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Improved Adhesion of Butyl Mastic Using Electron Beam Irradiation

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Abstract
Butyl mastic is used in a number of industrial areas and in particular in the production of adhesives. The performance of this material can be improved by using ionizing radiation. In this work we showed that the shear forces in butyl mastic were modified when the material was exposed to radiation, and as a consequence the adhesion strength was increased. This was done by comparing the shear forces of butyl mastic before and after the exposure to ionizing radiation. The result was an increase of 440% of the average shear strength between the sealant and the substrate after the irradiation. This work can be considered as an important contribution to the field of butyl mastic. sealants and adhesives.

Keywords
Butyl mastic, ionizing radiation, shear strength

1. Introduction

The use of adhesives dates back to antiquity. In the past adhesives were restricted primarily to certain natural polymers as a way to 'glue' related materials. Animal based-glues, asphalt, resins, and resins from trees were used well before the introduction of modern adhesives which are based on starch, natural latex, or synthetic polymers.

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The development of synthetic adhesives, which can be tailor made for particular applications, has led to significant improvements. Nowadays, several factors must be considered when choosing a particular type of adhesive:

- the adhesive must wet the surface of the substrate;
- if the surface is waterproof, and non-absorbent, the adhesive should be free of water or organic solvent;
- both adhesives and application methods should be of low cost;
- the patches should not be stiffer than the substrate to avoid stress concentrations;
- after hardening, the joints should be able to withstand normal environmental conditions.

Mastic sealants have been known for a long time. The first formulations were developed in the 1960s to supply sealants for automotive windshields, and later in the 1970s to supply sealants to the construction business. Nowadays there is a relatively high demand in the building industry for the use of aluminum structural parts and the joining and sealing of these parts make adhesives a very important economical item, especially as a result of their low cost. Adhesives can be applied to a large range of materials for sealing or caulking purposes as, hence, are of paramount importance in the building industry. The major advantages of modern adhesives are their large versatility and range of application.

The evaporation of a solvent is part of the curing process of butyl mastic, with the solvent acting as a vehicle to facilitate the application. This makes it a very practical product which is simple to apply.

2. Butyl Mastic

There are two methods of formulating a butyl rubber based sealant or butyl sealant. This sealant constitutes the main component of a rubber mastic system [1]. The first method uses solid butyl rubber with a high molecular mass which is mixed with solvents and plasticizing oils. The second method involves mixing the sealant with liquid polybutene. With this method the stickiness is the predominant feature, but it does allow for the mineral filler content to be increased, thus, reducing the overall cost. The product has its own curing process which involves the evaporation of the solvent. This feature is essential in some cases such as in the manufacture of certain gasket materials. It has been applied, for example, to join steel structures, aluminum parts, truck body parts, where there are small mechanical displacements. For larger displacements the adhesive could reach fatigue and compromise not only the bonding but also the seal or caulk. The mechanical handling and the resistance against water infiltration are some of the special characteristics of mastic.

The main application of mastic is in the joining of aluminum structural parts used in the construction industry. When the final cost is an important issue, the formulations must be evaluated [2]. During the 1970s there were formulations with up to 56.5% of mineral fillers, with a guaranteed shelf life of 1 year. Today with the advancement of new materials, the filler content can reach up to 79%. Furthermore as the product is 'undried', its shelf life can be as high as two years.

These sealants are known by different names: butyl or butyl based sealant, butyl mastic, mastic or simply, butyl mastic. In the market, they are known to have a basic formula consisting of butyl rubber polymer which constitutes a fraction, ranging from 1 to 3%, of the overall sealant. The remainder of the formulation is basically 25–30% mineral or natural oil, 60–79% mineral fillers, 0.05–3% pigment, 7–18% solvents and up to 2% of other additives. The amount of the secondary polymer, which is polybutene or polyisobutylene, varies from one manufacturer to another, and ranges from 1 to 2%. In summary, the mastic is an 'undried' product, with variable formulations containing a large amount of oil, and has no cross-linking agent [3]. It is the latter which makes the mastic 'undried' and crosslinking agents such as sulfur, zinc oxide, magnesium oxide are not included in mastic formulations. For this reason the butyl mastic has an appearance of soft rubber, like a gum. Some types of mastic contain additives which allow it to form a skin or 'shell' to protect it from ultraviolet rays. As an example, a product well known in Brazilian as 'Juntabel', has been extensively used in the construction industry since the 1980s. Other researchers [4] have defined mastic as a material containing drying oils or components of bitumen, which in time also form a skin on the surface. These products attached to various substrates, aim to fill corners or junctions with their special blends.

This paper aims to demonstrate the behavior of adhesion through the evaluation of shear strength acting on the surface of materials, as has been done for epoxies, with a consideration for the kinetic and curing procedures [5].

3. Experimental Procedure

Samples of mastic (Brascoved 1513/CV) were used to bond 2 mm thick aluminum plates (7075-T651 Alumi Copper, hardness 160 Hb). Each specimen was prepared as a layer of mastic between two aluminum plates according to ASTM D 1002 [6].

Tests were performed over a period of 28 days on the non-irradiated specimens. Another set of specimens were cured for 28 days, then irradiated, and tested after 48 h. In this study we used an electron accelerator model DC1500/25/4 JOB188 (Dynamitron®), with an energy of 0.5–1.5 MeV, a beam current to 25 mA, scanning from 60 to 120 cm, with a maximum beam power of 37.5 kW. Although the dose rate could range from 1.07 to 161.67 kGy/s, the specimens were irradiated with radiation doses of only 5, 10, 20 and 30 kGy at 22.61 kGy/s, because applications above 30 kGy would not be economically viable.

The specimens were irradiated in a direction perpendicular to the aluminum surface. The electron energy range was selected to allow the maximum penetration of the electron beam through the aluminum plates because in this region the stopping power of aluminum was small (1.5 MeV cm^2/g) and the electrons reach the mastic layer in the specimens. The irradiation was performed in air.

In order to evaluate the behavior of the adhesive forces on irradiated and on the non-irradiated samples, shear tests were performed using an Instron universal testing machine, model 3367, with a 30 kN load cell (courtesy of Brascola company). In the following discussion the two specimen sets were defined as ‘mastic without radiation’ and ‘mastic with radiation’.

Shear strength tests were performed on the ‘mastic with irradiation’ samples after 28 days plus 48 h. In the case, of the ‘mastic without radiation’ samples the tests were done after 7, 14 and 28 days in order to get a better understanding of the solvent evaporation curing process.

4. Results and Discussion

Figure 1 shows the shear strength for samples of ‘mastic without radiation’ for the three time periods. Ten samples were tested for each period of time. The effect of the curing time on the adhesive forces was noted. The average strength value after 28 days was approximately 0.39 kgf/cm^2; suggesting that the forces on the surface of the material without ionizing radiation, were very weak.

Figure 2 shows the shear strength for samples of ‘mastic with radiation’ after 28 days plus 48 h. Ten samples were tested and the sample doses ranged from 5 to 30 kGy. From Fig. 2 it can be seen that there was an improvement in the

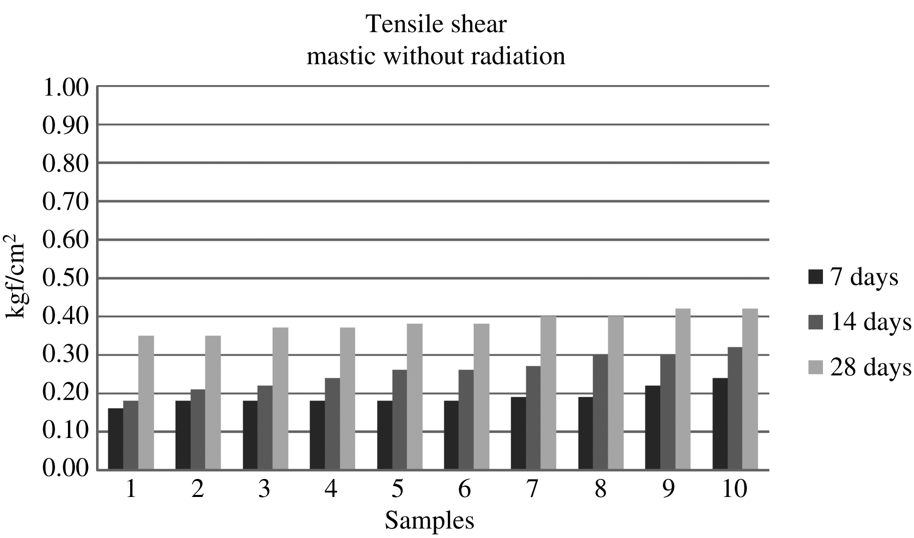

Figure 1. Shear strength of the butyl mastic without radiation.

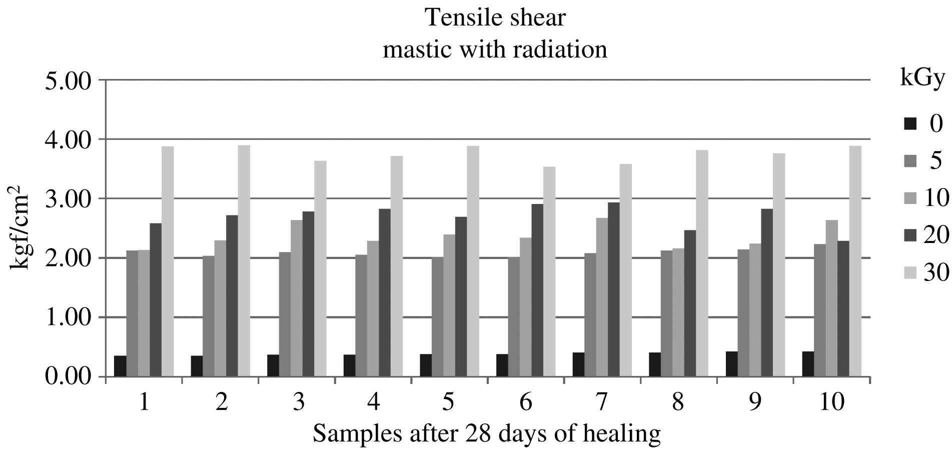

Figure 2. Shear strength of the butyl mastic with radiation.

shear strength as the irradiation dose increased. The adhesive forces increased significant as a result of the increase of the shear force and reached an average of 3.75 kgf/cm^2 after 30 kGy; this is almost 10 times the value for mastic without irradiation (0.39 kgf/cm^2). This result suggests that the forces on the surface of the material were improved after the irradiation with electrons; this we believe, is an important issue in the field of adhesives. This effect was related to the number of dative bonds of butyl which is larger then the London bonds. According to some authors [7–9] adhesion can be defined as the state in which two surfaces are held together by interfacial forces such as chemical or mechanical forces, or both. On the other hand, an adhesive material is defined as a substance capable of holding together different substrates by linking both surfaces. The shear tests were carried out to verify the modifications to the bonding shear strength of both surfaces attached to the aluminum plates. This work is particularly relevant because butyl mastic is widely used in buildings and in automotive applications. The experiments were limited to 28 days and the results from this work cannot predict any change in behavior after this period.

5. Conclusions

Butyl mastic can be used as a caulking material or sealant and is usually applied and cured at the room temperature. Butyl mastic is a product based on butyl rubber, or related types of rubber, and oils plasticizers, pigments and minerals. The main feature of this type of sealant, as used for sealing and caulking, is the curing by evaporation of solvent. This mastic, when exposed to ionizing radiation, behaves like an adhesive, with the ionizing radiation assisting the curing process by improving the bonding (adhesion). An improvement of 10 fold was achieved in the average shear strength between the sealant and the substrate after irradiation with electrons. The present work also shows that the butyl mastic can be used in environments where radiation is present such as in sealing windows of accelerators, etc. This can even lead to an improvement of the properties.

This work highlights the use of ionizing radiation in improving properties of materials and in the development of butyl sealants as adhesive sealants.

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