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EFFECT OF SILICA ADDITION IN THE ADHESIVE PRIMER LAYER OF COMPOSITE SYSTEMS USED TO REPAIR LEAK DEFECTS IN PIPELINES

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Abstract. *Fibre reinforced polymer composites are often used to repair through-thickness corrosion damage in metal pipes. Usually, a primer adhesive layer is initially applied to the surface of the pipe and then the composite reinforcement is applied in concentric layers. This paper investigates the possibility of adding silica as a low-cost thixotropic thickener to a commercial epoxy resin to facilitate the application of the prime layer while maintaining the strength and effectiveness of the repair after curing. The study of the influence of the silica concentration on the rheological behaviour of the adhesive before fully cured was performed using a rheometer. Hydrostatic burst tests were carried out on specimens with through-thickness defects, repaired using the same composite, but with two different initial adhesive layers (same surface finish and layer thickness, but with silica and without silica). The study shows that introducing a low fraction of silica facilitates the application of the primer and does not significantly affect the strength of the repair. The introduction of a small fraction of silica powder (1% by weight) in a commercial adhesive increases its apparent viscosity, facilitating its application as primer layer adhesive (an adhesive used in some repair systems to bond the composite laminate to the substrate) on the surface of corroded pipelines. This is particularly important in the repair of leakage defects in complex positions. The adhesive becomes fluid and easy to handle as it is applied to the pipe surface but becomes a gel when left undisturbed. Because of these qualities, this adhesive is less likely to drip from a brush than the adhesive without silica. Besides, the silica does not affect severely the mechanical performance of a composite repair system after the resin is fully cured. This thickener has proved to be remarkably effective in imparting thixotropic properties to coating masses. In the case of through-wall defects, the failure pressure is lower when using silica in the primer layer adhesive. However, this can be easily circumvented by increasing the thickness of the composite repair. Another interesting remark is that all the main conclusions in this paper apply for pipe repairs using bonded metallic or composite patches instead of composite gloves. The addition of such a low-cost thixotropic thickener makes the application of the adhesive layer in composite repair systems easier while maintaining good adhesive properties and avoiding leaking.*

Keywords: *Corroded Pipelines, Composite repair systems, Adhesion, Thixotropic Thickener, Silica*

1. INTRODUCTION

The use of composite repair systems in corroded metallic pipelines (da Costa Mattos et al 2009, 2012, 2014, 2016, Watanabe Junior et al, 2016, Sathler et al 2021) with through-wall defects requires an adequate surface preparation (in order to achieve the roughness levels required for the use of the selected adhesive) and the application of an epoxy adhesive with short curing time. The epoxy layer should have a smooth boundary for improved performance. Smoothing is needed to not allow gaps between the substrate and the composite glove (Watanabe Junior et al, 2016).

The present study is concerned with the introduction of a thixotropicizer and thickening agent in a commercial resin in order to prevent it from flowing on vertical surfaces (Coussot, 2014, 2018). A thickening agent or thickener is a substance which can increase the viscosity of a liquid without substantially changing its other properties. Silica powder is used as a low-cost post additive to adjust final viscosity. The concentration level is important as too high a level can affect the mechanical strength.

The focus is the effect of the addition of very fine silica powder (in an amount not exceeding 2% by weight) in the failure pressure of a composite repair system in a hydrostatic test. Initially it was analyzed the effect of different fractions of silica in the rupture force in tensile tests of bonded hybrid metal-composite joints. The study shows a good compromise between viscosity/mechanical strength for very low fractions of silica, with a better result being 1% by weight of epoxy resin. Then, hydrostatic test specimens with 10 mm, 15 mm and 25 mm holes were repaired (using the adhesive with 1% weight addition of silica and without silica). The failure pressure and corresponding critical energy to the delamination were analyzed. The study shows a good compromise between viscosity/mechanical strength for such a very low fractions of silica. The addition of such a low-cost thixotropic thickener would make easier to apply the adhesive layer in composite repair systems with good adhesive properties.

2. MATERIALS AND METHODS

The hybrid single lap joints used in this research were fabricated with a steel substrate and a composite substrate made of an epoxy resin reinforced with glass fibres and bonded with an epoxy-based adhesive. In the next sessions each of the components will be carefully described.

The composite substrate was an epoxy resin + bi-directional E-glass fibre composite, commonly used to repair and reinforce internal and external corrosions on pipelines. The metallic substrate selected for the fabrication of joints was ASTM 1020 steel. This steel has good machinability, is easy to form because its alloy is very ductile and can be welded easily. For these reasons it is widely used in industry and has been selected for use in this research. The properties of the composite and metallic substrates are presented in Tables 1 and 2.

Table 1. Composite substrate properties.

Property	ASTM 1020 steel
Tensile strength at the fibre directions (MPa)	440
Shear modulus (G_{31} , MPa)	2147
Elasticity Modulus at the fibre direction (E_{11}, E_{22} , MPa)	23335
Poison's ratio (ν_{12})	0.147

Table 2. Metallic substrate properties.

Property	ASTM 1020 steel
Yield strength (σ_y , MPa)	350
Tensile strength (MPa)	420
Young's modulus (MPa)	186000
Poison's ratio (ν)	0.3

The adhesive consisted of a two-part epoxy-based adhesive with excellent mechanical and corrosion resistance. The tests have been performed with the addition of pyrogenic silica powder (0%, 1% and 2% by weight. See Fig. 1). 5 Specimens were used for each silica fraction. The technical data about this pyrogenic silica are depicted in table 3.

Table 3. Pyrogenic Silica - Technical data

Property	Pyrogenic Silica
BET Surface (DIN ISO 9277/ DIN 66132)	170 – 230 m^2/g
Loss on drying (DIN EN ISO 787-2)	<1.5%
pH (DIN EN ISO 787-9)	3.8 – 4.3
Tamped residue (DIN EN ISO 787-11)	40 g/l
Sieve residue (DIN EN ISO 787-18)	<0.04%

Figure 1. Addition of silica in the adhesive

There is no specific standard for testing hybrid bonded single lap joints, the ASTM D1002-10 have been used, and the bonded area A is equal to L^2 (L is the width of the substrate (see table 4)

Table 4: Single lap joint specimen dimensions.

Component	Width (mm)	Length (mm)	Thickness (mm)
ASTM A1020	24.5	100	4.76
COMPOSITE	24.5	100	5.2

The first step to the preparation of the joints is to guarantee a minimum roughness is obtained on the surface of the metallic substrates, to obtain a proper adhesion between adhesive and substrates. For this matter it was used a shot peening machine with steel grit G40 and a compressed air pressure of 0.45 MPa. Later, the metallic substrates are cleaned with acetone. A Taylor-Hobson CCI-MP non-contact profiler was used to make sure the necessary roughness was obtained, and the results are presented in Figure 2. The parameter Rt (total roughness) was used, following the standard ISO 4288 and the values obtained were between 51.7 µm and 70.9 µm. Next, a base is prepared to maintain the alignment of the joints with holders being placed on the opposite side of the base of the joint to assure the desired adhesive thickness. A thin layer of adhesive is placed in the metallic substrate, then it is positioned in the cast and the composite substrate is carefully positioned over the metallic substrate. Finally, a steel plate closes the cast. Once the single lap joints are finalized, it is given a one-week period to make sure the adhesive is fully cured before the beginning of the tests.

All tensile tests were performed in a Shimadzu Servo-Pulser universal testing machine, with hydraulic grips and a 100 kN load cell. The tests were made accordingly to ASTM D1002, with a 140 millimeters distance between grips and a crosshead speed of 1.3 mm/min. Finally, to guarantee the alignment of the joints in the testing machine and avoid flexure loads, tabs were attached to its extremities.

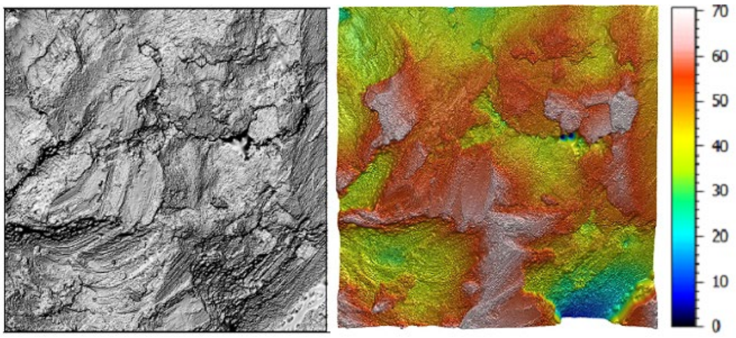

Figure 2. Roughness measurement.

The tensile tests were used to obtain a reasonable choice between 1% and 2% addiction in weight of silica in the adhesive. The best compromise between mechanical properties and adequate increase of viscosity was used to compare the failure pressure of composite repair systems in hydrostatic tests.

The hydrostatic specimens have 152.4 mm (6-inch) nominal size and 60 cm in length. In order to simulate a through-thickness damage, holes of 3 different diameters (10 mm ,15 mm and 25 mm) were drilled in the pipe walls. After applying the adhesive coating, a 3 mm thick composite reinforcement is applied in concentric coils. 3 tests were performed for each hole diameter. Thus, a total of 18 hydrostatic burst tests (3 different hole diameters and 2 adhesives) were performed according ASME PCC-2 and ISO Technical Specification 24817.

The external pipe wall surface was prepared using a bristle blaster tool for systems 1 and 2 and shot peening for repair system 3. Fig. 3 shows an example of a pipe specimen after the surface preparation process. Figs. 4 and 5 show, respectively, the application of adhesive layer and the application of the composite repair. All tests (tensile tests in lap joints and hydrostatic tests) were performed one week after the specimens´ fabrication.

Figure. 3. Pipe specimen after surface preparation.


Fig. 4. Application of adhesive layer.

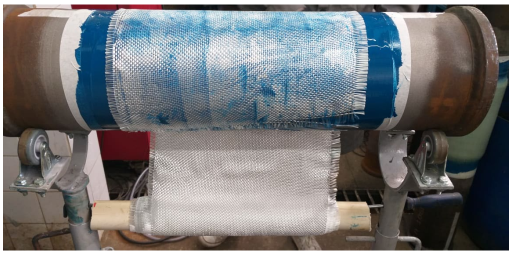
Fig. 5. Composite repair application.

3. RESULTS AND DISCUSSION

Table 5 presents the rupture force obtained in the tensile tests of single lap joint tests for different fractions of silica.

Table 5. Rupture force obtained for different fractions of silica.

	0% - Rupture force (N)	1% - Rupture force (N)	2% - Rupture force (N)
CP1	4717.69	4755.56	3865.58
CP2	4698.64	2968.89	3430.56
CP3	4113.21	5216.07	4039.35
CP4	6504.08	4324.80	3476.07
CP5	5695.48	4025.27	2755.71
MEAN	5145.82	4258.12 (-17%)	3513.45 (-31%)
Standard deviation	948.23	849.39	495.74

The addition of silica changes a lot the behaviour of the adhesive as it can be seen if Fig. 6.


Figure 6. Adhesive after the mixture with the thickener.

The addition of 1% of silica in weight seems a good compromise between strength (reduction of 17% in the rupture force and good applicability).

The hydrostatic burst tests have been performed with the addiction of 0% and 1 % of silica. Table 6 presents the experimental results.

Table 6. Rupture force obtained for different fractions of silica.

		0% - Failure Pressure (bar)	1% - Failure Pressure (bar)
10 mm	CP1	65.86	58.61
	CP2	59.42	54.34
	CP3	63.43	56.81
15 mm	CP1	59.81	53.42
	CP2	59.71	55.05
	CP3	56.26	54.22
25 mm	CP1	43.75	41.63
	CP2	41.18	40.40
	CP3	41.43	39.61

According to ASME PCC-2 and ISO Technical Specification 24817, a lower estimate γ_{LCL} of the energy release rate can be obtained assuming a Student distribution (due to the limited space the technical issues about Fracture Mechanics will not be discussed in this paper, see Sahtler et al, 2021) using the following equations:

$$\gamma_{LCL} = \left[\frac{\sum_{i=1}^{n} A(d_i)p_i}{\sum_{i=1}^{n} A(d_i)^2} - t_v\sigma\sqrt{\frac{1}{\sum_{i=1}^{n} A(d_i)^2}}\right]^2 \tag{1}$$

where,

$$A(d_i) = \sqrt{\left\{\frac{0,001}{\frac{(1-\nu^2)}{E_{12}}\left\{\frac{3}{512t^3}{d_i}^4 + \frac{1}{\pi}d_i\right\} + \frac{3}{64G_{13}t}{d_i}^2}\right\}} \tag{2}$$

$$\gamma_{mean} = \left(\frac{\sum_{i=1}^{n} A(d_i)p_i}{\sum_{i=1}^{n} A(d_i)^2}\right)^2 \tag{3}$$

$$\sigma = \sqrt{\frac{\sum_{i=1}^{n}(p_i - A(d_i)\sqrt{\gamma_{mean}})^2}{(n-2)}} \tag{4}$$

$$p_i = A(d_i)\sqrt{\gamma_i} \tag{5}$$

d_i is the hole diameter, p_i is the failure pressure associated to the diameter d_i and t is the composite thickness. Tables 7 and 8 present the lower estimate of

Table 7. Energy release rate obtained without silica.

Hole diameter	CP	0% - Failure Pressure (bar)	$\boldsymbol{\gamma_i}$ (J/m^2)	$\boldsymbol{\gamma_{mean}}$ (J/m^2)	$\boldsymbol{\gamma_{LCL}}$ (J/m^2)
10 mm	CP1	65.86	43.67	57.2	31.03
	CP2	59.42	35.48		
	CP3	63.43	40.42		
15 mm	CP1	59.81	88.01		
	CP2	59.71	87.71		
	CP3	56.26	78.01		
25 mm	CP1	43.75	177.77		
	CP2	41.18	157.29		
	CP3	41.43	158.82		

Table 8. Energy release rate obtained with 1% of silica.

Hole diameter	CP	1% - Failure Pressure (bar)	γ_i (J/m^2)	γ_{mean} (J/m^2)	γ_{LCL} (J/m^2)
10 mm	CP1	58.61	34.53	47.79	24.27
	CP2	54.34	29.65		
	CP3	56.81	32.44		
15 mm	CP1	53.42	70.18		
	CP2	55.05	74.72		
	CP3	54.22	72.30		
25 mm	CP1	41.63	160.36		
	CP2	40.40	151.24		
	CP3	39.61	145.31		

The lower estimate γ_{LCL} of the energy release rate used in projects is 31.03 (J.m^2) without silica and 24.27 (J.m^2) with 1% of silica. A 21.8% reduction, which is acceptable and that can be circumvented by increasing the repair thickness.

3. CONCLUDING REMARKS

The study shows that the introduction of a small fraction of pyrogenic silica (1% in weight) in a commercial adhesive allows enhancing the viscosity, making easier the application in the repair of corrosion defects in pipelines. This is particularly important in the case of through-wall defects in complex position. The adhesive becomes free-flowing and easy to manipulate while being brushed but that sets to a gel when it is allowed to remain at rest. Because of these qualities, this adhesive is less likely to drip from a brush than the adhesive without silica. Besides, the silica does not affect severely the mechanical performance of a composite repair system. The addition of such a low-cost thixotropic thickener makes easier to apply the adhesive layer in composite repair systems maintaining good adhesive properties. This thickener has proved to be remarkably effective in imparting thixotropic properties to coating masses.

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