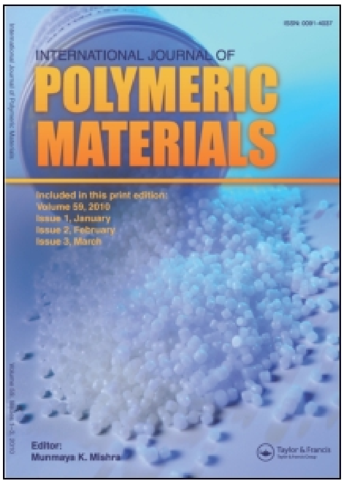

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Use of Epoxy Resin as an Adhesion Promoter in Liquid Polysulphide Sealants[†]

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INTRODUCTION

Curable liquid polysulphide polymer based sealants and caulking compositions have become popular in recent years. They are based on liquid polysulphide polymers and are being used especially in the building, automotive and marine industries, for castings e.g. solid rocket propellants, for leather and textile impregnating agents, for adhesives, coatings etc. The distinctive nature of these liquid sealants is their capacity for vulcanizing at room temperature with an addition of suitable curing agents with transition from the viscous-flow, pasty state to the rubbery and elastic stage, practically without any shrinkage.

Because of the wide range of applications in which these liquid polysulphide polymer based sealing and caulking compositions are used, they must be capable of bonding to various substrates e.g. metals, glass, concrete, wood etc. In the unfilled state, vulcanizates of liquid polysulphide have poor strength and adhesion to various

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substrates. The addition of appropriate fillers and adhesion promoters to the sealant increase the tensile strength and resistance to peeling from substrate substantially. Two basic methods have been recommended in the literature for obtaining adhesion, namely, the use of primers or the use of certain adhesion promoters. In specific purposes, the combination of these two methods have been used. A number of adhesion promoters have been evaluated in liquid polysulphide sealants but none of these have been found universally satisfactory. A small amount of certain phenolic resins in combination with liquid polysulphide sealant give adhesion to a variety of surfaces but show failure to adhesion after coming in contact with polar solvents such as water. In other words, phenolic resins provide only a polar, rather than a chemical type bonding of the cured sealant to the substrate. The polar solvent breaks down the polar bond thereby eliminating the use of sealants containing phenolic resins. A particular grade of polyvinyl acetate in combination with special phenolic resins, show good adhesion to a variety of surfaces, both wet and dry.[1] Some organo-silane compounds[2,3] have been proposed to overcome the difficulty encountered by using phenolic resins. Though they are reported to be good adhesion promoters they have some disadvantages also. These adhesion promoters are economically costly, affect the shelf life of the sealant making it more shorter and exhibit erratic adhesion results with different substrates.

Recently[4] abietate terminated polysulphide polymers, epoxy terminated polysulphide polymers and abietate terminated polyester polymers have been reported as adhesion promoters for polysulphide sealants.

Vulcanizates of liquid polysulphide polymers may be used for working in air and in liquid fuels of petroleum origin at temperature from −60°C to +150°C. Keeping this view in mind, the present work of studying the suitability of commercial epoxy resin as an adhesion promoter in a polysulphide sealant has been undertaken.

SEALANT COMPOUNDING AND CURING

The raw material except a liquid polysulphide polymer were procured from commercial organisations. A liquid polysulphide polymer (mol. wt. 2500–3500) was prepared in our laboratory. The final sealant has been prepared as a three component system i.e. (1) polymer paste (2) curing paste and (3) accelerator. The preparation of polymer paste and curing paste is very much similar to the methods adopted in the paint industry. The polymer paste consists of (1) liquid polysulphide polymer—80 parts by weight, (2) carbon black GPF—20 parts by weight, (3) Epoxy resin (epoxy equivalent 260)—4 parts by weight and (4) Thixotropic agent (silicon dioxide)—variable. Initially the mixing was carried out in a pug mixer and finally on the three roll paint mill till a creamy consistency was obtained. The particle size of the filler used is extremely important since it affects the end properties as well as the thixotropy of the system. The curing paste consists of (1) manganese dioxide—50 parts by weight and (2) plasticizer DBP—50 parts by weight. This paste was also prepared on a paint mill till a pasty consistency is obtained. The curing paste should be homogeneous without any lumps.

The sealant is prepared by mixing (a) polymer paste—100 parts by weight (b) curing paste—8–15 parts by weight and (c) accelerator (DPG)—variable (0.1 to 0.5 part) in a glass or porcelain dish. The complete curing of the sealant samples were carried out in air at room temperature (RT)(28–25°C) for 24 h and then at 70 ± 2°C for 24 h. The completeness of the curing process was indicated by the optimum mechanical properties, which do not vary with further curing. Two sealant formulations, Type A containing epoxy resin and Type B containing no epoxy resin were prepared and tested for their physico-mechanical properties in air. These properties are given in Table I. Both the sealants prepared had considerable flow without the thixotropic agent in their formulations. Too much of a flow of the sealant could be a drawback for its application to vertical and ceiling surfaces. Literature reveals some information for increasing the thixotropic properties of fluid substances by incorporating structurising additives[5–8] in small proportions into their compositions. A suitable thixotropic agent was chosen for the Type A (adhering type) sealant formulation and compounded in a liquid polysulphide polymer as mentioned earlier.

The effect of the thixotropic agent at different proportions on its flow and mechanical properties has also been studied. The flow of the sealant was determined on a device developed in our laboratory following the general principle mentioned in the literature.[9] The

TABLE I

Physico-mechanical properties of sealants at room temperature in air

Sr. No.	Physico-mechanical properties	Type A (adhering type)	Type B (non-adhering type)
1	Peel adhesion strength on anodized aluminium alloy kN/M	4.41–2.94	0.78–0.58
2	Tensile strength MPa	3.14–2.45	1.96–1.50
3	Relative elongation %	280–200	350–300
4	Residual elongation %	2–4	6–10
5	Hardness after 24 h at room temperature (RT) Shore A	42–40	40–38
6	Hardness after 24 h at RT + 24 h at 70 ± 2°C	58–57	56–55
7	Brittleness temperature °C, not less than	−40	−40
8	Pot life h.	1–6	1–6

device consists of an aluminium plate of 300×60×20 mm, at the top of the plate a hole of diameter 25 mm and depth of 10 mm. The plate is graduated in mm for measuring the flow of the sealant, the plate is kept in a horizontal position when the hole is filled up by the sealant to the level of the surface of the plate. Then the plate is placed in a vertical position and the sealant is pushed out by the plug. The sealant starts flowing under its own weight. The distance travelled by the sealant in 30 minutes time has been taken as a measure of its flow. The effect of the thixotropic agent on the flow of the sealant has been shown in Figure 1.

The curve obtained in Figure 1 shows that the addition of thixotropic agent in Type A sealant reduces the flow, thereby making it convenient for application on a vertical and ceiling surfaces. The effect of thixotropic agent on the mechanical properties of the sealant has been shown in Table II.

The results in Table II show that the incorporation of thixotropic agent does not affect the mechanical properties adversely including the adhesion.

The adhering type formulation (Type A) was subjected to detailed test in air and fuel of petroleum origin at different temperatures and time. The physico-mechnical properties of this formulation are given in Table III.

The results in Table III show that the type A sealant could be useful from −55 to +150°C in air and fuel of petroleum origin. The

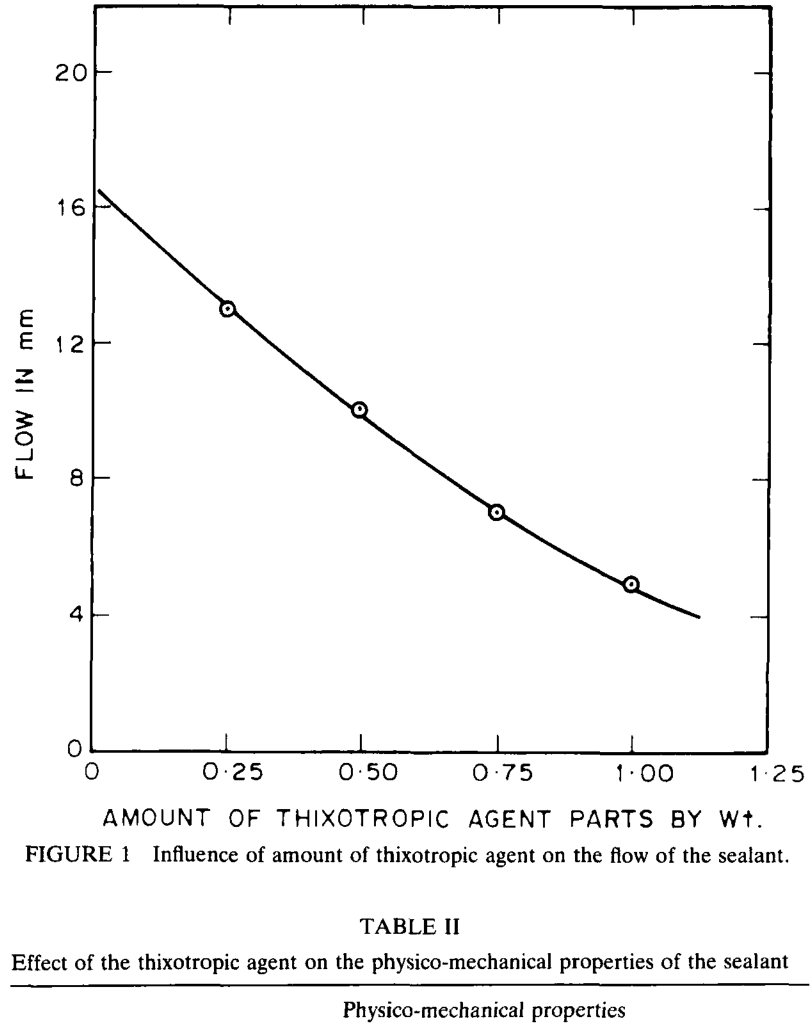

FIGURE 1 Influence of amount of thixotropic agent on the flow of the sealant.

TABLE II

Effect of the thixotropic agent on the physico-mechanical properties of the sealant

Amount of thixotropic agent parts by weight	Physico-mechanical properties				
	Tensile strength MPa	Relative elongation %	Residual elongation %	Hardness Shore A	Peel adhesion strength kN/m
0	2.79	200	4.00	54–53	4.22
0.25	2.74	210	3.00	55–54	3.82
0.50	2.84	220	2.00	55–54	3.92
0.75	2.99	240	2.00	56–55	3.92
1.00	3.08	260	2.00	57–56	3.73

TABLE III

Physico-mechanical properties of Type A sealant in air and fuel at different temperatures

	Test conditions			Properties					
Sr. No.	Temperature	Medium	Period h.	Peel adhesion strength on anodized aluminium alloy kN/m	Tensile strength MPa	Relative elongation %	Residual elongation %	Gravimetric swelling %	Fuel contamination mg/100 cc
1	25°C	Air	—	4.40	3.12	260	2.0	—	—
2	25°C	Fuel	50	4.00	2.95	210	4.0	0.10	50
3	70°C	Air	100	3.03	3.20	240	Negligible	—	—
4	70°C	Fuel	50	2.94	3.05	220	2.0	0.90	55
5	130°C	Air	100	3.04	3.42	230	2.0	—	—
6	150°C	Air	24	2.85	3.50	300	4.0	—	—
7	−55°C	Air	6	3.90	3.40	240	4.0	—	—
8	−55°C	Fuel	6	5.00	3.30	240	2.0	—	—
9	+70°C and −55°C	Fuel	30 min[a]	3.25	3.20	250	2.0	—	—
10	+100°C and −55°C	Air	20 min.[b] 30 min.	3.50	3.30	240	2.0	—	—

[a] 50 cycles, each cycle of 30 min. at 70°C and 30 min. at −55°C.

[b] 50 cycles, each cycle of 20 min. at −55°C, 20 min, at +100°C, 20 min. at −55°C and 30 min. at 25°C (room temperature)

epoxy resin incorporated into the sealant provides good adhesion in air as well as in fuel for a longer period. There is slight reduction in the relative elongation and marginal increase in tensile strength, which may be due to the hardening of epoxy resin in the sealant. The same sealant if diluted with solvents like acetone, ethyl acetate, cyclohexanone, methyl ethyl ketone in different proportions, can be used as a good coating to many substrates without altering its original properties except pot life or viability. To sum up, the sealant developed can be used as coating material as well as for sealing bolted, welded and other joints in air and fuel medium.

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