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EXPERIMENTAL STUDY ON THE AGING PERFORMANCE OF SECONDARY SILICONE SEALANT OF INSULATING GLASS UNITS

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ABASTRACT

Insulating glass units (IGUs) have been widely used as an energy saving building product. The air/water tightness performance of the edge seal system is the key to achieve the durability of IGUs. Nowadays, most edge seal consists of a dual sealant system, in which the secondary sealant is to retain the integrity of IGU. High temperature/relative humidity conditions and UV-exposure are the two most common factors that cause the detrimental aging effects upon the secondary sealant. In this paper, the cross-bonded test method is employed to examine the aging effects on the mechanical performance of silicone specimens. The tensile strength, shear strength, elongation rate at break and Shore hardness are measured for different aging periods. Aging rates are also calculated to investigate the aging sensitivity. It is found that most aging actions occur at the early age. The aging mechanisms are also discussed: excessive cross linking formation and the Si-O bond oxidation on the interface are deemed as two major reasons that attribute to the different aging behaviours of silicone sealants.

1 INTRODUCTION

Secondary insulting sealant plays an essential role for insulating glass units (IGUs) to ensure the structural integrity of the entire glass unit (as shown in Figure 1) [1]. Sufficient elastic recovery rate and adequate strengths are both essential for the secondary sealant to resist various loading actions. The durability requirement of IGU also relies on the secondary sealant to maintain stable properties, in particular, when experiencing cyclic or large deformation [2-4].

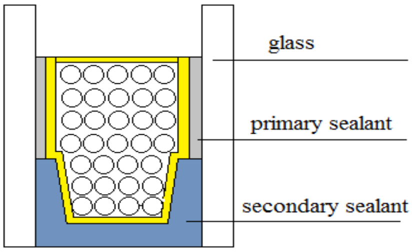


Figure 1. Cross section of the typical edge seal system of IGU

Nowadays, silicone sealant has become one of the most popular secondary sealants employed in the modern IGU products [3, 4]. As a type of organic polymer material, the mechanical properties of silicone sealant (strength, elasticity, hardness) are strongly dependent on the environmental conditions [5].

In most practical applications, these sealants are exposed to various natural environmental conditions, such as the variations of temperature, humidity and UV exposition. The sealant inevitably suffers the aging problem during the service time [6, 7]. In this paper, high temperature/relative humidity tests and UV-exposure tests are conducted for the secondary silicone sealant aiming to reveal the aging behaviours of the mechanical properties (e.g. elasticity, strength, elongation rate) under different aging conditions. Cross-bonded specimens are employed to measure the strength, elongation rate at breakage and the loading-displacement relation of the test specimens.

2 CROSS-BONDED TEST METHOD

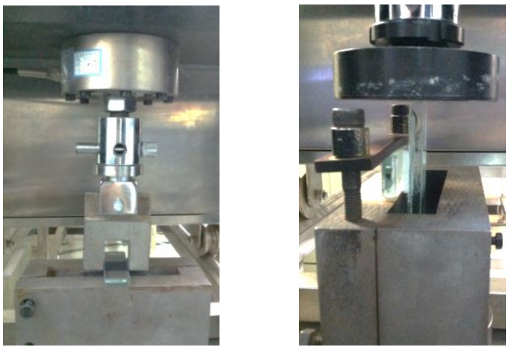
Cross-bonded test method introduced by ISO 13124 [8] is aimed to determine the tensile and shear bond strength of silicone sealants glued to other substrates, e.g. glass. A typical type of one-part silicone insulating secondary sealant is employed in this study. The geometry of test specimens is made as below (see Fig. 2): the silicone sealant is bonded with two glass bars which are perpendicularly placed as shown in Figure 2(a). The size of each glass bar is 75×24×6mm, and the bonded surface is 24(±1) × 24 (±1) mm. The thickness of sealant is 5 (±1) mm. A group of specimens, six for each test, are prepared (see Fig. 2b and c). The specimens were placed in the curing room for 28 days, with the relative humidity 50% and a constant temperature 25+-1^{o}C.

(a) Schematic diagram of test specimen (b) Moulding kits (c) Prepared cross-bonded specimens

Figure 2. Cross-bonded specimens

The prepared specimens will be subjected to two aging conditions prior to testing, which are high temperature and RH aging and UV exposure aging, respectively. According to BS ISO 23529:2010 [9], the standard laboratory ambient temperature is 25+/-2^{o}C, and the preferred relative humidity is 50%.+/-10%. The test procedures are as follows:

- Test specimens are mounted on the universal test machine for tensile and shear test (see Fig. 3).



(a) Tensile test (b) Shear test

Figure 3. Test set-up for cross-bonded tensile and shear tests

- Apply a load with a rate 5mm/min to the specimen until it fails.
- The load-displacement curves are recorded, from which the tensile and shear bond strength are calculated.

3 HIGH TEMPERATURE AND RH AGING

High-temperature and high relative humidity (RH) environments are common to the IGUs that are installed in tropical or sub-tropical regions. The excessive moisture and the oxygen in the air can react with the secondary silicone sealant and accelerate its aging process.

In this study, a high temperature and relative humidity test chamber (shown in Figure 4) is used to generate a temperature cycling regime with a high relative humidity (RH).

(a) High temperature and RH cycling test chamber (b) Control panel

Figure 4. Test apparatus

The relative humidity of the test chamber is above 95%. The temperature changes regularly with time and the cycling pattern is given in Fig. 5.

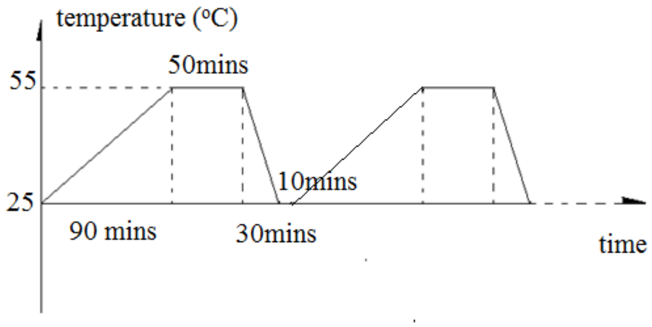


Figure 5. Climatic cycling condition in the test chamber

Aging period is taken into consideration as a parameter to study. The specimens are aged for 20 days, 40days, 60 days 80 days and 120 days, respectively. Each group contains six specimens. The aged specimens are tested with identical loading conditions.

As shown in Figure 6, the tensile and shear bond strength are markedly reduced under the high temperature and humidity environment. The tensile strength starts to decline sharply after the first 20 days and reduces by 86.18% in the 120 day total aging period. The sealant is almost completely de-bonded from glass substrate in the tensile test when the aging period exceeds 80 days.

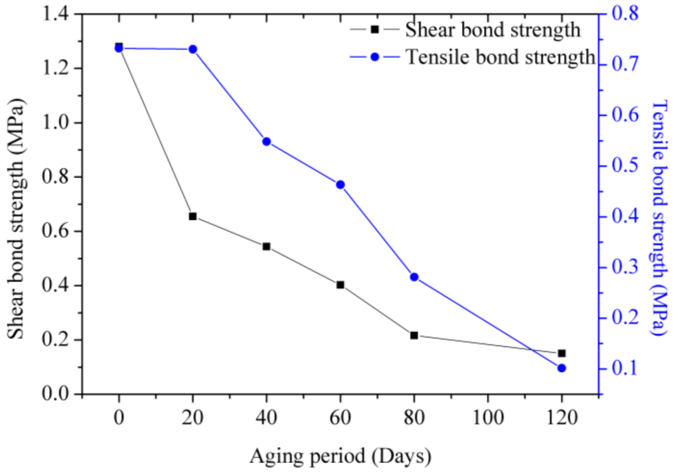


Figure 6. Tensile and shear strength variation with aging time

Similarly, the shear strength is also greatly reduced by 88.24% over 120 aging days. In contrast to the tensile strength that is only subjected to the minor change in the first 20 days, the decreasing rate for the shear strength is greatest in the first 20 days, which is followed by a mild drop until the failure at the end of aging period.

The bonding performance of silicone sealant depends on two main contributors, i.e., the interfacial adhesive behaviour and the sealant cohesive performance. The adhesion performance is mainly governed by the chemical bonds at the glass-sealant interface, while the cohesive performance relies on the chemical bonds and intermolecular interactions within the sealant. The chemical backbone of silicone sealant is a polydimethylsiloxane (PDMA) [5]. When subjected to the high temperature, the adhesion interface turns to be a high temperature anoxic environment. Since there is no sufficient oxygen to activate the methyl oxidation, the scission of the backbone Si-O bond will be induced by the high energy [11]. PDMA is degraded into a variety of LMW (low-molecular weight) polymers such as hexmethyl/octamethylcy - cyclotrisiloxane, and other oligomer chains. The Si-O bond scission at the interface due to high temperature is a main reason to the degraded bonding strength.

Within the sealant, the high humidity provides abundant moisture to activate the hydrolysis of PDMAs and hence promote the formation of more cross-linking. Meanwhile, high temperature will accelerate the oxidation in PDMA. Since the backbone of PDMA, Si-O bond, possesses the highest bond energy [10], the oxidation is more likely to happen to the methyl side groups with the lower bond energy and form hydroxyl ended siloxanes and the carboxyl (-COOH) group. The labile carboxyl group then converts into protons and carbon dioxide. The hydroxyl ended siloxanes will further develop cross-linking. The increase in the three-dimensional PDMA net structure results in a higher molecular weight, hardness, cohesive strength and lower elongation rate at breakage. The changes in these mechanical properties will also cause the loss of resilience.

An ageing coefficient is proposed to indicate the aging rate within a certain period, which is calculated as

$$V = \frac{(n_0 - n_t)/n_0}{t} \times 100 \tag{1}$$

where, V is the aging rate in unit of %/h, n_0 is the initial property, n_t is the new property after aging and t is the aging period (h).

The sensitivity analysis to the high temperature and RH is presented below in Figure 7. Positive values demonstrates the improved property and vice versa.

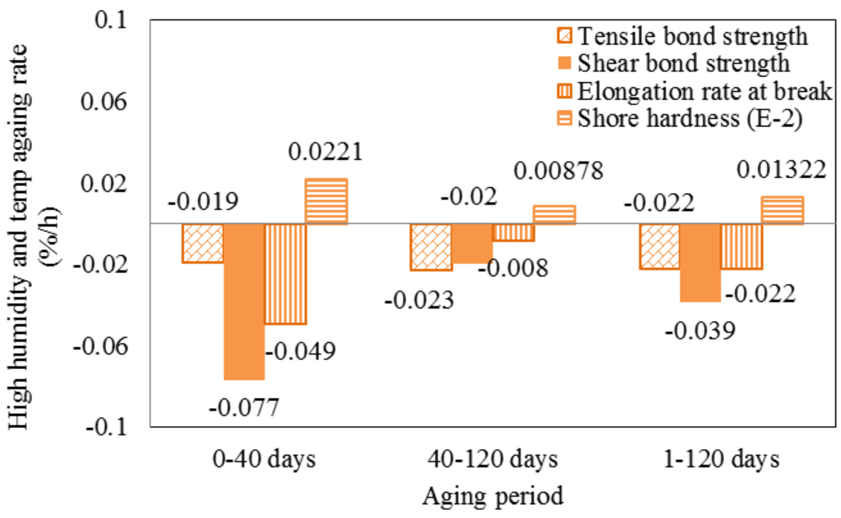


Figure 7. Sensitivity analysis to high temperature and RH aging condition

According to Figure 7, the tensile, shear bond strength and the elongation rate at breakage rate are degrading under the aging condition whereas the shore hardness is enhanced. Apart from the tensile strength, other properties are more sensitive in the initial aging period, i.e. 0- 40 day, in which the aging rate is higher than that in 40 to 120 days. On the contrary, the tensile strength aging rate at a later aging period is higher than the earlier stage. This is because the further cross-linking formation due to the methyl oxidation at the early stage plays a beneficial role to the tensile bond strength. Such beneficial effect counteracts the bond strength degradation caused by Si-O scission at the interface. In the latter aging periods, the action of Si-O bond cleavage governs the increase in the degradation rate.

4 UV EXPOSURE AGING TEST

Installed IGUs are always directly or indirectly exposed to sunlight. Sunlight spectrum consists of ultra-violet, infrared radiations and visible light. The energy of light is inversely proportional to its wave length. The wave length of ultra-violet radiation is lower than 280 nm, visible light between 380 to 780nm and infrared rays higher than 780nm. The energy of ultra-violet radiations is the highest and therefore imposes greatest threat to the sealant components. Normally the UV-exposure always combines with high temperature and hence forms the dual attack to IGUs. Therefore the dual environmental impacts should be considered in the test.

The specimens are placed in the UV chamber (shown in Figure 8) and exposed to an MLU ultraviolet radiator of 300W. The output power is no lower than 40W/m^2. The ambient temperature in the chamber is set to be 50±3^{o}C. Similar to the high temperature and RH aging tests, the specimens are grouped according to different aging periods, i.e. 20 days, 40 days, 60 days, 80 days and 120 days.

Figure 8 UV exposure chamber

The tensile and shear strengths are plotted against aging time in Fig. 9. The tensile strength is found to be increasing with the aging time initially and then experiencing a drop from 20 to 40 days before it increases again (see Fig 9). On the other hand, there is a sharp drop for shear strength in the first forty days, and then undergo a minor recover before it becomes stable. The overall rise of tensile strength is 49.04% while the decrease of shear strength is 40.64%.

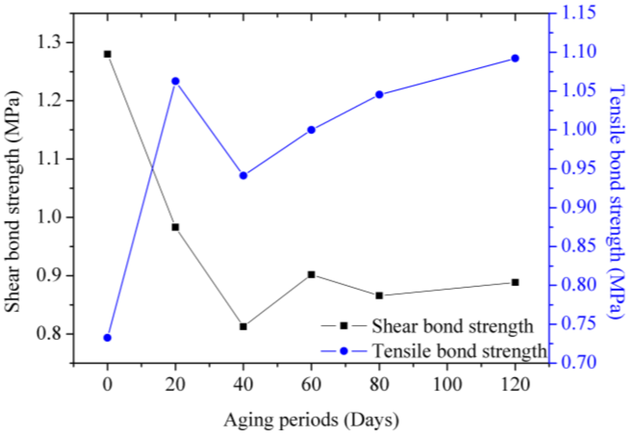


Figure 9. Tensile and shear strength variation with aging time

Opposite aging trends of tensile and shear strengths are attributed to the joint action of the strengthened cross-linking reaction and the Si-O bond cleavage. The UV exposure and high temperature provide sufficient energy to intensify the chemical reactions within the organic polymer sealant. Two types of reactions will take place simultaneously in different ambient conditions. Further cross-link reaction commences in the aerobic ambient where the methyl side groups are replaced with hydroxyl by oxidation. The cohesive strength is therefore increased. The Si-O scission occurs on the bond interface, i.e. relatively anoxic ambient, triggered by the high energy of UV radiator and higher temperature. The interfacial adhesive strength will decrease due to the cleavage of the interfacial chemical bonds. The results suggest that the cross-linking reaction is a primary influencing factor to the tensile bond strength, so it can be enhanced by an enhanced level of cross-linking reaction even though the Si-O bonds are weakened on the interface. It is also observed that the bond shear strength is more sensitive to the decomposition reaction of Si-O bond on the interface. When the Si-O bond scission occurs, the shear bond strength is directly affected negatively.

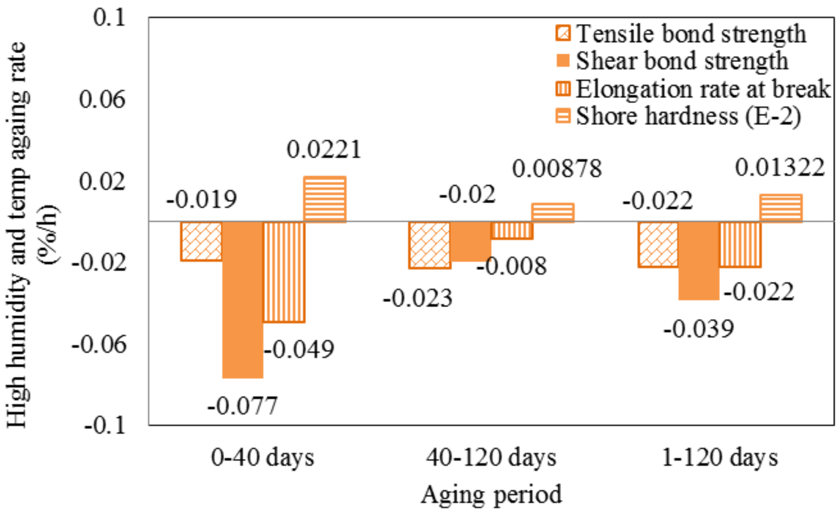


Figure 10. Sensitivity analysis for UV exposure aging condition

The aging sensitivity analysis is presented in Figure 10. The aging rates in the early age are much higher than the later period. So the aging environment is more influential to the sealant strength at the early age. The elongation rate at break increases slightly in the beginning and then significantly declines with the longer aging time. The increasing rate of Shore hardness remains stable over the entire aging period.

5 SUMMARY

In this paper, the aging behaviours of IGU's secondary silicone sealant are analysed experimentally. The effects of high temperature and high RH condition and UV exposure are examined to investigate the sealant aging behaviours under the environmental attacks during their service life. It is found that most aging behaviours take place at the early age e.g. 0-40 days.

The aging mechanisms can be ascribed to two main molecular reactions: the excessive cross-linking reaction because of methyl oxidation or the hydrolysis that leads to the loss of elasticity; Si-O bond scission on the sealant-glass interface due to high temperature or UV exposure resulting in the degradation of adhesion strength.

It is noted that further cross-linking reaction can reinforce the sealant cohesive strength, and hence increases the tensile bond strength even though the interfacial adhesion is deteriorated. The aging rates of each physical property of silicone sealant are also presented. The sensitive ranges can be found by comparing the aging rates in different aging periods.

ACKNOWLEDGEMENT

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