Effects of Waste Glass Powder Filler on Intumescent Coating for Steel Structures Application

Y M Azmi¹, F Ahmad*, S N Razak², M A H A Hadi¹, S Kabir¹ and G H Yeoh³

¹ Department of Mechanical Engineering, Universiti Teknologi PETRONAS, Malaysia
² Laboratoire Angevin de Mecanique, Procedes et Innovation of Arts et Metiers, France
³ School of Mechanical & Manufacturing Engineering, University of New South Wales, Australia

*Corresponding author: faizahmad@utp.edu.my

Abstract. Intumescent coating is a passive fire protection system used to prevent and resist the spreading of fire. The coating swells several times of its thickness on exposure to the fire, forming cellular foam and acts as an insulative barrier, thus delaying the heat transfer to the protected substrate. The study aims to analyse waste glass powder filler’s effect into intumescent coating formulation limited to one size of waste glass powder. Waste glass is a broken form of leftover by-product from industrial and domestic. Millions of tons of waste glass generated every year, adding with their non-biodegradable nature causing additional environmental pollution. Waste glass powder contains high silica composition known for its high thermal stability, weather resistance, hydrophobicity, low surface tension, and high oxidation resistance. The coating showed improved thermal stability, weather resistance, and adhesion when added into intumescent coating formulation. The lowest recorded backside substrate temperature 154.6 °C compared to coating without the waste glass filler, 333.7 °C. The coating also showed less than 0.1% total weight loss for samples with 4% wt. filler after immersion in seawater for 15 days compared to the control formulation, 0.19%. The char morphology analysis showed the emergence of silica and calcium elements occupying the expandable char’s gaps, thus strengthening the char structure.

1. Introduction

Fire losses are among the most disastrous accidents, be it from a human mistake or natural causes. The short time it took to spread and decrease escaping time, resulting in various protection types, was created to either slow it or prevent it. Two significant types of protection available in modern civilisation are active (AFP) and passive fire protection (PFP). Active fire protection requires an action taken to put out a fire, such as a sprinkler system, while passive fire protection prevents or resist the spread of fire. An example of PFP is an intumescent coating (IC). The intumescent coating was applied like paint into the protected substrate and acted to prevent the spreading of fire and delay the heat transfer rate. When exposed to fire, the coating reacts and swell from its original thickness, thus foaming a carbonaceous insulation barrier obstructing the heat transfer to the protected substrate[1]. According to Mariappan (2016), the intumescent coating mechanism comprises five steps. It starts with the decomposition of catalyst material into mineral acid. The acid reacts with carbonic by dehydration to form an ester, thus
decomposing and forming a carbonaceous char and releasing the acid. The resin material then melted, forming a skin over the char and the blowing agent releasing an inert gas, resulting in the expansion of char that act as efficient insulation[2]. The modernisation of intumescent coating has shown that the researchers eagerly interested in adding nanofillers for various purposes, such as smoke suppressant, corrosion resistance, thermal conductivity, etc. Filler generally incorporated into the formulation in the range of less than 40% of IC mass.

Glass is a unique multipurpose substance with more comprehensive applications naturally occurring when rock high in silicates melt at high temperature and cool before forming a crystalline structure. Waste glass is a broken form of leftover by-product from industrial and domestic. Millions of tons of waste glass generated every year, adding with their non-biodegradable nature causing additional environmental pollution. In Malaysia, the lack of glass recycling of glass product making a rising effect on the environment. The common types of glass found in domestic are soda-lime glass, lead-alkali-silicate glass and borosilicate glass. These glasses commonly manufactured to form a window, a bottle, glassware and laboratory equipment used in the house and other domestic industries, making the amount of waste glass rising every year.

Research nowadays used waste glass powder (WGP) in the cement and concrete industry. The pozzolanic reaction from the waste glass powder is highly sought due to its advantage of increasing the end product’s durability and providing compressive strengths. Decreasing the particle size of glass powder encouragingly increases pozzolanic reactivity [3]–[6]. Glass as a filler also reported by a researcher to help increase the thermal stability and dynamic thermal-mechanical properties and reduce the coefficient of thermal expansion[7]. Few studies on using the waste glass powder in the epoxy and ceramic coating reported, but little to none in the field of intumescent coating formulation studied[8]–[10]. This paper discussed the effect of waste glass powder as filler into intumescent coating limited to one size of waste glass powder. The effect of different size of WGP was not analysed.

2. Experimental Materials and Procedure

2.1. Materials
Bisphenol A diglycidyl ether (DGEBA) with Equivalent Weight (E.E.W) 188, expandable graphite (EG), and zinc borate (ZB) purchased from Mc-Growth Chemical Sdn. Bhd. Boric Acid (BA), melamine (MEL) bought from Sigma-Aldrich (M) Sdn. Bhd. Malaysia. Ammonium polyphosphate (APP) supplied by Clariant (Malaysia) Sdn. Bhd and waste glass obtained from factory glass & plastic packaging (Ipoh), Malaysia.

2.2. Experimental Procedures
All the materials weighed according to their respective weight percentage as per table 1. BA, ZB, Mel, and APP was ground for one minute before mixed with EG and epoxy in a sheer mixer machine at 30 rpm for 15 minutes. The hardener slowly introduced into the mixer, and the process continued for another 10 minutes. The coating then applied to ASTM A36 steel substrate before leaving to cure at room temperature.

Table 1. Intumescent coating with the waste glass powder formulation

| Samples | APP   | MEL | BA   | EG | Epoxy & Hardener | Waste Glass Powder |
|---------|-------|-----|------|----|------------------|--------------------|
| WG000   | 11.23 | 5.5 | 11.11| 5.5| 66.66            | 0                  |
| WG001   | 11.23 | 5.5 | 11.11| 5.5| 65.66            | 1                  |
| WG002   | 11.23 | 5.5 | 11.11| 5.5| 64.66            | 2                  |
| WG003   | 11.23 | 5.5 | 11.11| 5.5| 63.66            | 3                  |
| WG004   | 11.23 | 5.5 | 11.11| 5.5| 62.66            | 4                  |
| WG005   | 11.23 | 5.5 | 11.11| 5.5| 61.66            | 5                  |
3. Characterisation and Analysis

3.1. Thermogravimetric Analysis (TGA)
Perkin Elmer Pyris Thermogravimetric Analyser was used for this analysis to determine the residual mass and thermal behaviour of the waste glass powder using a heating rate of 10℃/min under nitrogen flow from room temperature till 900℃.

3.2. Field Emission Scanning Electron Microscope (FESEM) and Scanning Electron Microscope (SEM)
Zeiss SUPRA 55-VP FEGSEM were used to study the morphology structure of char’s surface and cross-section. The samples prepared by burning them in the Carbolite furnace for 1 hour at 600℃ before subjected to the analysis.

3.3. Fourier Transform Infrared Spectroscopy (FTIR)
The technique based on the functional groups’ IR absorption frequencies and spectra was recorded in 450 to 4000 cm⁻¹ using the KBR-FTIR method.

3.4. X-ray fluorescence (XRF)
This non-destructive analytical technique used to determine the elemental & oxidation composition of materials. The samples were analyse using Bruker S8 Tiger.

3.5. Direct Fire Test (UL94 Lab scale) and Control (Furnace) Fire Test.
The cured samples exposed to UL94 in a lab-scale test to evaluate the coating's thermal performance. The samples back substrate was attached with three thermocouples and hold with a retort stand. The coating sample then exposed to butane gas fire, and the back substrate's temperature and the physical observation recorded for 1 hour.

A control fire test was performed inside a Carbolite Gero Furnace to examine the morphology and expansion of char in a controlled environment. The cured coating burned for 1 hour at a temperature of 600℃ with a 20℃/min rate. The samples thickness before and after burning recorded, and intumescent factor (IF) was calculated based on the following equation:

\[ IF =\frac{(D_2-D_0)}{(D_1-D_0)} \]  

\[ D_2 \] is the thickness of the char with the substrate, \[ D_1 \] is the thickness of coating with the substrate, and \[ D_0 \] is the thickness of the steel substrate.

3.6. Water Immersion Test
The test performs according to ASTM D870-15, Standard Practice for Testing Water Resistance of Coatings Using Water Immersion[11]. All the cured intumescent coating samples were weighted before immerse into seawater obtain from Teluk Batik, Perak. The samples weight was recorded every three days for 15 days, and the last weight recorded after the samples left for drying for 24 hours.

4. Results and Discussions

4.1. Waste Glass Powder Analysis
The waste glass powder was subjected to characterisation analysis to study the material's thermal stability and composition. The graph in figure 1 displayed a low decomposition percentage of 0.78% at the 800℃, even with the sharp slope of -0.0014. This lower value of slope indicates the material is thermally stable and could enhance the intumescent coating's thermal performance. Note the average temperature of the back substrate intumescent coating reported by Ahmad (2015-2017) group was less than 400℃ at the end of 1-hour exposure to fire[12]–[14].
The raw samples sent for X-Ray Fluorescence (XRF) to determine the elemental composition of the waste glass powder. The data obtained listed in table 2 and Table 3. As expected, the waste glass powder contains high silicon with 33% of the total elements, with the possibility of the emergence of SiO$_2$ at 70.60%. Silicon was known for having high thermal stability, weathering resistance, hydrophobicity, low surface tension and high oxidation resistance when added in fire resistance material [15]. In addition, the powder also contains 13% of calcium element with the possibility of the minor components of sodium, aluminium, magnesium, phosphorous, tungsten, iron, potassium, titanium and sulphur.

![Figure 1: TGA thermogram of raw waste glass powder](image)

| Waste Glass powder | Element Composition (%) |
|--------------------|-------------------------|
|                    | Si  | Ca  | Na  | Al  | Mg  | P    | W    | Fe   | K    | Ti   | S    |
|                    | 33  | 13.5| 3.75| 0.778| 0.563| 0.453| 0.438| 0.273| 0.129| 0.119| 0.116|

| Waste Glass powder | Oxides Composition (%) |
|--------------------|-------------------------|
|                    | SiO$_2$ | CaO | Na$_2$O | Al$_2$O$_3$ | P$_2$O$_5$ | MgO | WO$_3$ | Fe$_2$O$_3$ | SO$_3$ | TiO$_2$ | K$_2$O |
|                    | 70.60    | 18.90 | 5.06 | 1.47 | 1.04 | 0.93 | 0.55 | 0.39 | 0.29 | 0.20 | 0.16 |

4.2. Direct and Control Fire Test

A control fire test performed to evaluate the char expansion with IF values as an indicator. The sample without filler (WG000) showed a brittle like char structure with shrinkage at the substrate's corner. This is not the case for the samples added with waste glass powder. The char expanded fully, and no shrinkage detected between the samples. The leftover char shown in figure 2 still intact with the substrate even after the cross-section cut indicates an excellent adhesive bond successfully created between the substrate and coating ingredient. Chowaniec (2019) studied on adhesive properties of waste glass powder added reported to cause an increase of pull-off strength of the coating. The microstructural analysis of the samples showing waste glass powder penetrates the coating to the substrate and fill the pore, thus strengthening the adhesive[10]. The Intumescent factor used to determine the ratio of the char with the coating. According to Jimenez (2006), char expansion is an essential parameter for evaluating coating performance but not necessary. If a brittle char forms when exposed to fire, it tends to break when subjected to external force (e.g., jet fire), thus decreasing efficiency in the turbulent regime of
This type of protective coating not giving viable security to the steel structure. No significant improvement in the char expansion when waste glass powder added to the formulation.

The average value obtained from samples WG001 to WG005 is 7.55, with WG004 having the slightest highest value of 7.93 compared to WG000 (without WGP) with an IF value of 10.44 in figure 3. Even though calcium reported to increase the char’s expansion [17], this is not the case for this research as this might due to less calcium composition in waste glass powder (18.90%) shown in previous table 2. The coating also prepared for direct fire test to evaluate their performance against real fire. A 10 x 10 x 1.5 cm coated substrate used for this test. After an hour, the result tabulated into the graph in figure 4. Sample WG005 recorded the highest temperature obtain after an hour duration with 370.3°C, follow by WG000 with 333.7°C. The other samples containing waste glass powder filler showing temperature less than 300°C with WG002 recorded as the lowest (154.6°C) follow by WG001 (230.4°C), WG003 (257.1°C) and WG004 (259.7°C).

**Figure 2:** Cross-section Expanded char with and without waste glass powder

The samples excellent performance with waste glass filler might come from the high content of silica with 70.06%. Silica was known for its high thermal stability and the credit gain from the Si-O-Si bond structure[15]. Despite no char expansion improvement when WGP added into the formulation during the control fire test, the direct fire test showing an opposition with an excellent lower temperature sustain for an hour exposure to fire. This showed that the char exhibit a denser structure that delays the heat's penetration to the protected substrate, thus improving the coating's thermal performance.

**Figure 3:** IF values of char expansion with and without WGP.

**Figure 4:** Temperature VS Time graph of intumescent coating with and without WGP.

4.3. Water Absorption of Coating (ASTM D870-15)
The cured intumescent coating fully immersed in seawater for 15 days, and weight loss recorded after drying with a dry cloth every three days. The samples were then left to dry at room temperature 24 hours before weighted again. The bar chart in figure 5 shows a weight-loss trend at the end of 15 days with an additional 24 hours of room temperature dried. The samples with 3% of waste glass powder had the highest final weight loss between samples with filler with 0.128%, while WG004 had the lowest value.
with 0.078%, followed by WG002 with 0.089%, and WG001 with 0.103%. WG005 exhibits an increase of weight by 0.035%. Meanwhile the intumescent coating without additional filler exhibits a 0.19% weight loss after dried at room temperature for 24 hours. In the first nine days of immersion, all the coating exhibits permeation and migration simultaneously. In permeation, molecules and ion from seawater penetrate the coating structure leading to an increase of the weight in the coating while hydrophilic fire retardant additives migrate from the coating and are solved in water during the migration process, resulting in a weight loss of the coating [17], [18]. The sample reached equilibrium after nine days of immersions except for WG005, with a significant drop in weight observed. This might occur due to the overloading of waste glass powder added into the formulation, resulting in a higher migration rate, allowing the mineral of salt to accumulate into the pore structure and increasing the samples' final total weight. Overall, the average of less than 0.1% total weight loss and gain for all the samples with waste glass filler proves that the material's water resistance properties improved by adding waste glass filler. However, all the samples demonstrate a suitable water resistance property as it is lower than the maximum requirement weight loss of 3%.

![Figure 5: Weight loss after immersed in seawater for 15 days and 24 hours dried](image)

4.4. IR analysis
IR analysis performs to determine the functional groups of the intumescent coating shown in figure 6. A nearly similar trend of spectra peak observes from the figure. A broader and prominent peak was detected between 3100 to 3600cm\(^{-1}\) for the sample with waste glass powder compare to WG000. This presence of broadband may due to O-H stretching from strong intramolecular and intermolecular hydrogen bonds[19]. O-H group reported encouraging the hydrogen bonding between the substrate and the coating itself[20].
4.5. Morphology of Char

Sample WG002 chosen as the best samples having the lowest back substrate temperature recorded against direct fire with 154.6°C, having the second-lowest for total weight loss on water absorption test and an average of IF 7 value on char expansion. The sample then sent together with the sample without waste glass powder (WG000) for FESEM analysis to further analyse their morphology structure. The interior char morphology of the samples with and without waste glass powder present in figure 7. The sample without the filler showing an empty and open space on the expandable graphite worm-like structure. This situation opposed WG002 with 2% of waste glass powder with an agglomeration of small round shape occupying expandable graphite structure gaps. This shape likely identifies as silica that reported by other researchers [21], [22]. The small flakes like shapes observed in figure 7b, possibly from calcium elements [23].

Figure 6: IR spectra of intumescent coating with and without waste glass powder filler

Figure 7: Morphology image of intumescent char samples without filler, WG000 (a) and sample with filler, WG002 (b)

5. Conclusions

Waste glass powder contains as high as 70% silica and has high thermal stability, with a percentage of more than 90% when decompose to 800°C. When added into the intumescent formulation, the coating’s thermal stability increases where the lowest back substrate temperature recorded was 154.6°C compare to coating without the waste glass, 333.7°C. The morphology analysis also shows the emergence of silica and calcium elements that occupy and close the char structure gap, thus slowing the heat transfer to the protected substrate. The coating showed a less than 0.1% total weight loss for samples with less than 4% wt filler after immersed in seawater for 15 days compared to the control with 0.19% wt. The
char subjected to a cross-section cut, and all the samples still left with char intact to the substrate. This leftover char intact with the substrate indicates an excellent adhesive bond formed between the coating and substrate. The coating's good adhesive bond is shown by the O-H functional group's high intensity promote the hydrogen bonding between those two. Sample WG002 with 2% of waste glass powder was chosen as the best samples with the lowest back substrate temperature recorded against direct fire with 154.6℃, and having the second-lowest for total weight loss on the water absorption test and an average of IF 7 value on char expansion. Hence, this study's findings indicate that waste glass powder's addition to intumescent coating formulation effectively enhances the intumescent coatings' thermal and mechanical properties. It is recommended analysing further the current work, including the char's compactness measurement using a rheometer as the char's measurement has a significant effect on the flow heat to the substrate.

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