Arabic was obtained from Borno state and Zaria market. Wood (hardwood and plywood), ceramic tiles and textile (cotton material), were obtained locally in Zaria. White duplicating (A3) sheets were used as paper substrate while standard microscopic slides were used as glass substrate. Zinc oxide is of M&B while phenol is from BDH.

Sample Preparation

The hardwood substrate was planed while the plywood was used as supplied. All substrate surfaces with the exception of paper were cleaned by wiping them with a damp cloth to remove dust, oil, grease or other contaminants. The substrates were then allowed to dry in a convection oven for 5min.

Preparation of Gum Arabic Stock Solution

200cm^3 of water was added to 90.0g of raw gum Arabic from which wood or other extraneous particles had been removed as much as possible, by hand, before weighing. The mixture was vigorously stirred for one hour followed by the addition of 100cm^3 of water. Vigorous stirring of the solution was continued until all

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gum had dissolved to give a thirty weight percent (wt %) gum Arabic solution. The gum solution was filtered through two metal sieves and its pH was determined with Hydrion paper.

5cm^3 of the stock solution was introduced into several small plastic bottles each fitted with a screw cap. Various amounts: 0.075g, 0.15g, 0.225g, 0.30g and 0.375g of zinc oxide were added to the portions of gum Arabic solution in the plastic bottles to give 5, 10, 15, 20 and 25 wt% of zinc oxide respectively. No additive was added to the control sample. Each mixture was vigorously stirred followed by the addition of two drops of phenol as preservative. The viscosities of the gum formulations were measured on a Brookfield Viscometer at 29^{0}C.

Bond Strength Determination

The strength of paper – substrate bond was estimated by determining the Pull Force (PF), on a specially constructed wooden pull apparatus illustrated in Fig. 1, which is a local adaptation of the 180^0 Test equipment for peel or stripping strength of adhesive joints described in ASTM method D 0903. The substrates were attached and held on the centre – support of the wooden pull apparatus while various masses were incrementally added on the scale pan until paper – substrate failure was observed. Bond strength was then calculated thus:

$$\frac{\text{Mass (g) (at bond failure)}}{1000} \times 9.80\text{N (Force of gravity)}$$

30wt% gum Arabic solution was used as standard.

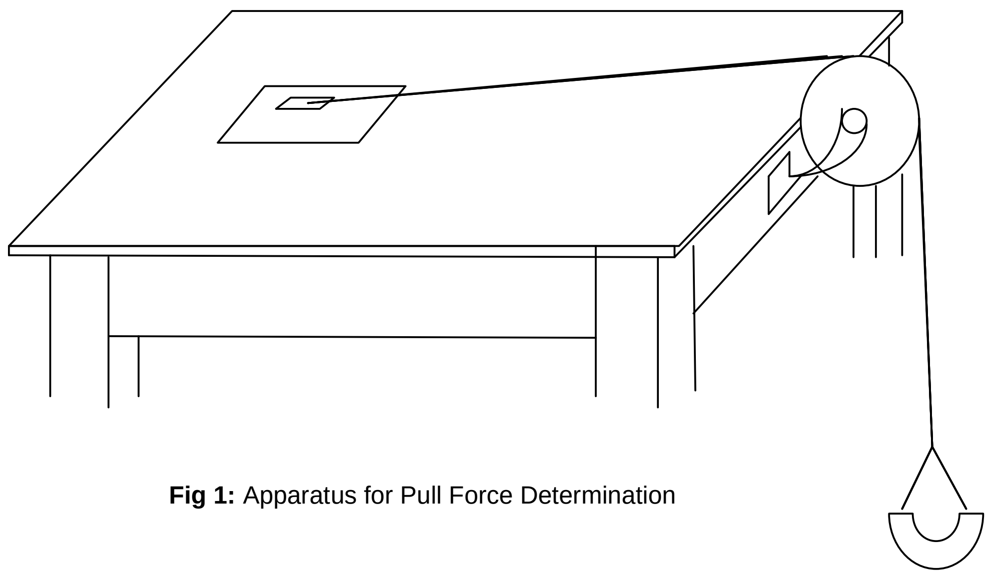

Fig 1: Apparatus for Pull Force Determination

Determination of Bonding Time

The various formulations were applied uniformly on wood, ceramic, glass and textile substrates while a paper strip (11 x 55mm) was used as a common adherent to the substrates. The samples were allowed to dry at ambient temperature (20 – 30^0 C) for 36hr. Within the drying period, samples were withdrawn at 6, 12, 24 and 36hr intervals and the Pull Force was used to determine the time at which maximum bond strength was attained.

RESULTS AND DISCUSSION

The bond strength of the various gum Arabic formulations determined for wood, ceramic, glass and textile is shown in Table 1. The standard 30wt% gum Arabic solution applied on wood, glass and textile attained maximum bond strength in 12hr while that applied on ceramic attained maximum bond strength in 24hr. The formulations containing zinc oxide attained maximum bond strength in 24hr on wood, ceramic and textile but 12hr on glass (Table 1). The formulation containing 5wt% zinc oxide also attained maximum bond strength in 12hr. Zinc oxide increased the viscosity of gum Arabic solution and viscosity increased with zinc oxide concentration as indicated in Table 2.

Zinc oxide increased Pull Force (PF) at all concentrations on wood and the highest PF of 2.85N was obtained at 20wt% zinc oxide as shown in Fig. 2. In adhesive formulations, zinc oxide applied as filler, increases viscosity and controls penetration associated with porous substrates (Jarowenko, 1977). Wood is porous and consists of cellulose, hemicelluloses and lignin which are all highly complex polar compounds. Zinc oxide might have increased the bond strength on wood as a result of dipole-dipole interactions formed between gum Arabic, zinc oxide and the polar groups on wood. According to Scheikl and Dunky (1998), the penetration behaviours of liquids into wood surface depend on the different diameters of wood cells and viscosity and molecular size of the penetrating liquids. Hence, zinc oxide increased the viscosity of gum Arabic solution which in turn increased physical adsorption and enhanced Van der Waals interactions between the highly branched complex gum Arabic molecules, zinc oxide and polar groups on the wood, at the interface which subsequently increased bond strength on wood.

Furthermore, intrinsic adhesion might have been enhanced on wood by mechanical interlocking of gum Arabic solution with pores on the wood. Hardly surprising as Bogner (1993), had reported correlations between bond strength and increased penetration of adhesive into wood. Wood had a cohesive failure in the paper adherent.

In the formulations applied on ceramic, PF was increased at all zinc oxide concentrations, while 15wt% zinc oxide had the highest PF of 3.20N as shown in Fig. 2. Traditional ceramic consists mainly of silica (SiO_2) and alumina-silicates such as clay ($3Al_2.2SiO_2$), both of which are highly polar compounds. Zinc oxide-modified gum Arabic improved bond strength on ceramic due to dipole-dipole interactions as well as electrovalent bonds formed between gum Arabic, zinc oxide and polar groups on ceramic. In an investigation involving albumin adhesion on ceramics, Krajewski (1998), concluded that the chemical nature of the substance constituting the ceramic was mainly responsible for adhesion. As stated by Davis (1985), the chemical properties of an adherent surface govern the interactions of adherent with primer or adhesive as well as long-term resistance of the bond to environmental degradation. Furthermore, Venables (1979) concluded that the morphology of the adherent controls the degree of physical interlocking with primer or adhesive such that a microscopically rough surface can provide a strong and more durable bond than a smooth surface. Intrinsic adhesion was therefore enhanced on the porous ceramic by mechanical interlocking of gum Arabic molecules with asperities on ceramic surface. Correlations between ceramic porosity and adhesion have been published by Packham and Johnston (1994). The ceramic-paper bond had a cohesive failure in the paper adherent.

Zinc oxide in the formulation gave low bond strength on glass as seen in Fig. 3. According to Bikerman (1968), surface modifications or molecular reorientation in adherents could give rise to weak boundary layer at the interface which could lead to bond failure at the interface. It can therefore, be deduced that the repulsive intermolecular forces resulting from weak boundary layer at the interface between zinc oxide-modified gum Arabic and sodium silicates contained in glass, decreased the bond strength on glass. A good adhesion promoter that has an excellent adhesion to glass and other silica-based substrates has been patented (Rodriques and Long, 2007). Furthermore, Aculon (2011) has developed specialized nanotechnology coating and treatment that increase adhesion on glass. These promoters eliminate the need for more costly pre-treatment or surface roughening that is often needed to promote adhesion of various substances to glass surface. The low bond strength on glass might also be due to bond failure resulting from localized stresses in adhesive film. Glass had a cohesive failure in the paper adherent.

Increasing zinc oxide concentration uniformly increased PF on textile and the highest PF of 2.80N was obtained at 25wt% zinc oxide as shown in Fig. 3. In order to improve the properties of fibres in many applications, surface treatments have been used to modify the chemical and physical structures of the surface layers of textile fibres. Some recent developments in surface modification of textile fibres have been reviewed by Lou (2002). Cotton textile is cellulose which consists of a linear chain of glucopyranose residues with many polar hydroxyl groups in the chain. The dipole-dipole interactions between gum Arabic, zinc oxide and polar groups on textile increased bond strength on textile. Furthermore, intermolecular interactions between the adherents were enhanced as zinc oxide concentration was increased (Fig. 3). According to Holme (1999), satisfactory adhesion to fibres, yarns and fabrics depends on the nature of the fibre surface, the presence of natural or added impurities and the effects of physical and chemical treatments. Such chemical treatments may modify the fibre surface energy and the wetting and spreading processes such that obtaining satisfactory adhesion to textile materials becomes difficult. All textile substrates had an apparently clean surface after peeling which implied an adhesive failure at the interface.

Table 1: Bond strength of Gum Arabic Formulations on Wood and Ceramic Substrates

Substrate	Composition Wt%	**Pull Force (N) at**			
		6hr	12hr	24hr	36hr
Wood	30GA	1.80	2.50	2.40	2.45
	5ZnO	1.60	2.40	2.80	2.80
	10ZnO	1.50	2.20	2.55	2.45
	15ZnO	1.55	2.40	2.60	2.60
	20ZnO	1.50	2.30	2.85	2.60
	25ZnO	1.60	2.35	2.60	2.55
Ceramic	30GA	1.30	2.00	2.45	2.45
	5ZnO	2.05	2.80	2.55	2.45
	10ZnO	1.95	2.85	3.15	3.00
	15ZnO	1.80	2.75	3.20	3.10
	20ZnO	2.00	2.85	2.90	2.85
	25ZnO	1.90	2.70	2.75	2.70

Glass	30GA	1.85	2.60	2.50	2.55
	5ZnO	1.30	1.75	1.50	1.55
	10ZnO	1.40	1.85	1.55	1.60
	15ZnO	1.40	2.00	1.80	1.70
	20ZnO	1.50	2.00	1.65	1.75
	25ZnO	1.50	2.05	1.80	1.75
Textile	30GA	1.50	2.15	1.95	2.00
	5ZnO	1.55	2.25	2.40	2.40
	10ZnO	1.55	2.30	2.50	2.40
	15ZnO	1.80	2.55	2.60	2.60
	20ZnO	1.70	2.50	2.75	2.70
	25ZnO	1.80	2.60	2.80	2.70

GA = gum Arabic, ZnO = zinc oxide

Table 2 Viscosities of modified gum Arabic solutions

Composition, Wt%	Viscosity, cP
30GA	1350
5ZnO	1400
10ZnO	1525
15ZnO	1620
20ZnO	1775
25ZnO	1905

GA = gum Arabic, ZnO = zinc oxide

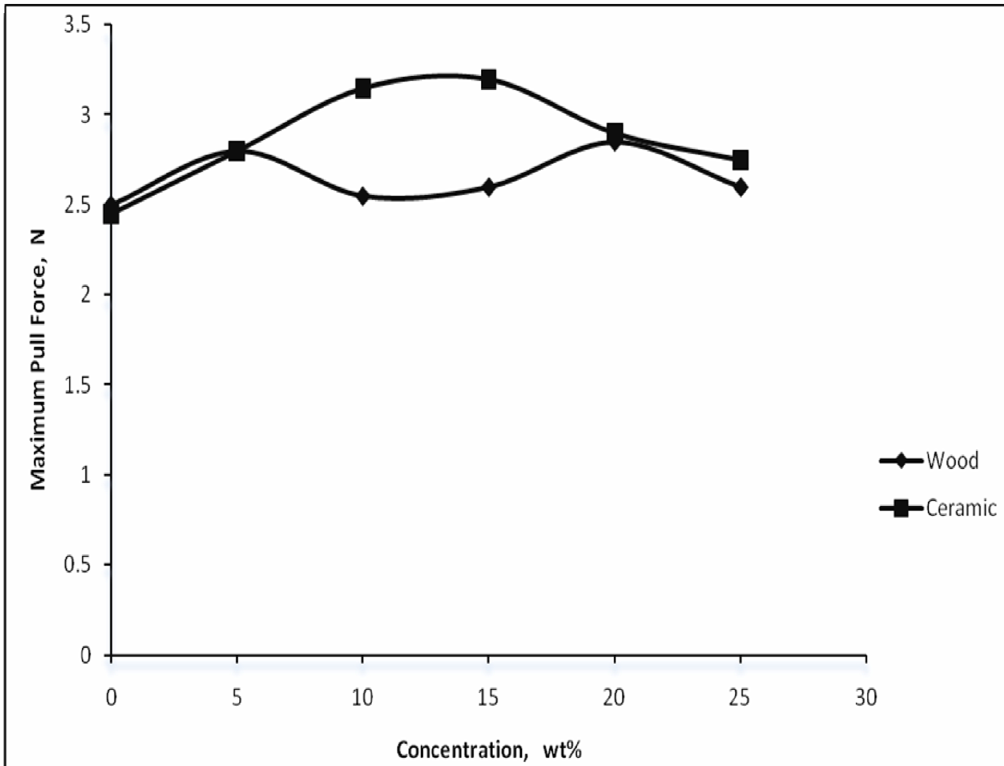

Fig. 2 Maximum Pull Force vs concentrations of zinc oxide in 30wt% gum arabic solution, on wood and ceramic

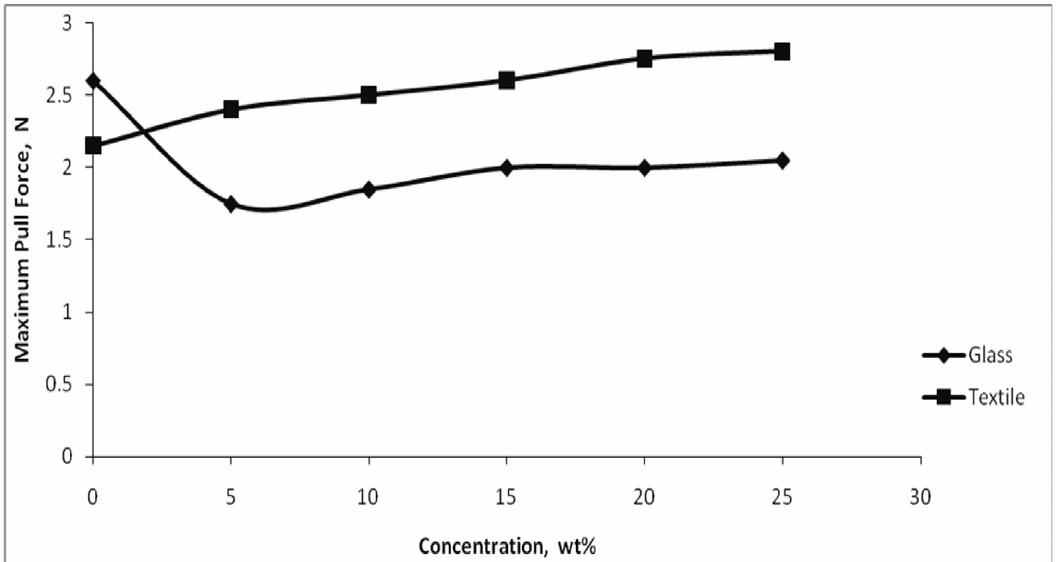

Fig. 3 Maximum Pull Force vs concentrations of zinc oxide in 30wt% gum arabic solution, on glass and textile.

CONCLUSION

Zinc oxide increased the bond strength of gum Arabic on wood, ceramic and textile and thus the formulation can be used for enhanced paper label on the substrates. The low bond strength obtained in glass was perhaps due to bond failure arising from localized stresses in adhesive film. Owing to the apparently clean surface obtained after peeling from textile, zinc oxide-modified gum Arabic solution can therefore be used as cheaper substitute for pressure-sensitive adhesive label for textile.

REFERENCES

Anderson, D. M. W., Millar, J. R. A. and Wang, W. 1991. Gum Arabic (Acacia Senegal): Unambiguous identification by ^{13}C-NMR spectrography as an adjunct to the Revised JECFA Specification and the application of ^{13}C-NMR spectra for regulatory purposes. Food Add. Contam. 8, 405-421.

Aculon Incorporated, 2011. _www.aculon.com/Adhesive-promotions. Retrieved 12th April, 2011.

ASTM D 0903 Test Method for Peel or Stripping Strength of Adhesive Bonds.

Bikerman, J. J., 1968. The Science of Adhesive Joints, 2nd Ed. Academic Press New York.

Bogner, A., 1993. Wettability of wood and adhesion. Drvna industrija 44, 139-143.

Davis, G. D., Ahern, J. S., Matienzo, L. J and Venables, J. D., 1985. Use of Hydration Inhibitors to Improve Bond Durability of Aluminium Adhesive Joints. J. Mater. Sci. 20, 88-98.

Holme, I. 1999. Adhesion to textile fibres and fabrics. Int. J. of Adhesion adhesives. 19, 455-465.

Jarowenko, W. 1977. Handbook of Adhesives 2nd Ed. I. Skiest ed. Van Nostrand Reinhold New York. p192.

Krajewski, A., Piancastelli, A. and Malavolti, R., 1998. Albumin adhesion on ceramics and correlation with their Z-potential. Biomaterials 19, 637-41.

Lou, S., Van, O. And Wim, J. 2002. Surface Modification of textile fibres for improvement of adhesion to polymeric matrices: A review. J. Adhesion Sci. Technol. 16, 1715-1735.

National Adhesives, 2006. American National Starch and Chemical Company U.S.A.

Packham D. E. and Johnston, C. 1994. Mechanical adhesion: were McBain and Hopkins right? Int. J. Adhesion Adhesives 14, 131-135.

Rodriques, K. A. and Long, J. C. 2007. Adhesion Promoter. US Patent 7270853.

Rohm and Haas, 2004. Water-based Adhesives: Viable Alternative to Solvent-Based Adhesives. www.azom.com/Details.asp? Retrieved 24th January 2008.

Scheikl, M. and Dunky, M. 1998. Measurement of Dynamic and Static Contact Angles on Wood for the determination of its surface Tension and Penetration of liquids into wood surface. Holzforschung 52, 89-94.

Skeistand, I. and Miran, J. 1990. in I. Skeist ed, "Handbook of Adhesives" 3rd Ed. Van Nostrand Reinhold New York pp 3-20.

Venables, J. D., McNamara, D. K., Chen, J. M., Sun, T. S. and Hopping, R. L. 1979. Oxide morphologies on aluminium prepared for adhesive bonding. Appl. Surf. Sci. 3, 88-98.