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Review

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The chemistry insight: epoxy sealant as an alternative remedial operation for well integrity

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Abstract: Epoxy resin is commonly used in the oil and gas industry due to its excellent toughness, low shrinkage, good adhesive strength, and relatively good thermal resistance. It is used for water shutoff, zonal isolation, cementing, enhanced oil recovery, and preventing leakage in wells. This paper reviews the chemistry aspect of using an epoxy resin system as a sealant to prevent well leakage and it offers insights into the chemistry of the epoxy resin system, as applied in previous studies. The paper also unveils the reasons for the application of this system from the chemistry perspective, allowing this aspect to be better understood. Success in the investigated cases depended on the formulation design. The epoxide and hydroxyl functional groups have been found to contribute substantially to the excellent performance of the sealant system. Furthermore, the amine curing agent triggers the abrupt reaction of the oxirane ring to stabilise when the cured sealant is perfectly applied. Based on the findings, it is suggested that other types of epoxies, namely epoxidised oils, require further study. Finally, in terms of safety and

sustainable energy, it is suggested that more curing agent and diluent studies are undertaken.

Keywords: epoxy; oxirane ring; remedial operation; well integrity; well leakage.

1 Introduction

Well leakage is a major issue in the oil and gas industry, with large amounts of production lost due to leakage problems. Many wells are abandoned or shut off as remedial operations involve considerable lengths of time and costs (Dusseault et al. 2014; Vicente Perez et al. 2017). Various environmental and operational factors cause these detrimental leaks (Li et al. 2016). Environmental factors like erosion and corrosion occur at the borehole during production. In addition, the high-temperature and high-pressure conditions at the subsea level cause the system to become worn (Jones et al. 2014). During operation, the cement eventually cracks and causes annular leaks via the leakage pathway (Genedy et al. 2014). Leakage may lead to a potentially catastrophic blowout, the impact of which might be disastrous to humans and the environment. Thus, leakage has to be resolved effectively.

Leak repair can be divided into mechanical solutions and chemical solutions. However, chemical solutions are preferable due to the lower cost and ease of such operations. Various methods can be applied as chemical solutions for remedial operations. Basically, chemical solutions provide a sealant that is used as a repair material. The use of several types of polymer-based sealant has been reported in such applications: epoxy, polyacrylamide co-polymerised tert-butyl acrylate (PatBA), methyl methacrylate (MMA), and crosslinked nitrile butadiene rubber latex (XNBRL).

Epoxy resin systems have gained the most attention in research into leak repair. However, most studies have not comprehensively described the chemistry of the epoxy used. The three main advantages of using epoxy as a sealant are its excellent adhesion energy, low shrinkage, and good chemical resistance. One of the vital characteristics of a

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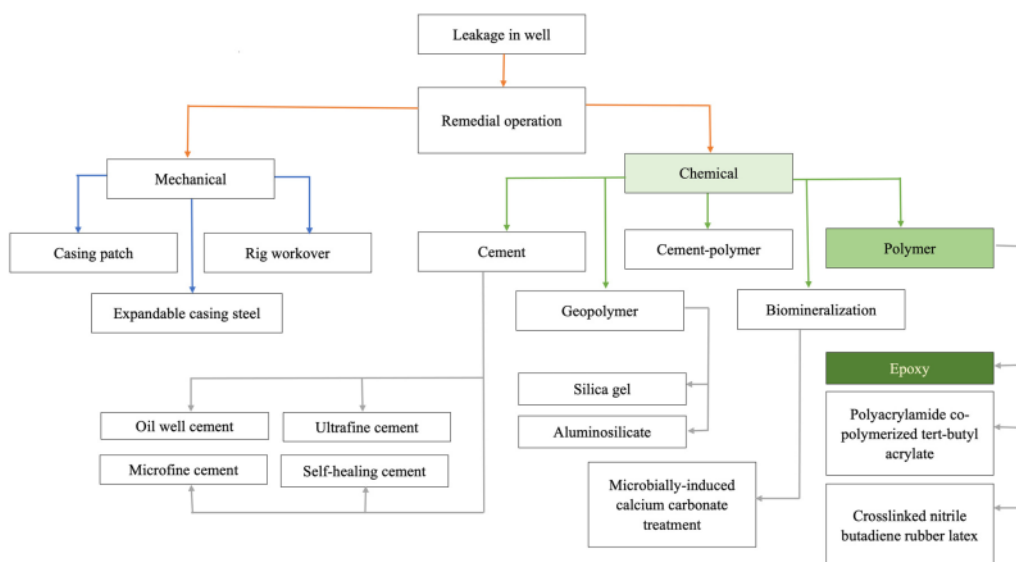


Figure 1: Overview of remedial operation for leakage in well.

sealant is to seal the substrate joints to prevent the passage of water and other substances (Lu et al. 2021). Thus, having excellent adhesion properties is a key characteristic of a sealant. Epoxy is a low-shrink polymer that performs excellently in concealing a leak (Genedy et al. 2017). Having relatively good chemical resistance properties is another factor making epoxy a good choice as a sealant in well leakage applications (Elyas et al. 2018).

This paper will review sealant studies in relation to their chemistry insights. For instance, Yousuf et al. (2021) reviewed well leakage remediation from the cement failure perspective and outlined other types of available sealant. Wu et al. (2020) wrote about sealant technologies for leak repair in relation to cement and generalised other types of sealants. The current paper highlights the chemistry relationship between the epoxy system and leak repair applications in the oil and gas industry. An overview is also provided of the remedial leak repair methods that have been undertaken to restore well integrity. The focus is on the epoxy resin system, its chemistry, and its properties with regard to its application. Leaks, which have a major impact on safety and finances, can be resolved effectively using a basic epoxy system. An overview of the layout of well leakage remedial operations is described; this is represented in Figure 1.

1.1 Leakage in wells

A well is comprised of the wellhead, wellbore, and reservoir. A hydraulic system is used throughout the well

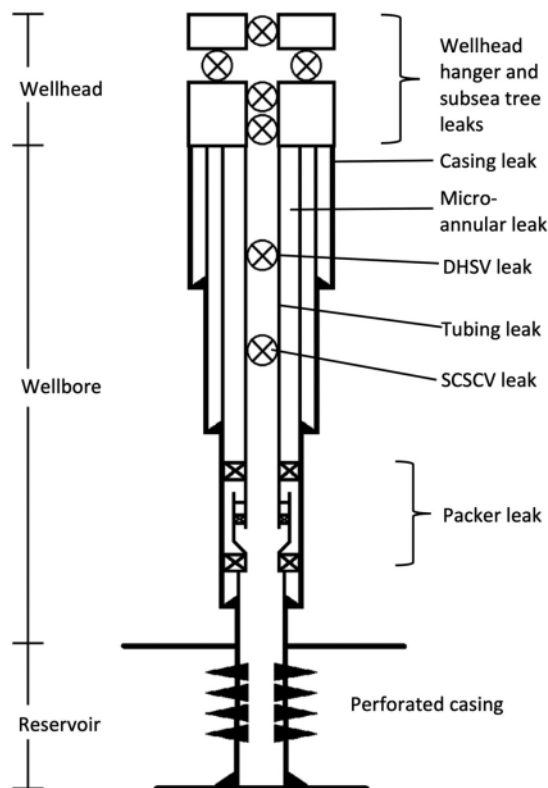


Figure 2: Schematic diagram of types of well leakage.

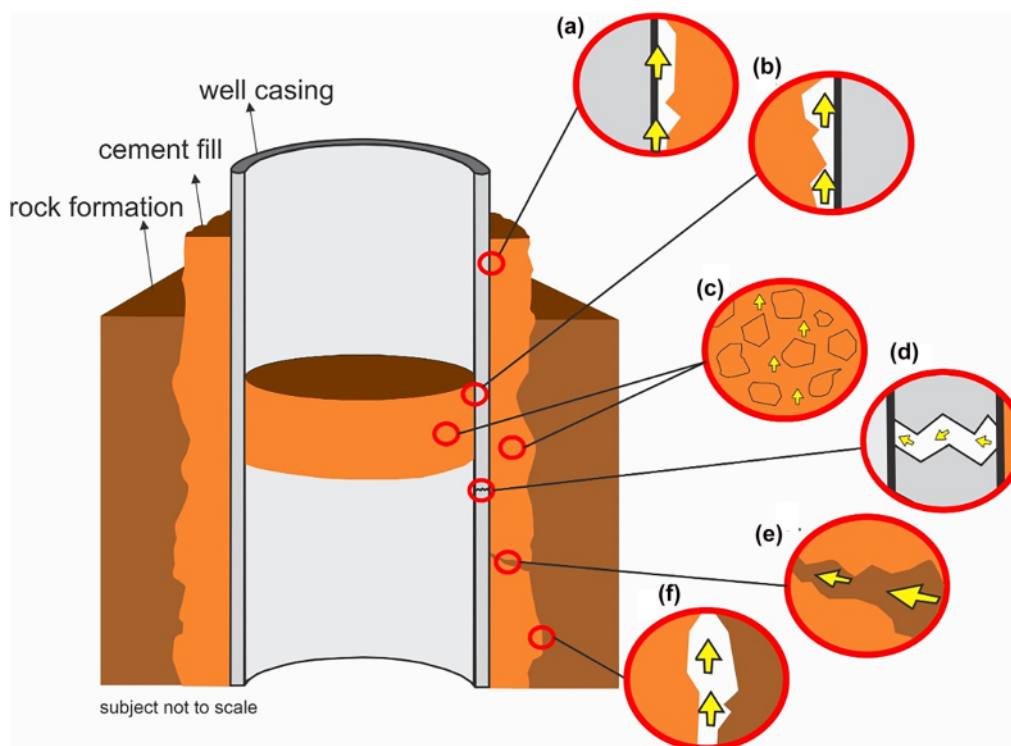


Figure 3: Leaks in plug-in well's schematic diagram.

(a) casing-cement interface, (b) casing-cement plug-in, (c) cement annulus, (d) casing-casing annulus, (e) fractured cement, (f) cement-formation interface (Randhol et al. 2009).

system. Wellhead hanger and subsea tree leaks occur in the wellhead segment. Micro-annular leaks, casing leaks and tubular leaks can be detected in the wellbore. Leakages involving valves are subsurface-controlled subsurface safety valve (SCSSV) leaks and downhole safety valve (DHSV) leaks. In the reservoir segment, the leaks that normally occur are packer leaks, umbilical leaks, subsea connector leaks and pipeline leaks (Rodrigues et al. 2005). These leaks are illustrated in Figure 2.

Figure 3 illustrates the leakage pathways that occur in plug-in wells. Interfaces between rock formations and the cement, the cement and the well casing, and the well casing and the plug-in material could become leak pathways (Randhol et al. 2009). The most critical leakage pathways in the wellbore are the rock-cement and steel-cement interfaces (Genedy et al. 2017). Long exposure to corrosive gases and poor cement formation lead to corrosion. This subsequently causes the casing leaks that are most frequently reported. Other plausible reasons for such leaks are incorrect casing grade options, defective casing due to the unsuitable use of torque, and worn drill pipes (Jasper 2012).

1.2 Remedial operations for leaks

Remedial operations for leaks are divided into mechanical solutions and chemical solutions. The former use mechanical equipment to close the opening caused by fissures or cracks (Jasper 2012). Meanwhile, chemical solutions use chemicals as repair materials, which function as sealants to seal off leakage pathways (Genedy et al. 2019). Mechanical and chemical solutions have advantages and disadvantages and each may be the

Table 1: Advantages and disadvantages of using mechanical and chemical solutions.

Solution	Mechanical	Chemical
Advantage	No chemical hazard No leak reoccurrence	Cost effective Effective installation Accessible
Disadvantage	Costly Ineffective installation Reduction in diameter of borehole	Chemically hazard Possibility of leak reoccurrence

appropriate method to deploy. Table 1 shows the advantages and disadvantages of using mechanical and chemical solutions for leak repair.

1.2.1 Mechanical solutions

Various methods have been employed as mechanical solutions to the issue of sealing leakages. The casing patch is an old method of repairing leaks, specifically casing leaks (Neely 1985). However, the casing patch is not an effective remedy for leaks (Todd et al. 2018). An expandable steel casing is another tool used to mechanically repair a leak; this is an improved version of the casing patch (Saltel et al. 2015). The disadvantage of using an expandable steel casing is that it decreases the internal diameter of the borehole (Alanqari et al. 2019). Using a remotely operated vehicle (ROV) with a rig workover is another example of a mechanical method of repairing leaks (Xu et al. 2020a). However, mechanical solutions involve considerable lengths of time and costs. The efficiency of the remedial work is sometimes compromised. Consequently, other complications might occur during the repair interval and/or production (Alanqari et al. 2019).

1.2.2 Chemical solutions

Cement squeeze and polymer-based sealants are common repair materials that have been studied and applied to restore well integrity. Ostensibly, reports indicate that several wells have successfully used cement and polymer as repair materials for remedial jobs (Rodrigues et al. 2005; Slater et al. 2001). Oil well cement has evolved to address the leakage issue in wellbores. Not limited to using conventional Portland cement, cement squeeze technology has advanced to the use of glass-beads cement, elastic and foam cement, cement-resin composite, and the latest invention, smart cement (Jimenez et al. 2016). In addition, gelant and biomineralisation are other chemical solutions that are more environmentally friendly selections as leak repair options (Kruszewski et al. 2021; Phillips et al. 2015, 2016; Wiese et al. 2019).

Cement squeeze uses cement slurry as the repair material. During its application, the cement is pumped into the leakage pathway and later squeezed to make it into filtrate cake. Thus, it can seal off the existing repair leak. The types of cement used are Portland cement, microfine cement, ultrafine cement, self-healing cement (SHC), and hybrid cement-resin (Abid et al. 2020; Arif Khattak et al. 2015; Baldissera et al. 2017; Cavanagh et al. 2007; Yousuf et al. 2021). Two advantages of using cement are its low cost and technical feasibility (Alkhamis et al. 2020). It is used

for primary cementing tasks to fill gaps between channels in the borehole, for zonal isolation, and to repair casing leaks due to corrosion (Shryock and Slagle 1968). It is suitable for sealing leaks as it can fill voids by forming a solid matrix in the leak area and has relatively good compressive strength (Wiese et al. 2019). Based on research, cement squeeze is an effective short-term solution for large leaks, specifically those more than 50 μm in size (Fernandez et al. 2019). However, cement squeeze has its drawbacks. Its large particle size limits its potential to penetrate leaks of smaller size (Wiese et al. 2019). Cement has a relatively high injectivity rate due to its non-compliance viscosity. Viscosity modifications of cement slurry are highly limited due to its particle size and rheological behaviour (Alanqari et al. 2019). For long-term solutions, cement squeeze has not been regarded as a suitable option for leak repair. It is recommended for use only when the well cannot be slimmed (Al-Dossary et al. 2017).

Geopolymer is another chemical used for leak repair. Generally, geopolymer is composed of inorganic-based materials, mainly aluminosilicates. Initially, the dissolution process of silicates takes place when an alkali activator invades the material precursor that is to be depolymerised. Once depolymerised, oligomers in a liquid state (gel), which are shorter chains of silicates, are transported to penetrate the aperture. The oligomers rearrange their molecules to reform a covalent bond. Finally, a solid geopolymer is formed (Vrålstad et al. 2019). The silica gel and alkali-activated alumina silicate possess high strength, flexural strength, and good thermal resistance. They behave as non-Newtonian fluids to penetrate the leak area (Wiese et al. 2019). Furthermore, geopolymer sealant is non-toxic and economically feasible (Kruszewski et al. 2021). The shortcomings of this type of sealant are that the solidification process is fast and difficult to control, and the sealant shrinks due to the syneresis effect (Vrålstad et al. 2019).

Another interesting remedial method using a biochemical solution is a biofilm with microbially induced calcium carbonate precipitation (MICP) (Kirkland et al. 2019; Phillips et al. 2015, 2016). MICP is produced by the inoculation of *Sporosarcina pasteurii*, which produces a urease enzyme in the hydraulically fractured core. The enzyme catalyses the hydrolysis process of urea. The hydrolysis process forms carbonate and ammonium ions. Subsequently, carbonate ions react with calcium in favourable alkaline conditions to form a precipitate of calcium carbonate (Phillips et al. 2015). Figure 4 illustrates an image of MICP treatment in CO_2 sequestration. Leakage from the functional aquifer can be prevented with a biomineral seal. This reported

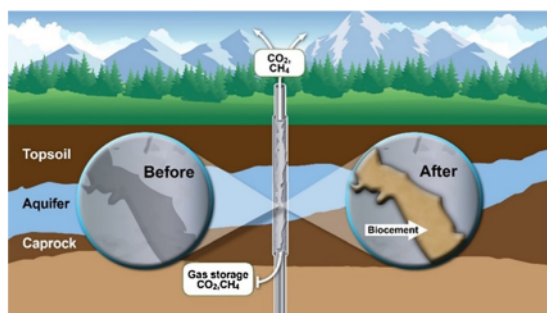


Figure 4: An illustration of MICP formation in a wellbore cement defect (Kirkland et al. 2019).

sealant is only available for limiting the seepage of CO_2 from a storage reservoir. Inoculation may be attempted at a reservoir in the presence of the mineral content of the rock. The success of the MICP treatment depends on the calcium ion concentration, pH conditions, and availability of nucleation sites (Kirkland et al. 2019). In other words, the application is suitable only in very limited conditions and requires specific resources.

2 Polymer-based sealant for leak repair

Polymers are applied in many industries. Some polymers have strong adhesion properties, excellent compressive strength, low shrinkage properties, and better processability, making them useful sealing materials (Saba and

Table 2: Type of sealant, chemical based and their advantages and disadvantages in application.

Type of sealant	Chemical based	Advantages	Disadvantages	References
Cement	Oil well cement	Relatively good compressive strength Feasible (application and economically) Suitable for larger leak size (more than 100 μm) Low cost	Non-flexible viscosity Ineffective for leak size smaller than 100 μm Low adhesion energy Shrinkage	(Alkhamis et al. 2020; Genedy et al. 2014; Wu et al. 2020)
Heavy-weight cement-resin	Hybrid epoxy and cement	Relatively good compressive strength Relatively good chemical resistance Lightweight	Penetration limited to larger aperture size	(Yami et al. 2017)
Geopolymer	Aluminosilicate/silica gel	Good compressive strength Good flexural strength Highly thermal resistance Highly ductile	Dependency on alkalinity	(Kruszewski et al. 2021; Wiese et al. 2019)
Biominalisation	MICP	Environmentally friendly	Dependency on concentration of calcium ion and inoculation of the microbes	(Kirkland et al. 2019; Phillips et al. 2013, 2015, 2016)
Polymer	Epoxy Novolac epoxy PAAtBA XNBRL MMA	Adjustable viscosity (non-Newtonian fluid) Excellent compressive strength Good adhesion energy Flexible Chemical resistance Good thermal resistance	Creep Hazardous	(Alkhamis et al. 2019; Alsaihati et al. 2017; Elyas et al. 2018; Jones et al. 2014; Vicente Perez et al. 2017) (Genedy et al. 2014, 2017) (Shamlooh et al. 2020; Vasquez et al. 2005; Vasquez and Curtice 2015; Vasquez and Santin 2015) (Xu et al. 2020a,b) (Genedy et al. 2019)

Jawaid 2018). In addition, some polymers can withstand conditions of high temperature and high pressure (De Leon et al. 2016). Viscoelasticity is a distinguishable property of polymers. It imparts flexible processability to suit the application requirements. A polymer solution or polymer melt behaves according to non-Newtonian flow and depends on the shear rate (Fried 2014). A polymer can be blended with other materials, such as fillers, to improve its properties. These properties are the advantages of using polymers as sealants to restore well integrity.

Reports have indicated the use of different polymers for leak repairs in well remedial operations. For instance, PATBA has been studied as a thermally activated sealant for this application. It has been reported as a shallow-penetration conformance sealant (SPCS). The sealant is workable at a temperature working range of 4 °C–204 °C. It is injected into the targeted leak area. Once activated by temperature, a three-dimensional structure (3-D) is formed in the area (Vasquez and Curtice 2015). Another study reported successfully using PATBA to repair leakage pathways caused by casing leaks and poor zonal isolation behind the casing. A low viscous solution of PATBA was injected into the leak formation. Hydrogel was formed by thermal activation to seal off the fractures and channel. It managed to withstand pressure at a differential pressure of 2600 psi during the application (Vasquez and Santin 2015).

Pressure activated sealant (PAS) was introduced in the 1990s to seal micro-defect leaks and production equipment. A novel multifunctional sealant was invented based on the PAS principle. This invention was to further improve the traditional PAS, which has been limited to plugging activities. The improved version of PAS introduced an XNBRL incorporated with solid materials—sepiolite fibre and mussel powder—to produce PAS composites (Xu et al. 2020a, b). The researchers inferred the *in situ* sealing mechanism and enhanced its process so it could be used for leaks with larger dimensions. The study combined the liquid and solid phases of the materials to better retain the capacity to cope with larger leaks. This also ensured that the latex could penetrate long leakage paths before it effectively coalesced and agglomerated in the leakage pathway. A mechanism involving the liquid-solid transition phase was introduced in this study. It was hypothesised that the factors that may affect the plugging performance of the composite sealant were the types of solid materials, its microstructure, the geometries of the cured sealant's pores and differential pressure during the sealant's deployment. It was proven that the solid materials used in the dispersed phase also played an important

role in repairing larger leaks, based on the aforementioned theory (Xu et al. 2020a). An extensive study of PAS using XNBRL claimed that too few studies had been conducted on the sealing mechanism, sealant design and evaluation of PAS (Xu et al. 2020b).

The potential of MMA and nanomodified methyl methacrylate (NM-MMA) as repair materials were studied by Genedy et al. (2019). A performance comparison was made between four types of sealants to evaluate their penetration into 30 µm cracks. NM-MMA is a mixture of MMA incorporated with 0.5% alumina nanoparticles (ANPs). It was proven experimentally that NM-MMA is much tougher and more ductile, and it has better bond strength than microfine cement. Based on a microscopic study, it has been highlighted that MMA and NM-MMA managed to penetrate uniformly into the microcrack, whereas microfine cement and novolac epoxy could not. In addition, NM-MMA has improved creep recovery, reduced creep compliance, and an increased degree of crystallisation. A higher degree of crystallisation indicates increases in the ductility and toughness of the sealant. However, neither the use of MMA nor NM-MMA has been reported in field applications. Exceptionally, the use of MMA has been reported in terms of its incorporation into cement by adding carbon nanotubes (CNTs). This report can be accessed in the article by Mohamadian et al. (2020).

To date, the epoxy resin system has been the most studied and applied polymer-based sealant for leak repair applications to restore well integrity. With its advantages over other reported polymer-based sealants, epoxy is potentially better for such applications. An epoxy resin system has better processability as it uses at least two components, epoxy resin and a curing agent (Elyas et al. 2018; Genedy et al. 2014, 2017). The adjustable viscosity uses a reactive diluent, which is highly related to the injectivity during the sealant's deployment and flowability in the leak area (Alkhamis et al. 2020; Alsaihati et al. 2017; Elyas et al. 2018; Genedy et al. 2017; Jones et al. 2014). The selection of the curing agent to harden the resin and the ratio determination are highly important as they allow sufficient time for the gelation and curing processes during application (Alsaihati et al. 2017; Licari and Swanson 2011). An epoxy resin system possesses the optimum sealing properties, such as excellent adhesion energy, good compressive strength and relatively good chemical resistance (Pham and Marks 2005). Those factors make it a prime option. Table 3 shows the factors involved in selecting an epoxy resin system as a sealant, based on its properties.

Table 3: Factors influencing the selection of epoxy resin system.

Factors	Reason	References
Density	Able to penetrate into the leakage pathway	(Alsaihati et al. 2017; Jones et al. 2014)
Gel/thickening time	Provide ample time for the fluid to properly engage into leaks	(Elyas et al. 2018; Jones et al. 2014)
Withstand relatively high pressure	To adapt with the condition of the well during application	(Alsaihati et al. 2017; Jones et al. 2014)
Compatibility with other fluids in application such as brine, hydrocarbons and water	No reaction to negate the application work	(Jones et al. 2014)
Adhesion	Bonding strength with casing and function as sealing material	(Alsaihati et al. 2017)
Thermal resistance	High T_g will ensure the range of application and reliability in long term application	(Alkhamis et al. 2019; Alsaihati et al. 2017)
Compressive strength, blocking and injectivity	Functionality as sealing material	(Alanqari et al. 2019; Elyas et al. 2018)
Corrosion resistance	Functionality as sealing material	(Elyas et al. 2018)
Viscosity	Flexibility to modify to suit the requirement and correlate with high injectivity rate	(Alkhamis et al. 2019; Elyas et al. 2018)

3 Epoxy resin system

Generally, epoxy resin is a thermoset polymer. Also known as epoxide, it is classified under the wider family of ether. The oxirane ring or ethylene oxide group contained in the chemical structure instils certain properties that make it useable for high-end applications, such as those in high temperature and high-pressure conditions. It consists of a three-membered ring that makes the group reactive, unlike other types of ether. The three main types of epoxies are cycloaliphatic epoxy resins, epoxidised oils, and glycidated resins (Massingill and Bauer 2000). Commonly used in well leakage applications is the epoxy-type glycidated resin bisphenol-A diglycidyl ether (BADGE or DGEBA). This comprises two epoxides, two open-ring ethers, and two phenyls, as pictured in Figure 5.

BADGE can be synthesised by reacting bisphenol-A with epichlorohydrin in basic or acidic conditions. The oxirane ring is naturally reactive due to its structural configuration, which strains the oxygen bond to the carbon-carbon. Therefore, it becomes reactive so as to stabilise the chemical structure (Fessenden et al. 1998). Having such chemistry, the epoxy resin demonstrates characteristics, such as resistance to corrosion and chemicals, as well as low shrinkage properties. In addition, it possesses outstanding adhesion energy and good electrical properties (Fried 2014). From a

financial perspective, the application of an epoxy resin system offers better cost savings than other methods and resin systems (Singh et al. 2019).

Alanqari et al. (2019) chose BADGE for their research works. Two formulations utilise BADGE as the resin. The researchers investigated the thermal properties of the epoxy resin system. Variations were observed in the glass transition temperatures (T_g). The T_g of epoxy hardened with aliphatic amine was 120.5 °C; when hardened with aromatic amine, the T_g was 147.5 °C. The formulation between the two formulations was somewhat vague. Another finding by Alkhamis et al. (2019) used BADGE as the base resin, whereby the viscosity of the neat resin was 11,000–14,000 cp. Theoretically, the molecular weight determines a polymer's physical properties, such as its viscosity and brittleness (Garcia and Soares 2003). The average molecular weight of a polymer is presented by the n value, which is the repeating unit of the monomer. If the n value is higher, the viscosity may also be higher.

Correspondingly, epoxy equivalent weight (EEW) is an important parameter to take into account before formulating the epoxy resin system. The EEW is the weight of resin per epoxy group containing the polymer molecules. The EEW determines the stoichiometric ratio between the epoxy resin and the curing agent, which affects the physicochemical properties of the system. Each hydroxyl ion

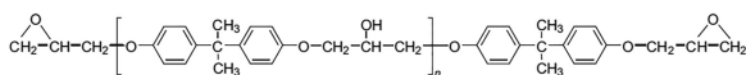


Figure 5: Chemical structure of BADGE polymer with n is the repeating unit (Gibson 2017).

Table 4: Viscosity and EEW of epoxy resins (Licari and Swanson 2011).

Epoxy resin type	Examples: resin/ supplier	Viscosity @25 °C (mPa.s)	Epoxy equivalent weight (gm)
High purity diglycidyl ether of bisphenol-A	EPON [®] 825/Hexion ^a	5000–6500	175–180
	D.E.R [™] 332/Dow Chemical Co.	4000–6000	171–175
Diglycidyl ether of bisphenol-A (standard, undiluted resin, general purpose)	EPON [®] 828/Hexion ^a	11,000–15,000	185–192
	D.E.R [™] 331/Dow Chemical Co.	11,000–14,000	182–192
Reaction product of epichlorohydrin and bisphenol-A to provide low viscosity without diluent	D.E.R [™] 330/Dow Chemical Co.	7000–10,000	176–185
	D.E.R [™] 354/Dow Chemical Co.	3400–4200	167–174
Diglycidyl ether of bisphenol-F	EPON [®] 862/Hexion ^a	2500–4500	165–173
	D.E.R [™] 351/Dow Chemical Co.	4500–6500	169–181
Reaction product of epichlorohydrin with bisphenol-F and bisphenol-A (achieves performance properties of bisphenol-A combined with low viscosity of bisphenol-F)	EPON [®] 58005/ Hexion ^a	3000–8000	325–375
Elastomer-modified epoxy functional adduct formed by reacting DGEBA with 40% carboxyl-terminated butadiene-acrylonitrile elastomer			

^aEPON is a registered tradename of Hexion Specialty Chemicals. EPON epoxy resins were originally developed by Shell Chemical Co.

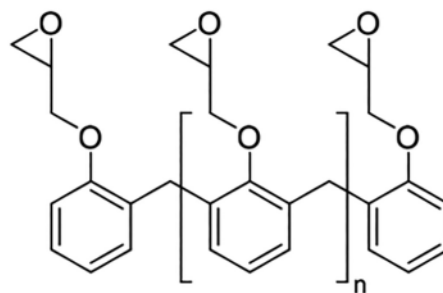
Table 5: Advantages and disadvantages of epoxy resin system as sealant to repair leak.

Advantages	Disadvantages
Excellent adhesion energy	Not able to withstand extreme temperature
Low curing volume shrinkage	Release hazardous fume during processing and curing
Good chemical resistance	Not compatible with water
Good compressive strength	
Adjustable viscosity	
Ease of processability	
Cost-effective	

from the epoxy group reacts with one hydrogen atom from the amine group of the hardener (Garcia and Soares 2003). Therefore, the EEW should be known before formulating the amount of amine hardener that will be reacted with the epoxy resin. The out-turn of the non-stoichiometric reaction is that the resin system could be either brittle or not cured. Brittleness and an uncured system occur due to excess amine hardener and insufficient amine hardener, respectively (Licari and Swanson 2011). Essentially, the supplier of epoxy resin provides the EEW for the formulators. For instance, Table 4 shows the EEW for various commercial epoxy resins used for adhesive application and in relation to the viscosity. Based on the table, EEW is proportionately related to viscosity. Table 5 summarises the advantages and disadvantages of using epoxy resin as a sealant.

BADGE has limited usage in higher applications involving temperatures of more than 147.5°C (Alanqari et al. 2019). This drawback means that novolac epoxy offers a potential back-up solution for its negation. Inversely,

most epoxy resin is very low in flexibility. The compact epoxide structure causes the brittleness (Licari and Swanson 2011). Less hazardous than BADGE, novolac epoxy resin (Figure 6) is another option as a glycidyl ether type of epoxy. This combination of phenol resin and bisphenol-A performs better. Although more expensive

**Figure 6:** Polymeric chemical structure of novolac epoxy resin (Unthank et al. 2019).

than BADGE, it is worth using in applications. Novolac epoxy imparts better chemical resistance, thermal resistivity, and toughness, and its application has proven successful in flame retardant products. This improvement in properties is caused by the functionality of the epoxides group, which can be from two to six (Pham and Marks 2005).

Two studies have been conducted to compare novolac epoxy with other sealing materials (Genedy et al. 2014, 2017). An initial study by Genedy et al. (2014) used novolac epoxy and polysulphide siloxane epoxy incorporated with nanoparticles. Novolac epoxy reinforced with nano-silica and nano-alumina was found to impart better bond strength to cement and steel when compared with micro-fine cement. The nanocomposites are also injectable into the leak area. With polysulphide siloxane epoxy, nano-silica and nano-alumina adversely affect the bond strength of the nanocomposites. However, polysulphide siloxane epoxy incorporated with CNT showed an improvement in bond strength compared with microfine cement but no effect was identified in novolac epoxy incorporated with CNT.

Subsequently, Genedy et al. (2017) studied the bond strength performance with shale by comparing neat novolac epoxy, novolac epoxy incorporating various percentages of ANPs, and microfine cement. The microscopic sealing ability was also compared. From the experimental study, it was suggested that novolac epoxy incorporated with 0.5% ANPs is a good repair material for shale micro-annulus. When compared with other samples, it showed the best improvements in bond strength and flowability. Moreover, it was able to seal the gap between the shale-cement interface.

3.1 Curing agent (hardener)

A curing agent or hardener is a reagent used to crosslink with the main polymer to form a three-dimensional network structure of cured resin. It is an oligomer that can be obtained from numerous types of functional groups. The functional group determines the final properties as it participates in the curing reaction. Various curing agents can be used for the epoxy curing reaction, namely amine hardener (Alkhamis et al. 2019; Alsaihati et al. 2017), acid anhydride hardener (Barabanova et al. 2019), phenol (Shibata and Ohkita 2017), and mercaptan hardeners (Eom et al. 2013).

To thermoset a resin system, the resin has to be hardened to prevent a reversible reaction of the polymerised polymer. In this case, epoxy performs this role. The

reaction with amine hardener results in the full reaction of both reactants with no creation of by-products (Gibson 2017; Hodd 1989). The addition of amine hardener to the epoxy resin system is illustrated in Figure 7. Initially, an active hydrogen atom from a primary amine reacts with a hydroxyl group (–OH) from one of the oxirane rings, causing the ring to open. Secondly, another active hydrogen from the amine group reacts with the subsequent hydroxyl from the oxirane ring of the epoxy resin. Theoretically, an active hydrogen reacts with an oxirane ring. Finally, once the amine has fully reacted with the epoxy, the etherification process or crosslinking occurs. This reaction is highly exothermic, specifically when epoxy is crosslinked with the amine type of hardener. Stoichiometrically, an active hydrogen atom reacts with one oxirane ring of the epoxy resin, but, in practice, there will be a deviation. This might be influenced by the epoxy-hydroxyl reaction to the epoxy secondary amine reaction and in the presence of a catalyst (Hodd 1989). Observations of properties and characterisation are important to elucidate the best formulation of the epoxy resin system.

Hypothetically, aliphatic amine hardener tends to react faster than aromatic amine hardener, resulting in a speedy curing reaction with the aliphatic type. Aliphatic amine with less steric hindrance likely tends to react with the hydroxyl group of the epoxy. Meanwhile, the aromatic amine hardener, with its bulky structure, is hindered from reacting quickly with the hydroxyl group (Alanqari et al. 2019; Alkhamis et al. 2019). Experiments have proven that the amine functional group accelerates the curing reaction of epoxy resin due to the hydroxyl group in the reaction matrix. As the additional reaction is favourable in the

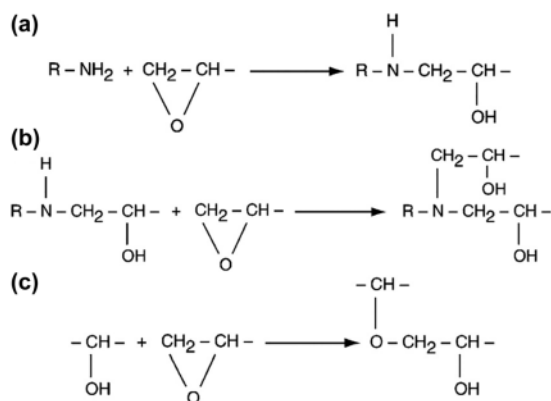


Figure 7: Stages of curing chemical reaction of epoxy resin and amine group hardener from (a) primary amine $-NH_2$, (b) secondary amine $-NH$, and (c) etherification process (Ramsdale-Capper and Foreman 2018).

epoxy curing reaction, the amine functional group is liable to attack the hydroxyl group for the ring opening. Therefore, the cured epoxy is thermoset and chemically stable (Massingill and Bauer 2000).

The curing process of the epoxy resin system involves a conversion from a liquid phase to a solid phase. As the reaction proceeds, the liquid state encounters an increase in viscosity until the gelation of the system occurs. At the time of gelation, the rubbery state of the polymer becomes a glassy state. Subsequently, the curing process is completed once the amine hardener has fully reacted. Nonetheless, the curing process of the epoxy resin system is a complex reaction. It can be evaluated quantitatively by characterising the heat released, as well as the changes in viscosity and specific volume. Experiments have proven that the cured epoxy volume shrinks disproportionately with the degree of conversion (Li et al. 2012). However, the mechanical properties, such as flexural strength, impact resistance, and flexural modulus improve (Hodd 1989).

Corresponding to the EEW, the amine hydrogen equivalent weight (AHEW) is a substantial parameter for determining the stoichiometric ratio of the epoxy resin system. The AHEW refers to the weight of amine containing an active hydrogen atom. The AHEW should be provided by the supplier or it can be determined by titration (Garcia et al. 2007). From known EEW and AHEW values, the amount of amine hardener needed (A_c) can be determined, as shown in Eq. (1).

$$A_c = \frac{\text{AHEW}}{\text{EEW}} \times 100 \quad (1)$$

A study on the epoxy resin system was conducted to repair a leak that occurred at the external casing packer (Jones et al. 2014). This study empirically compared the epoxy resin system with Portland cement in terms of its application performance. Two samples of the epoxy resin system were studied. The first sample (R1) was a mix of epoxy resin (proprietary) and diethyltoluenediamine (DETDA), which was the hardener (Figure 8a). The second sample (R2) was a mix of R1 and a multifunctional primary amine (proprietary). It was found that R2, the supplemental base resin, was able to increase the compressive strength of the initial epoxy system (R1). The system managed to withstand pressures as high as 9000 psi in a laboratory simulation. Besides, the added primary amine reduced the viscosity of the R1 system at low temperatures, but exactly which temperature was not stated. Furthermore, it was able to increase the thickening time at higher temperatures. In addition, the system was compatible with other fluids during placement and curing, such as brine, hydrocarbons, and mud.

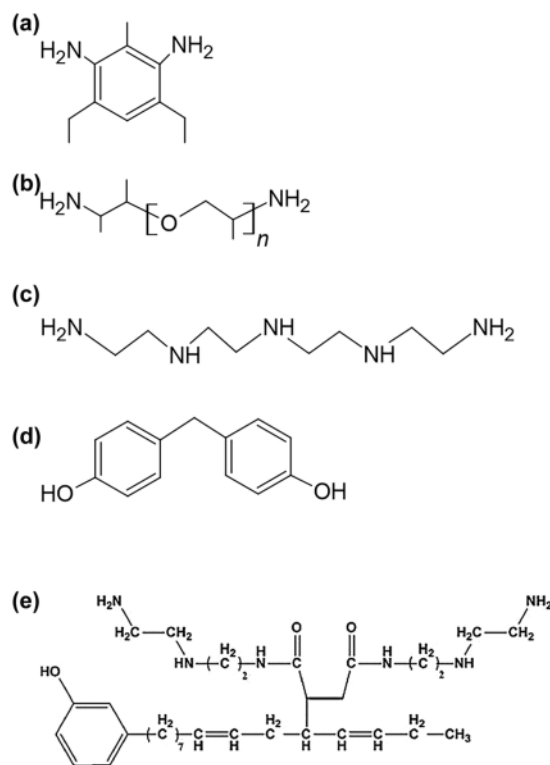


Figure 8: Chemical structures of (a) DETDA, (b) poly(oxypropylene) diamine, (c) tetraethylenepentamine (TEPA), (d) phenol (4,4'-methylene bis- and (e) cardanol-polyamide based (Balgude et al. 2017).

Meanwhile, Genedy et al. (2014) and Alanqari et al. (2019) used a combination of aromatic amine and cyclic aliphatic amine, which was the hardener. They controlled the gel time or thickening time of the resin system with the chosen hardeners. For instance, Alanqari et al. (2019) studied the difference between two formulations using two types of amine hardeners: poly(oxypropylene) diamine (Figure 8b) and a combination of 2-methyl-4,6-bis(methylthio)benzene-1,3-diamine (Figure 9a) and 4-methyl-2,6-bis(methylthio)benzene-1,3-diamine (Figure 9b). Epoxy cured with poly(oxypropylene) diamine has been applied on-field but due to its limited temperature resistance, a combination of aromatic hardeners was used to counter the issue. It has been observed that the T_g of an epoxy system cured with aliphatic amine and aromatic hardeners were 120.5°C and 147.5°C, respectively. This implies that the aromatic amine hardeners used to cure the epoxy resin could potentially be applied at deeper sections of the well.

The vapour pressure of the aromatic amine hardener causes fume release during handling. As an alternative,

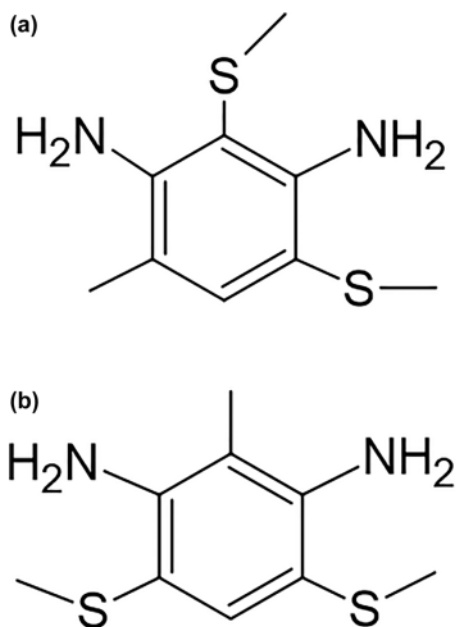


Figure 9: Chemical structures of (a) 2-methyl-4,6-bis(methylthio)benzene-1,3-diamine and (b) 4-methyl-2,6-bis(methylthio)benzene-1,3-diamine.

reactive polyamides (Figure 8e) could be used, as they produce much less fumes than the aromatic amine. They also offer more flexibility and better toughness for the cured epoxy (Hodd 1989).

3.2 Reactive diluent

Neat epoxy resin can be highly viscous, which causes difficulties during handling and application. A diluent is a liquid with low viscosity and, as a free-flowing fluid, can reduce the viscosity of the neat epoxy. There are two types of diluents. A non-reactive diluent is normally an organic solvent or plasticiser that is highly volatile. This type of diluent has adverse effects on the resin properties. It does not participate in the curing reaction but will impede the performance of the mechanical and thermal properties. It reduces the functionality of the epoxy resin, thus decreasing its performance (Pham and Marks 2005).

Preferably, a reactive diluent chemically combines with the epoxy resin while simultaneously improving the resin properties. Even though it is relatively volatile compared to epoxy resin, it is at least far less volatile than a non-reactive diluent. A reactive diluent containing a reactive functionality with low molecular weight epoxy can

improve the process of curing and application (Karami et al. 2017). Furthermore, it enhances the mechanical and thermal properties once the epoxy resin system has been cured (Huo et al. 2018). Thus, the system is toughened by the epoxy matrix (Sahoo et al. 2015). A diluent also helps to improve adhesion and flexibility, in addition to increasing the solvent resistance (Alsaihati et al. 2017).

Viscosity levels of neat epoxy resin, especially BADGE, could reach 11,000–14,000 cps (Alkhamis et al. 2019). This characteristic will cause a high injectivity rate during the application of a sealing material. To reduce its viscosity, a

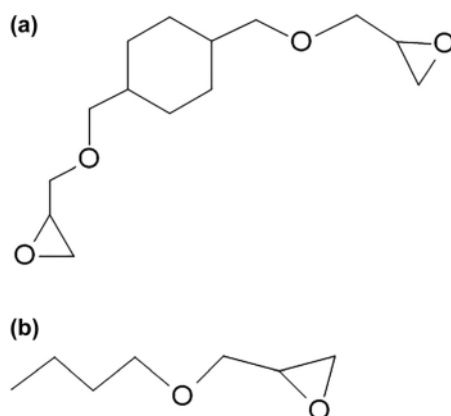


Figure 10: Chemical structures of aliphatic epoxy-based reactive diluent: (a) CHDGE, (b) BGE.

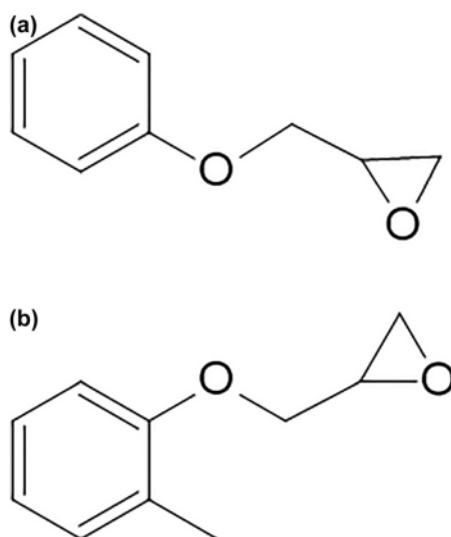


Figure 11: Chemical structures of aromatic epoxy-based reactive diluent: (a) phenyl glycidyl ether, (b) *o*-cresol glycidyl ether.

Table 6: Epoxy resin system as sealant for leak repair.

Researchers	Epoxy resin	Curing agent	Reactive diluent	Other chemicals	Focus of characterisation
Jones et al. (2014)	Diglycidyl ether based (proprietary)	Multifunctional primary amine	R2 (proprietary)	No	Gel time
Genedy et al. (2014)	Polysulfide silane epoxy Novolac- epoxy	DETA Phenol, 4,4'-(1-methylethylidene) bis Tetraethylenepentamine	Not stated	MWCNTs Nano silica Nano clay ANPs	Bond strength
Genedy et al. (2017)	Novolac epoxy	Polyamide hardener	Not stated	ANPs	Interfacial bond strength test
Alsaihati et al. (2017)	Epoxy (proprietary)	DETDA	CHDGE BGE	No	Rheological and mechanical properties
Elyas et al. (2018)	Epoxy – two types (proprietary)	Aromatic amine	Not stated	No	Gel time, injectivity and blocking test
Alanqari et al. (2019)	BADGE	2-methyl-4, 6 bis(methylthio) benzene 1,3-diamine 4-methyl-2, 6 bis(methylthio) benzene 1,3-diamine Polyoxypropylene diamine	CHDGE BGE	No	Thermal analysis (T_g)
Alkhamis et al. (2019)	BADGE	DETDA	CHDGE	No	Rheological property and curing time

DETA, diethyltolueneamine; MWCNTs, multi-wall carbon nanotubes; DETDA, diethyltoluenediamine; CHDGE, cyclohexane dimethanol diglycidyl ether; BGE, butyl glycidyl ether; BADGE, bisphenol A diglycidyl ether.

diluent is essential to control the range required. Cyclohexanedimethanol diglycidyl ether (CHDGE) (Figure 10a) is widely used as a reactive diluent. Other than CHDGE, a possible alternative is butyl glycidyl ether (BGE) (Figure 10b), an aliphatic type of short chain epoxy. BGE effectively reduces the viscosity of BADGE, but hazard and volatility issues make it unfavourable. Phenyl glycidyl ether (Figure 11a) and *o*-cresol diglycidyl ether (Figure 11b) are aromatic ethers that effectively bring down the viscosity, but they are noxious (Pham and Marks 2005).

Alsaihati et al. (2017) prepared two types of epoxy resins that used different diluents. One of the epoxy resin systems used BGE and the other used CHDGE. Both systems were cured with the same hardener, DETDA. However, neither specific observations nor quantitative data were made available to compare the two formulations in findings. The same diluents were used by Alanqari et al. (2019) without further elaborating on a comparison between them. The study intended to find a solution to the current epoxy resin system, which has limited temperature application. Alkhamis et al. (2019) opted to use CHDGE as a diluent because it decreases the viscosity of the epoxy resin with the optimum loss of properties. Table 6 shows the epoxy resin system designed by several researchers as a leak repair solution and the focus of their experimental work.

3.3 Reinforcement fillers

Generally, a filler is a solid particulate incorporated into the polymer matrix to enhance the performance of the polymer, while it also has economical value. Fillers can be selected based on their chemical group, particle sizes, and morphology (Ooi et al. 2013; Satterthwaite et al. 2012; Tanimoto et al. 2013). The filler selection is crucial, especially for its functional aspects and reinforcement. When a filler is added within a reasonable limit, it infuses into the polymer matrix and strengthens the polymer structure. The composite imparts better toughness, increases wear resistance, enhances durability and processability, and seals off pores (Burriss and Sawyer 2006; Derazkola and Simchi 2018; Genedy et al. 2014; Zhu et al. 2021). The adverse effect of a filler depends on the processing and application. Some fillers have a thixotropic effect, which may increase the viscosity of the composite (Julkapli et al. 2015). To prevent this, an optimum dosage must be pre-determined before formulating the product. Numerous fillers, specifically reinforced fillers, will strengthen the lattice structure configuration; however, exceeding the filler dosage will impair its crucial function (Fried 2014).

Today, fillers of nanometre size are widely incorporated into polymer resin. CNT (Lehman et al. 2011), nano silica (Aggarwal et al. 2015), nano clay (Chan et al. 2011),

and nano calcium carbonate (Boyjoo et al. 2014) are examples of nanoparticles that are commonly studied. The major reason for selecting nanoparticle fillers is their particle size. Reducing the filler particles to nanometre size significantly improves the performance of polymer composites. The far larger area of surface interaction improves the processability, structure, and adhesion energy of the matrix system (Akpan et al. 2019). The improper selection of nanoparticles may hamper the system during application and compatibility issues may arise in the system. Additives can be used to improve the compatibility of the nanoparticle with the resin system. Syneresis, or liquid separation, is a common issue encountered when nanoparticles are incorporated into a polymer matrix. Besides, the viscosity tends to increase and agglomeration formation can be expected due to the finer particle size (Julkapli et al. 2015). The increased viscosity is due to the increase of frictional forces caused by the intact large surface area (Licari and Swanson 2011).

Genedy et al. (2014) used four different types of nanoparticles in their work: multi-wall carbon nanotubes (MWCNTs), nano silica, nano clay, and nano alumina. Two epoxy-based resins were studied by incorporating them with these nanoparticles. The properties of the nanocomposites were compared with those of microfine cement. A contrasting effect was observed in the viscosity of both resins with the addition of nano-fillers. The polysulphide siloxane epoxy encountered a viscosity reduction whilst the novolac epoxy experienced an increase in viscosity. Nano-silica and nano-alumina considerably improved the bond strength of the novolac epoxy to a steel surface. MWCNTs quite significantly improved the bond strength of polysulphide siloxane epoxy to steel. However, MWCNTs did not significantly affect the novolac epoxy's bond strength to steel.

4 Epoxy sealant properties

Analyses of the epoxy sealant properties are important for the formulation design, processability, and application. For instance, physical properties, such as density are crucial to ensure the sealant placement can be successfully conducted (Jones et al. 2014). The sealant has to be denser than water to displace the remainder in the fracture. It also has to be less dense than conventional cement to prevent the formation from cracking further. Investigations by Jones et al. (2014) and Alkhamis et al. (2019) obtained epoxy resin system densities of 9.19 lb/gal and 8.76 lb/gal, respectively. Thus, it is vital to employ the sealant within a

range such that it is denser than other substances but less dense than conventional cement.

From the viscosity measurement, the injectivity factor and flow rate can be determined. These characteristics are important to ensure the liquid sealant placement is successful. The sealant liquid must fill the leakage path to excess. Thus, when the liquid flows through the differential pressure at the leak area, the liquid plates out and solidifies to form a solid sealant. Meanwhile, the excess sealant liquid flows out from the system (Rodrigues et al. 2005). The viscosity of neat epoxy resin is adjustable. Therefore, it is preferable that the viscosity measurement is conducted at an early stage to determine the viscosity range. Epoxy resin generally exhibits non-Newtonian flow. It is shear-dependent and temperature-dependent. It is a shear-thinning fluid; when shear stress is high, the viscosity is low. The viscosity increases proportionately with an increase in temperature. Thus, it is suitable for a thermal-activated sealant and the temperature at a downhole, which can activate the working temperature for the system (Jones et al. 2014).

To further investigate the rheological behaviour, the storage modulus and loss modulus can be evaluated. These two properties play important roles in pre-determining the ability of the sealant once it has been applied. The storage modulus and loss modulus anticipate the performance during application. In correlation with shear stress, they provide information about the performance and life span of the liquid sealant (Genedy et al. 2017).

The gel time or thickening time of the epoxy resin system can be controlled to provide ample time for the resin to fully occupy the leakage pathway before curing. It has been stated that the epoxy resin system can withstand the pressure testing requirements and hydraulic fracturing operations. In addition, the system is compatible with other fluids during placement and curing (Alanqari et al. 2019; Elyas et al. 2018; Jones et al. 2014).

To assure the performance of the cured sealant, the compressive strength needs to be evaluated. The sealant must seal off the crack and fracture without becoming detached. It brings the crack surfaces into contact. Thus, the sealant itself must not become detached, regardless of the surrounding pressure and chemicals. Compressive strength tests determine the ability of the cured sealant to remain in place after sealing the leak by simulating the pressure in the well or at the applied area. Besides, the blocking test and injectivity test introduced by Alkhamis et al. (2020) suffice to determine the performance of the sealant from a mechanical perspective. They characterise the sealant need during application and post-application.

Thermal analyses include the T_g , melting point, and crystalline temperature. These evaluations are important to determine the gel time and the point at which the temperature suits the application of the resin system. By determining the T_g , the gel time of the liquid sealant can be predicted. At high-temperature applications, the polymer system tends to gel up. This assists in determining the temperature range to be applied to the resin system. Some polymers will have a distinctive crystalline temperature. This may help to determine the curing temperature of the polymers and the point at which the polymer matrix will be in a totally solid state. A study has obtained the T_g of the epoxy resin system as 149 °C, which imparts relative thermal resistance for remedial application tasks (Alkhamis et al. 2019).

Morphology analysis provides confirmation of the structure formed. The formation of the structure, the particle size, and the shape of the sealant system can further determine the physical properties of the system. This can be achieved using scanning electron microscopy (SEM) and transmission electron microscopy (TEM) (Alanqari et al. 2019; Genedy et al. 2017). Morphological evidence obtained by Genedy et al. (2019) magnified the figure of the penetration of NM-MMA into 30 μm cracks.

5 Potential future work

5.1 Work on other types of epoxies

Although many studies have been undertaken on the epoxy resin system for the oil and gas industry, there remains room for improvement to leak repair applications. BADGE has attracted attention in terms of application. Genedy et al. (2014, 2017) can be credited for exploring other types of glycidated epoxy to obtain better performances. As hazards are a concern, epoxidised oils can potentially be reviewed in order to achieve better safety at work. Even though cured epoxy is stable and not hazardous to the user, during the production of the epoxy resin system a hazard risk arises as BADGE is hazardous to health and the environment (Thermo Fischer Scientific 2021). It is also a carcinogenic substance and causes endocrine disruption to humans (Flint et al. 2012).

Besides the epoxy resin, the curing agent and reactive diluent can also be further explored. Today, other than safety concerns, society is concerned about energy sustainability. The epoxy resin, curing agents and diluents used today generally originate from petroleum resources. Therefore, it is favourable to switch from the use of petroleum resources to natural and renewable resources.

5.2 Implementing industry standards

Test methods for sealant should be based on industry standards, such as those of the American Petroleum Institute (API), the Standard by Norwegian Industry (NORSOK), the American Society of Testing and Materials (ASTM), ISO (The International Organization for Standardization) and the National Association of Corrosion Engineers (NACE). For instance, many standard methods are available for elastomeric assemblies for oil and gas applications (Patel et al. 2019). ASTM D445 (ASTM International 2021) and ISO 3204 (International Organization for Standardization 2020) are available for viscosity measurement of petroleum products. Test methods for the compressive strength of polymers are stipulated in ASTM D695 (ASTM International 2015) and ASTM D638 (ASTM International 2014) for tensile tests. Thus, it is suggested that industry standards are used for the characterisation of sealant that conforms to the quality requirements.

5.3 Testing for the life span of the sealant

To the best knowledge of the author, no comprehensive report has been issued on the life span of a cured epoxy sealant that has been successfully applied in the field. Studying the life span of the sealant would make a major contribution to planning and future work. Creep, a property of a polymer, occurs due to pressure and time. Therefore, creep behaviour can predict the life span of a solid sealant using the acceleration method (Martins et al. 2011). This provides information about the ageing time of the sealant from a simulation test at the area of application. Dynamic mechanical analysis provides data on the storage modulus and loss modulus of the cured sealant. The ageing time can be properly determined. With the combination of these characteristics and creep behaviour, better predictions of life span can be made.

6 Conclusions

The epoxy resin system has gained considerable attention as a sealing material for well integrity remedial operations. The chemistry aspect of the system must be studied and reviewed to ensure the superior performance of the system. Notably, the proper selection of the epoxy type, curing agent, and diluent play important roles in determining the properties of the required sealant. Knowing the chemistry and its relationship with performance will allow room for improvement in terms of safety concerns or sustainable

resources. This may also allow better knowledge to be obtained. Research on the replacement of commercial epoxies with renewable resources, implementing industry standards for sealant characterisation and application, and determining the life span of the sealant may allow sealant conformance to be introduced.

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