Effect of type and content of tackifier on adhesion of natural rubber and reclaimed natural rubber based sealant

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Abstract. Adhesion property of natural rubber (NR) and reclaimed natural rubber (RNR) based sealant with concrete was studied here. The effect of tackifier type and loading on the tensile properties of the rubber based sealant sandwiched between two flat cements was evaluated. There are three different tackifiers including Coumarone-indene resin (CI), petro resin (PE) and gum rosin (GR). The result initially showed that at the 20 phr of tackifier both NR and RNR sealant mixed with CI consumed the highest force to separate the rubber based sealant from the concrete. This might be due to the highest compatibility between CI and rubber. Regardless of the tackifier type, all NR based sealants showed the cohesive failure while all RNR based sealants only showed the adhesive failure. Moreover, the NR based sealant seemed to be stronger than the RNR based one. When considering the effect of CI loading on the adhesion, it was shown that for both NR and RNR based sealants, the highest stress was observed when the rubber based sealant loaded with the 20 phr of CI.

1. Introduction

Rubber adhesive and sealant are highly flexible material. They are used to join components or fill gaps between seams or on surfaces. Sealant features a reactive adhesion property adopted as a joint water-stop material for underground concrete structures, and used to prevent water leakage and protect reinforcing bars from centralizing cracks in mass concrete.

In general, both natural rubber based on polyisoprene and synthetic rubbers including silicone, polyurethane, polysulfide, styrene-butadiene rubber (SBR), butyl, acrylic or polyacrylate, isoprene, polyisobutylene, ethylene-vinyl acetate (EVA), vinyl and nitrile compounds are widely used in this application. Specifically for civil engineering, nowadays the rubber based sealant made from the reclaimed butyl rubber has commercially come into wide use in Japan, largely replacing conventional PVC. Even though the reclaimed butyl rubber based sealant has been commercially used for many years [1]. The excellent feature is that sealant adheres to green concrete during it’s hardening process. This adhering mechanism is scientifically solved. It is
based upon existence of active factors (carboxyl factor, etc.) generated during the production of sealant and while special treatment is given to butyl rubber. It was reported that metal oxide containing in the cement becomes metal water oxide by influence of water, and ion reaction is created together with active factor of sealant, thereby allowing chemical compound to be formed. Also, in addition to physical adhering mechanism of other anchoring effect, sealant adheres to green concrete through this feature [2]. The development of the rubber based sealant from other rubbers is recently concerned.

In the present study, the aim is to study the effect of tackifier type and loading on the adhesion between the rubber and concrete. Two kinds of natural rubber including virgin natural rubber and reclaimed natural rubber were used here. Coumarone-indene resin, petro resin and gum rosin were used as tackifier. A systematic study was carried out to understand the roll of tackifier type and content on the adhesion properties.

2. Experiment
2.1 Material
Natural rubber (NR) grade STR 5L and reclaimed natural rubber (RNR) were purchased Sakdarungreaungkij Co., Ltd. Three tackifiers including coumarone-indene resin (CI), petro resin (PE) and gum resin (GR) were kindly supplied by Innovation Technology Co., Ltd.

2.2 Rubber Based Sealant Preparation.
Rubber based sealants were prepared by mixing 100 phr of rubber with 0, 10, 20 and 30 phr of tackifiers and other fillers in an internal mixer (MX500-D75L90) at a high temperature. The total mixing time in the internal mixer was 30 min. After discharging, the rubber compound was sheeted off to form the rubber sheet sample with a thickness of 4-5 mm using a two-roll mill.

2.3 Adhesion Testing.
To determine the adhesion between the rubber based sealant and concrete, a PVC pipe was as a mold. Firstly, the PVC pipe was placed on the sealant sheet. Then, the wet mixed cement was poured into the PVC pipe. The sample was left under air until the cement completely solidified. Secondly, the same procedure was repeated on the another side of rubber based sealant. The maximum load needed to separate the rubber based sealant from the concrete was determined
using an Instron model 5843 at a crosshead speed of 5 mm/min with the load cell of 1 kN. The failure behavior was observed and noted.

### 3 Result and Discussion

#### 3.1 The Effect of Tackifier Type on the Adhesion.

The adhesion properties of the NR and RNR based sealant when loaded with the 20 phr of tackifier were shown in Table 1.

| Property        | Samples     |   |   |   |   |   |
|-----------------|-------------|---|---|---|---|---|
|                 | NR/CI       | NR/PE | NR/GR | RNR/CI | RNR/PE | RNR/GR |
| Maximum load (N)| 211.17      | 93.38 | 170.73 | 118.38 | 117.95 | 112.95 |
| Strain at break (%) | 3500       | 2043 | 1500 | 917 | 1875 | 428 |
| Failure behavior | cohesive    | cohesive | cohesive | adhesive | cohesive | adhesive |

From the above results, in the case of NR based sealant, it can be clearly seen that the rubber based sealant loaded with PE required the lowest maximum load to separate the rubber from the concrete. Therefore, the NR based sealant loaded with PE tackifier was the weakest sealant. The PE tackifier is synthetic tackifier consisting of long aliphatic chains [3]. The reason why PE tackifier gave the rubber sealant with the lowest adhesive strength might be attributed to the high chain entanglement between the rubber chains and the long molecule of PE, thus increasing the viscosity of the sealant and then inhibiting the sealant from wetting on the concrete. The NR based sealant loaded with CI and GR tackifier showed much higher adhesive strength than the one loaded with PE. This is due to the molecule of these two tackifiers are much smaller than the PE. However, the NR based sealant loaded with CI showed the highest adhesive strength. This might be due to the chemical structure of CI consisting less polar functional groups than that of GR, thus causing CI more compatible with the NR and then giving the NR sealant with higher adhesive strength.

The RNR used in this study is the cured rubber that was mechanically masticated to break down the crosslinking and shorten rubber chain, making the rubber be able to reprocess. However, the RNR still consists of crosslinking point, causing RNR in having higher viscosity than the NR. In this part, it was seen that the tackifier type hardly affected on the adhesive strength of the sealant [4]. This might be because the amount of tackifier used in this part was too low to loosen the
viscosity of the RNR. Therefore, the RNR based sealant loaded with different tackifiers showed the similar adhesive strength [5].

When considering the type of rubber, it can be found that the NR based sealant showed the higher adhesive strength than the RNR based one. This might be due to the existence of partial crosslinking network inhibiting the wettability of the rubber onto the concrete, thus giving the lower adhesive strength. Most of the NR based sealant showed the cohesive failure, while all RNR based sealant except the one loaded with PE showed the adhesive failure. The failure behavior of the tested sample was shown in Figure 1. The cohesive failure indicated that the interaction between the rubber and concrete is stronger than the strength of the rubber. On the other hand, the adhesive failure indicated that the interaction between the rubber and concrete is poorer than the strength of the rubber.

![Figure 1](image)

**Figure 1.** Failure Behavior of Sample (a) Cohesive failure (b) Adhesive Failure (diameter 40 mm)

### 3.2 The Effect of Tackifier Loading on the Adhesion.

In this part, only the rubber based sealant loading CI was focused. The loading of CI was varied from 0 to 10, 20 and 30 phr. The effect of CI loading on the adhesion properties was depicted in Table 2.

| Property                      | Samples          | Max. Load (N) | Strain at Break (%) | Failure Behavior |
|-------------------------------|------------------|--------------|---------------------|------------------|
|                               | NR/CI0 | NR/CI10 | NR/CI20 | NR/CI30 | RNR/CI0 | RNR/CI10 | RNR/CI20 | RNR/CI30 |
| Maximum load (N)              | 103.17 | 138.79 | 211.17 | 109.07 | 62.13 | 70.94 | 118.38 | 80.34 |
| Strain at Break (%)           | 1233   | 2250   | 3500   | 2750   | 425  | 1050  | 917    | 1400  |
| Failure behavior              | cohesive | cohesive | cohesive | cohesive | adhesive | adhesive | adhesive | adhesive |

![Table 2](image)
From the above result, for both the NR and RNR based sealant, it can be seen that the increase of CI content from 0 to 30 phr with the increment of 10 phr caused the maximum load or adhesive strength pass through the maximum. The presence of tackifier initially enhanced adhesion. However, when tackifier was overloaded, it caused the decrease of adhesion. This is generally due to the dilution effect of the rubber. The CI content giving the highest adhesive strength was at 20 phr. At each CI content, the NR based sealant gave higher maximum load than the RNR one.

4. Conclusions
This study was focused on the development of the rubber formula for the rubber based sealant application. The best rubber formula can be considered from the highest adhesive strength between the rubber and the concrete. The result showed that amongst three different tackifiers including CI, PE and GR, CI gave the rubber based sealant with the highest adhesive strength. Although the NR is much more expensive than the RNR, the NR based sealant showed much higher adhesive strength than the RNR one. The rubber based sealant filled with CI exhibited the highest maximum tensile load at the 20 phr of tackifier. Therefore, from this study the best rubber formula for the sealant application is the NR filled with the 20 phr of CI.

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