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Research article

Deterioration behavior of aged magnetorheological elastomer under harsh marine environment

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Abstract. Silicone rubber magnetorheological elastomers (SR-MREs) are increasingly recognized for their resilience in marine conditions, offering prolonged service life and durability. This study evaluates the one-month durability of silicone rubber magnetorheological elastomers (SR-MREs) under seawater conditions. Results revealed a 6% reduction in hardness and an 8% decrease in Young's modulus compared to unimmersed samples. Morphological and attenuated total reflectance-Fourier transform infrared spectroscopy (ATR-FTIR) analyses supported these findings, revealing surface defects and chemical bonding changes. The immersed SR-MRE displayed a notable 250% increase in elongation at break, highlighting enhanced elasticity. Rheological properties revealed complex mechanical behavior, with an initial increase in storage modulus from 0.25 to 0.38 MPa in the presence of a magnetic field, followed by a gradual decrease to 0.15 MPa at 0 A and 0.52 MPa at 5 A with strain. Additionally, this study proposes an illustrative mechanism to elucidate the relationship between seawater elements and SR-MRE behavior, enhancing our understanding of its mechanical properties and degradation in marine environments, thus highlighting SR-MRE's potential as a durable material compared to traditional rubber composites.

Keywords: *degradation, durability, magnetorheological elastomer, microcrack, rheological properties, seawater immersion*

1. Introduction

Throughout maritime history, materials utilized in marine applications have continuously evolved to withstand the severe conditions of harsh seawater environments. The South Sea, known for its severe atmospheric conditions characterized by high temperature, humidity, and salinity, referred to as the "three highs" environment, significantly impacts initial material properties [1]. While metals such as high-carbon steel and iron metal were commonly used in marine construction, they are susceptible to rust, leading to structural damage and reduced lifespan. The presence of microcracks within rust layers compromises protective capabilities such as coating [2]. Specialized coatings offered protection but had limitations, especially in dynamic applications [3]. Then,

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composites and polymers emerged as corrosion-resistant alternatives, overcoming metals in durability against saltwater, ultraviolet (UV) radiation, and chemicals. Their versatility allows custom design, improved resistance, prolonged service life, and easy installation despite potential brittleness [4–6].

Polymer-based materials, particularly elastomers such as neoprene and ethylene propylene diene rubber (EPDM), are preferred for flexible marine applications [7]. These materials are modified to maintain their elasticity while enhancing strength and durability against saltwater exposure. Out of all rubber types, silicone rubber magnetorheological elastomers (SR-MREs) stand out due to their distinctive ability to tune their properties in response to a magnetic field, presenting novel possibilities for diverse applications, particularly in marine engineering.SR-MREs stand out among elastomers due to their unique tunable properties in the presence of a magnetic field [8–10]. SR-MREs can undergo rapid changes in stiffness and damping, potentially adaptable for marine structures requiring on-demand mechanical property adjustments. However, it is crucial to note that SR-MREs may be susceptible to deterioration in specific service environments, potentially leading to material failure over time.

Environmental factors, including temperature, moisture, humidity, and chemical exposure, can contribute to the mechanical degradation of SR-MREs [11–13]. This aging process, especially in outdoor conditions, involves complex chemical reactions that gradually degrade the material. Exposure to outdoor conditions introduces temperature changes, moisture, humidity, and exposure to chemicals, initiating chemical reactions that gradually lead to the degradation of materials over time. Notably, seawater further speeds up this deterioration process by worsening the negative effects caused by outdoor exposure conditions [14, 15]. Multi-factor (time, temperature, and concentration) aging tests were conducted [16] to investigate the effect of seawater on the physical/mechanical degradation of the material. The findings reveal that combined aging affects the surface morphology of SR-MRE due to the intrusion of water molecules into the SR-MRE material, further accelerating the aging. This results in swelling and physical defects, such as microcracks and pitting, further weakening the SR-MRE material [17].

The study on the durability of SR-MRE in terms of aging under several environmental conditions like temperature, UV light, and natural weathering has been observed. According to Aziz *et al*. [18], temperature changes affected the phase shift angle (δ) at high-strain regions, indicating significant alterations in the deterioration process of SR-MRE. Thermal aging led to a 7% decrement in the hardness of soft SR-MRE. The Payne effect was noticed in both samples of SR-MRE across all strain levels. As the magnetic field strength increased, there was a decrease in the storage modulus, which was attributed to the disruption of the matrix chain and the interaction between the matrix and particles. Another study conducted by Wibowo *et al*. [19] highlighted the impact of UV exposure and water degradation processes, which led to the breakdown of particle bonds within the MRE sample and, at the same time, revealed a notable degradation in microstructure and a significant reduction of up to 80.5% in rheological properties. Johari *et al*. [20] observed surface crystallinity and shrinkage were identified as key factors contributing to the formation of erosion and flaw lines, resulting in 20% decrease in elongation performance. In a recent study [21] conducted in a saltwater environment, water molecules such as Na^+ and Cl^- ions increased the water diffusion into the matrix, allowing matrix molecules to move with each other, which consequently softened the matrix. While prior research explored SR-MRE durability in saltwater, its behavior in seawater remains unreported. Seawater composition's unpredictability, influenced by atmospheric conditions, can impact SR-MRE. A previous study [22] revealed significant polymer crystalline structure impact after exposure to magnesium (Mg) for durations of 3 and 6 months, elevating melting point (T_m) and increasing stiffness. Also, several studies [23, 24] investigated the effect of both artificial seawater and natural seawater on the morphological and structural properties. These studies conclusively demonstrated that the existence of other metal salt ions within seawater has a significant effect on the evolution of the corrosive interface and induces severe microcracking on the surface of polymer and rubber in seawater. Despite existing research on the influence of sodium chloride (NaCl) on MREs in marine environments, there is a significant gap in understanding how various metal compositions present in seawater impact the real-time performance and durability of MREs.

Addressing this gap, this study comprehensively evaluates SR-MRE aging behavior in seawater to

gain accurate insights into deterioration mechanisms, emphasizing their application in marine components like semi-active engine mounting, damping systems and other applications in seawater. By replicating realistic marine conditions, it seeks to provide detailed insights into long-term durability patterns. This research focuses on assessing the impact of seawater conditions, consisting of elements potentially conducive to biological attacks, on the aging process of SR-MRE samples, with an emphasis on obtaining additional data and validating results against controlled laboratory conditions. Overall, this research aims to bridge the gap in understanding SR-MRE durability in real-world marine scenarios, ultimately contributing to the development of more reliable materials for the marine industry.

2. Methods

2.1. Sample preparation

The matrix used in this study was room temperature vulcanization (RTV) SR NS625A/Nippon Steel, purchased from Nippon Steel Co., Tokyo, Japan. SR-MRE consisted of 70 wt% CIP and 30 wt% of SR as a matrix. This percentage was determined to provide the optimal balance between a stable response to magnetic stimulation and a higher magnetorheological (MR) effect. By using 70 wt% of CIPs, the SR-MRE exhibited favorable rheological properties when subjected to magnetic fields, resulting in enhanced controllability and responsiveness, and was deemed to be the most suitable for achieving the desired MR performance in the material. CIP type OM grade supplied by Badische Anilin Soda-Fabrik (BASF) from Ludwigshafen, Germany, with size and density of 1–10 μm and 7.874 g/cm^3, respectively, was selected as magnetic particles. The preparation of the SR-MRE-based SR samples is shown in Figure 1.

There were several steps involved in the sample preparation. First, 70 wt% of CIP was added to the SR matrix and mixed for 10 minutes with a mechanical stirrer (WiseStir, HS-30D, Daihan Scientific Co., Ltd, Semenyih, Selangor, Malaysia) at a speed of 200 rpm at 25 °C until it appeared visually homogenous. After that, 1.5% of the curing agent NS 625 B (Nippon Steel) was added, and the mixture was stirred for 1 min before being poured into the mould. After 4 h, the SR-MRE specimen was taken out of the mould and cut into a circular shape with a 20 mm diameter and a 1 mm thickness for rheology testing.

2.2. Seawater immersion procedure

SR-MRE samples were soaked in seawater for 30 days. The seawater used to immerse the samples was collected from Bagan Lalang, Selangor, Malaysia and had an average pH measurement of 7. The average ion composition of seawater was C, O, Si, Fe, Na, Cl, and Mg ions, which was characterized by energy-dispersive X-ray spectroscopy (EDX) analysis. The tank size used in this study was 38 cm in length, 24 cm in width, and 38 cm in height. Then, the immersed samples were dried at room temperature for 24 h before proceeding to physical characterization. The pH value of the seawater remained constant at 7 throughout the immersion period of SR-MRE, indicating the remarkable pH stability of seawater. Seawater possesses a natural buffering capacity, which enables it to resist significant pH alterations even in the presence of exogenous materials like SR-MRE. This buffering capacity helps maintain the pH level unchanged by neutralizing any potential changes caused by the interaction between the SR-MRE and the seawater. The minimal chemical interaction between the SR-MRE and seawater further contributes to the preservation of the seawater's pH, resulting in

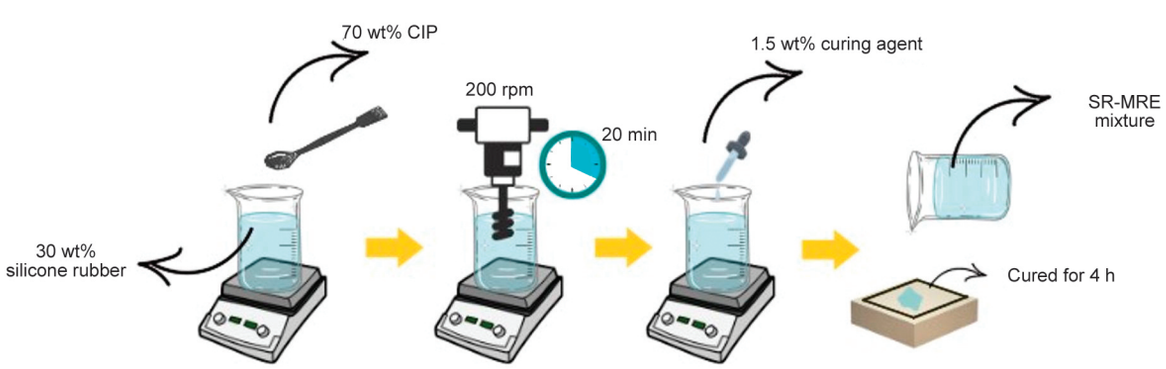

Figure 1. Sample preparation SR-MRE sample.

little to no observable changes during the immersion period.

2.3. Microstructure characterization

Surface and cross-section images were obtained using scanning electron microscopy (SEM, Vega 3, Tescan Analytic, France) at 15.0 kV, with samples coated in a gold layer to prevent charging during analysis. The cross-section analysis examines particle distribution, while surface observation identifies smooth regions and defects post-fabrication and immersion.

2.4. Hardness analysis

Meanwhile, SR-MRE hardness was assessed using a Shore-A durometer (Model HBA 100.0, Sauter, Balingen, Germany) in accordance with ASTM D2240. Five random points on each sample's flat, clean, and smooth surface were tested, and the average data across positions were recorded.

2.5. Mechanical analysis

The mechanical properties of both unimmersed and immersed SR-MRE samples were assessed by using a tensile test in accordance with the ASTM D412 testing standard. The SR-MRE samples were cut into dumbbell shapes from the sheets using dies designed for tensile cutting. The samples were symmetrically clamped on the top and lower grippers to ensure even stress distribution over the sample's cross-section. The tensile test was conducted at a controlled room temperature of 27 °C with a loading speed of 500 mm/min using a universal testing machine (Shimadzu AGX-S, Kyoto) in quasi-static uniaxial mode.

2.6. Water uptake analysis

Moreover, the water uptake test assessed SR-MRE's water absorption after 30 days in seawater. The initial mass was measured using a precise analytical weight balance (AUW220, Shimadzu, Kyoto, Japan), which served as the dry sample baseline. After immersion, samples were weighed again, and water uptake was determined by calculating the difference between final and initial masses, expressed as a percentage (Equation (1)):

$$M\,[\%] = \frac{M_t - M_0}{M_0} \cdot 100 \tag{1}$$

The water uptake percentage of the specimens was calculated by using Equation (1), where M [%] is the water uptake, M_t is the weight of the wet sample at a given time, and M_0 is the initial weight of the sample.

2.7. Chemical analysis

The ATR-FTIR analysis was carried out at room temperature using a PerkinElmer Spectrum 100 FTIR spectrometer (USA). FTIR spectra were logged in between 4000 and 800 cm^{-1} at a resolution of 2 cm^{-1} with 10 scans.

2.8. Rheological analysis

The SR-MRE sheet was punched to a 20 mm diameter and 1 mm thickness using a hollow hole punch tool. Samples were then subjected to oscillatory shear testing at 25 °C, while magnetic flux density values were automatically recorded by a Teslameter connected to a rheometer for various applied currents. Rheological properties related to strain sweep were varied from 0.01 to 10% at a constant frequency of 1 Hz using an oscillation mode rheometer (MCR 302, Anton Paar, Graz, Austria). By varying the applied current from 0 to 5 A, the rheological properties were obtained, and the test was repeated three times to ensure that the data was consistent.

3. Results and discussion

3.1. Morphological characterization of unimmersed and immersed SR-MRE

The fabricated unimmersed and immersed SR-MRE samples were determined with morphological analysis to identify any imperfections or defects that could arise during the fabrication process. Figure 2 depicts cross-section observation for the micrograph analysis of the unimmersed and immersed SR-MRE samples. As shown in Figure 2a, small agglomerations and voids were observed in the unimmersed sample. Any fabrication of SR-MRE has the potential to have agglomerations and voids. Based on Figure 2a, this investigation successfully shows the occurrence of small agglomerations and voids in the SR-MRE immediately after the fabrication process. These defects, identified in the unimmersed sample, have the potential to influence the bonding between the matrix during subsequent testing stages. In contrast, Figure 2b shows that after being immersed in seawater for 30 days, the micrograph analysis of the SR-MRE samples revealed a reduced number of voids and agglomerations compared to the unimmersed samples. It may be due to swelling of the SR-MRE, which may have caused the matrix to expand

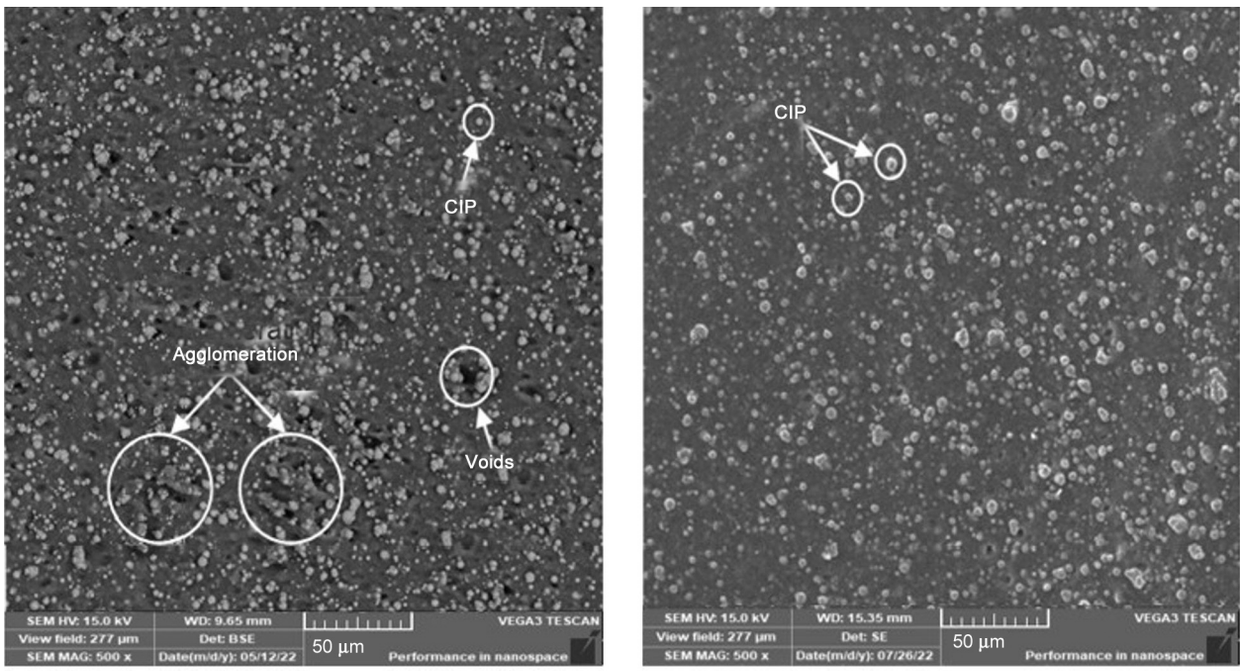

Figure 2. Micrographs analysis of a) unimmersed and b) immersed cross-section SR-MRE samples.

and fill the voids in the matrix [25]. This aligns with the findings of Muñoz *et al*. [25] and Karmaker *et al*. [26], both of whom observed that water absorption induced swelling in natural reinforcement fibers within epoxy resin and polypropylene composites. Muñoz *et al*. [25] reported a decrease in voids and an improvement in cohesion between fibers and the matrix in epoxy resin composites following water absorption-induced swelling, attributing this reduction to the expansion of the matrix, filling the voids. Similarly, Karmaker *et al*. [26] investigated whether fiber swelling due to water absorption could fill the gaps between jute fibers and the polypropylene matrix. They reported a similar effect, where the swelling of fibers led to enhanced cohesion and filling of empty spaces previously occupied by the fibers [26]. The surface analysis of the unimmersed and immersed SR-MRE samples has shown some unique observations as compared to the cross-section of SR-MRE, which is depicted in Figure 3. The unimmersed SR-MRE had a smooth surface, as shown in Figure 3a. However, based in Figure 3b, after 30 days of immersion, some surface defects such as stretched network, pitting, and microcrack were observed for the SR-MRE. These defects could be due to the interaction between the seawater and the SR-MRE, which contains various ions and contaminants such as carbon (C), oxygen (O), silicon (Si), iron (Fe), sodium (Na), chloride (Cl), and magnesium (Mg) ions, that can react with the SR-MRE and weaken the material's mechanical properties. As the immersion time increased, the amount of seawater moisture infused into the sample was also increased [27, 28]. The presence of these defects could accelerate further deterioration due to seawater disruption that led to the occurrence of 'pathways', which allowed the oxygen to diffuse into the SR-MRE surface specimen. Moreover, it was assumed that the seawater disruption would cause a "chain movement" in the SR-MRE during immersion, thus might lead to the interfacial debonding between matrix and magnetic particles. The debonding phenomenon leads to a reduction in material hardness, which corresponds to a decrease in hardness [29]. This correlation further strengthens the understanding of the relationship between swelling behavior and mechanical properties in SR-MRE materials.

The previous study discussed how immersion in salt water changed the property of SR-MRE and generated micro defects on its surface, where erosion lines appeared on the surface [21]. Therefore, increasing the saltwater content was believed to elevate the risk of pitting corrosion, particularly at damaged spots. Hence, this pitting corrosion was believed to cause the impurities to completely penetrate the SR-MRE samples within a short time, indicating higher seawater absorption [30]. Additionally, defects known as micro-cracks on the surface of SR-MREs were

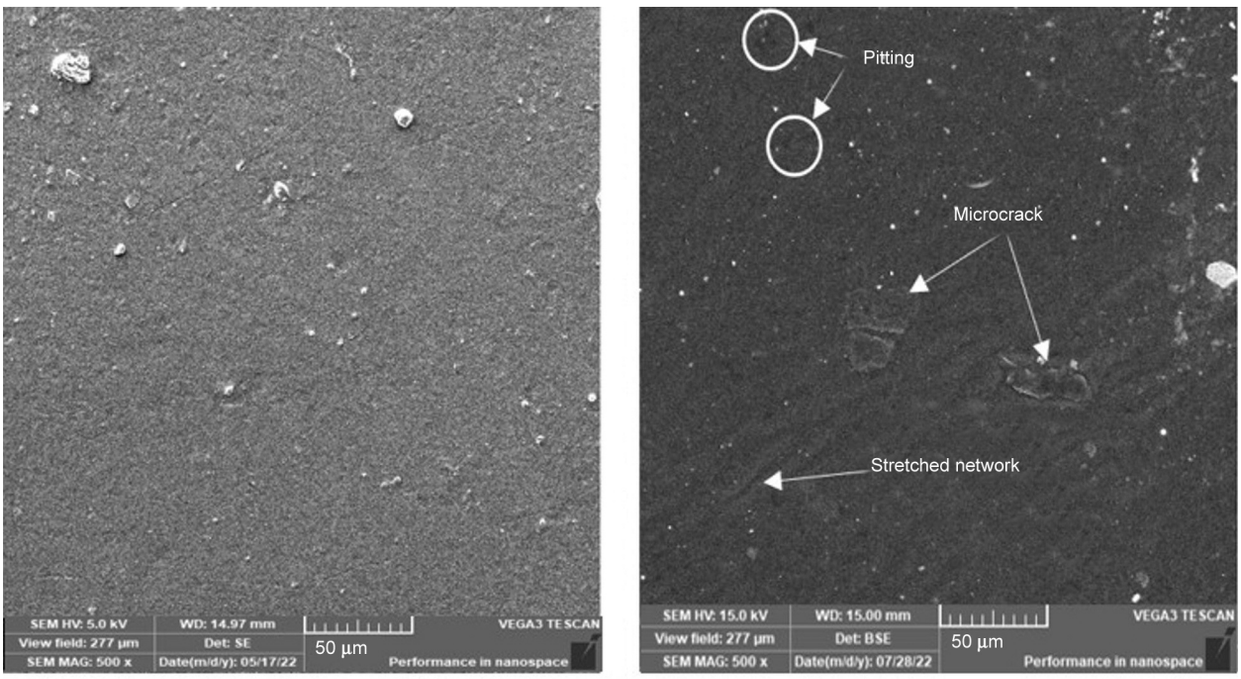

Figure 3. Surface micrographs of a) unimmersed and b) immersed surface SR-MRE samples.

observed, and this phenomenon may be attributed to osmotic stresses. The immersion of SR-MRE in seawater for 30 days is likely to result in a hypotonic condition. This is because seawater has a lower concentration of solutes compared to the internal environment of the SR-MRE material. As a result, water is likely to flow into the material, causing it to swell and expand. Consequently, water may penetrate into the material, leading to swelling and expansion. The accumulation of molecules within these micro-voids may trigger osmotic cracking, emerging as a primary factor in material degradation. EDX analysis was conducted to provide evidence of the presence of seawater ions on the surface of SR-MRE.

Table 1 presents a comprehensive overview of the detailed EDX analysis for both unimmersed and immersed SR-MRE samples. Four types of elements, C, O, Si, and Fe, were detected for unimmersed SR-MRE. After 30 days of immersion, the samples seemed to exhibit the new formation of new elements such as Na, Cl, and Mg. As seen in Table 1, C and Si are the predominant elements in the immersed SR-MRE, constituting approximately 59.11% of the total mass and 67.31% of the total atoms. These elements are integral components of the polymer matrix in SR-MREs and play a significant role in determining their mechanical and structural properties. The presence of Cl and Na in the immersed SR-MRE indicates the incorporation of ions from seawater into the material during immersion. They are commonly found in seawater, and their detection suggests that the material has interacted with the surrounding marine environment. Besides, O is also detected in the immersed SR-MRE, which is likely derived from both the polymer matrix and absorbed moisture from the environment. Oxygen plays a crucial role in various chemical reactions and may contribute to the degradation processes observed in the immersed SR-MRE.

Table 1. Elements in the immersed SR-MRE.

Element	Mass [%]	Atom [%]
C	30.05	47.62
Si	29.06	19.69
Cl	15.21	8.17
Na	13.03	10.79
O	10.64	12.66
Fe	1.11	0.38
Mg	0.89	0.69

Additionally, the presence of Mg and Fe in small quantities suggests possible contamination or trace elements present in the material. These elements may have originated from the manufacturing process or external sources. Fe content, which was predominately determined by the CIP, was assumed due to the oxidation layer that formed on the surface of the CIP [31]. When CIP is immersed in seawater, it may undergo reactions with oxygen and moisture present

in the environment, leading to the formation of an oxide layer on its surface. This oxide layer may contain iron oxide compounds, contributing to Fe content detected by EDX. Another possibility was that during the immersion in seawater, the CIP protruded on the surface of the sample due to polymer matrix shrinkage. Seawater diffusion through the interface into SR-MRE accelerated the hydrolysis of the samples, similar to other ions from seawater. There were also cracks, voids, and deterioration of the interface between the CIP and matrix, which led to degradation [32].

In addition to the microstructure analysis, a comparative analysis of unimmersed and immersed SR-MRE under seawater conditions was discussed in Table 2. The other properties, such as hardness and tensile properties of unimmersed and immersed SR-MRE, were also observed.

Besides, the results revealed that the unimmersed sample's mean average hardness was 57.5 HA, with a standard derivation of 0.7. As can be seen in Table 2, initially, the unimmersed samples exhibit a mean hardness of 57.5 HA, while the immersed samples, after 30 days of immersion in seawater, show a mean hardness of 54.0 HA, indicating a 6.5% hardness depletion. Khairi *et al*. [21] demonstrated a 15% reduction following immersion in saltwater. Similarly, in an investigation focusing on seawater, there was a consistent reduction in hardness. This parallel trend indicates a coherent impact of aqueous environments on the mechanical properties of SR-MRE samples. This reduction in hardness is attributed to water absorption from seawater, causing plasticization effects and weakening of the rubber-particle interactions. Aligned with Wang *et al*. [33] revealed that the plasticization of the matrix led to the appearance of microcracks. This absorption might lead to the plasticization effect coincidently because of the medium absorption, which resulted in the reduction of hardness in SR-MRE samples. The moisture-induced plasticization, which exhibited weak bonding, involves the disruption of van der Waals bonds between the rubber chains and rubber-particle interaction.

Based on Table 2, before immersion, the unimmersed samples exhibited lower elongation at break, suggesting greater flexibility and deformability under tensile stress. However, upon immersion, the samples displayed decreased stiffness accompanied by an increase in elongation at break. This change in mechanical behavior can be attributed to several factors, including water absorption, ion-polymer interactions, and the presence of contaminants. Water absorption leads to swelling, causing the disentanglement of polymer chains and an increase in flexibility while decreasing stiffness. Also, a decrease in Young's modulus observed in the immersed SR-MRE samples compared to the unimmersed ones could be attributed to several factors. One possible explanation is the ingress of water into the material, leading to a phenomenon known as "plastification". This process involves the water molecules interacting with the polymer chains, causing them to become more flexible and reducing the overall stiffness of the material. Additionally, hydrolysis, which is the chemical breakdown of polymer chains by water molecules, could also contribute to the decrease in elastic modulus. The presence of water in the immersed samples may facilitate hydrolytic degradation of the polymer matrix, leading to a reduction in stiffness. The tensile test was also carried out to understand the different effects between unimmersed and immersed SR-MRE. Figure 4 depicts stress-strain curves, which illustrate the relationship between applied stress and resulting strain for both unimmersed and immersed SR-MRE samples under seawater conditions. The curves for

Table 2. Comparative analysis of unimmersed and immersed SR-MRE under seawater conditions.

MRE comparative analysis of MRE properties in seawater		
Properties	**Unimmersed**	**Immersed**
Microstructure (Cross-section)	Agglomerations and voids	Reduced number of voids and agglomerations
Microstructure (Surface)	Smooth surface observed	Stretched network, pitting, and microcrack
EDX analysis	C, O, Si, Fe	C, O, Si, Fe, Na, Cl, and Mg
Hardness	Mean: 57.5 HA, SD: 0.7 HA	Mean: 54.0 HA, SD: 0.7 HA
Tensile	Elongation at break: 238% Young's modulus: 0.59 MPa SD: 8	Elongation at break: 251%, Young's Modulus: 0.54 MPa, SD: 6

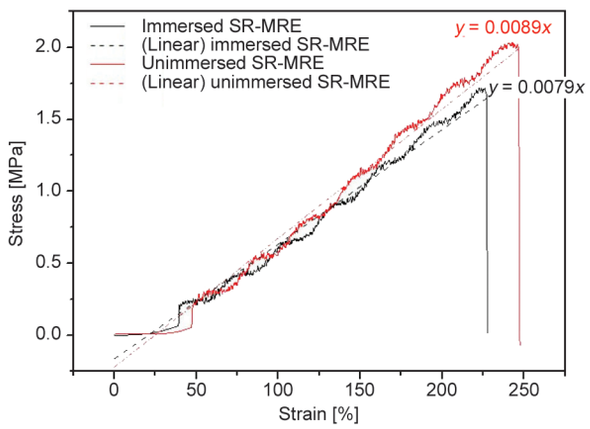

Figure 4. Stress-strain curves of unimmersed and immersed SR-MRE samples under seawater condition.

immersed SR-MRE samples consistently exhibit lower stress values across varying levels of strain compared to unimmersed samples. This indicates that the immersed SR-MRE samples experience reduced resistance to deformation or lower mechanical strength when exposed to seawater compared to their unimmersed counterparts. the stress-strain curves depicted in Figure 4 are directly related to the mechanical properties of the material, including Young's modulus and elongation at break. Young's modulus represents the stiffness of the material, and a lower stress value on the stress-strain curve suggests a decrease in Young's modulus. Additionally, elongation at break indicates the material's ability to stretch or deform before failure. The lower stress values observed in the stress-strain curves for immersed SR-MRE samples suggest reduced stiffness, which aligns with a decrease in Young's modulus. Furthermore, the lower stress values may also imply increased elongation at break, indicating enhanced flexibility or ductility of the material when immersed in seawater compared to the unimmersed samples.

3.2. Moisture absorption

The immersion test was conducted to assess the water absorption properties of SR-MRE. The samples were immersed in a container filled with seawater for a duration of 30 days at room temperature. The weight difference before and after immersion was recorded to calculate the moisture absorption, represented as a percentage of weight gain. The weight changes overtime was taken after 30 days immersion. Initially, the average mass of the SR-MREs before immersed in seawater from different batches was recorded at 14.600 g. After immersion, the masses increased slightly to 14.625 g, indicating water uptake. The percentage average of SR-MREs increase in weight due to water absorption was calculated, resulting in values of 0.17%. These results indicate that all SR-MRE samples from three different batches exhibited minimal water uptake after 30 days of immersion in seawater. This suggests that the immersion applied to the SR-MRE material had minimal impact on its water absorption properties. The results highlight the potential of SR-MRE as a viable material for marine applications that require resistance to water absorption.

3.3. Attenuated total reflectance – Fourier-transform infrared spectra

Figure 5 demonstrates the peaks representing the unimmersed and immersed SR-MRE bonding characteristics. The transformation of the functional group for both unimmersed and immersed SR-MRE was analyzed by using ATR-FTIR analysis. The ATR-FTIR spectrometer was used for the measurement technique with a spectra measurement range from 4000 to 600 cm^{-1}. The results in Figure 5 revealed that the organic compound produces some characteristic spectra in the infrared region. Generally, absorbance refers to the peak intensity observed in the spectra, which is indicative of the concentration of functional groups present in the sample. Therefore, a higher absorbance corresponds to a greater ability to absorb spectra and signifies a higher concentration of functional groups. The presence of NaCl molecules resulted in the absence of vibrational states in the considered wavenumber range. Thus, the interaction of Na^+ and Cl^- ions with surface hydroxyl groups (adsorption effect) and with adsorbed H_2O molecules determined the spectral

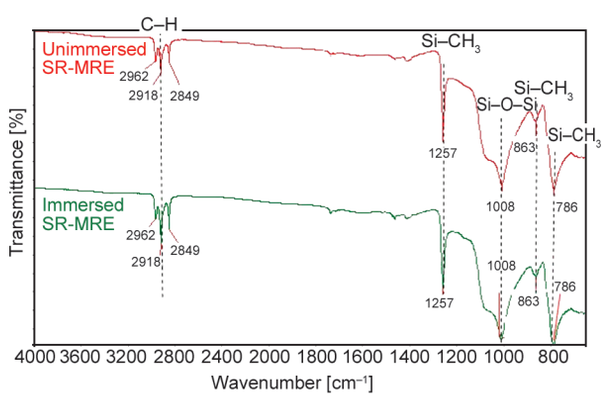

Figure 5. FTIR spectra of unimmersed and immersed SR-MRE samples of 30 days of immersion in seawater under seawater conditions.

effect of the appearance of NaCl molecules (hydration effect). Both effects were observed in the range of 2750–4000 cm^{-1} wavenumber [34]. The observed effects are relevant to the study of the effect of NaCl adsorption on the IR spectra of SR-MRE.

The absorption peaks at 2849, 2918, and 2962 cm^{-1} were due to the symmetric and asymmetric stretching vibrations of the methyl ($–CH_3$) group. The peak at 1257 cm^{-1} corresponded to the $Si–CH_3$ bond. Moreover, the peak at 1008 cm^{-1} was attributed to the existence of Si–O–Si, and the strong vibration of $Si–(CH_3)_2$ [35]. There were no apparent changes in the Si–O–Si bond between unimmersed and immersed SR-MRE. This could be due to the immersion duration of SR-MRE was not enough to induce Si–O–Si chain scission. Bleszynski and Kumosa [36] found that the chain scission of the Si–O–Si backbone of silicone rubber, occurring after approximately 10 weeks, is responsible for material damage, which involves the damage affecting the nonpolar methyl groups responsible for the hydrophobic properties of the RTV material. Compared to the immersed sample's FTIR spectrum with the unimmersed sample, no appreciable changes were detected in the intensity of any five peaks of SR-MRE. The summary of the FTIR results for unimmersed and immersed SR-MRE, the peaks in the absorption band diagram and their corresponding infrared wavenumbers are shown in Table 3.

Table 3. FTIR absorption peaks of unimmersed and immersed SR-MRE.

Functional group	Wavenumber [cm^{-1}]	Characteristics
$Si(CH_3)_2$	700–870	CH_3 rocking vibration
Si–O–Si	1000–1100	Si–O stretching vibration
$Si–CH_3$	1255–1270	CH_3 symmetrical deformation vibration
C–H in CH_3 (stretching)	2960–2962	C–H stretching vibration

3.4. Rheological properties of SR-MRE

Figure 6 presents a comparison of the storage and loss moduli of unimmersed and immersed SR and SR-MRE samples under both 0 and 5 A conditions. Regarding the storage modulus, unimmersed SR exhibits a broad linear viscoelastic (LVE) region, reflecting its inherent flexibility and ability to deform under stress. Conversely, after 30 days of immersion, the storage modulus of SR decreases by approximately 7%, attributed to weakened intermolecular interactions resulting from water uptake. The storage and loss modulus results of unimmersed and immersed SR provide a baseline for understanding the intrinsic mechanical properties of the material. The broad linear viscoelastic (LVE) region observed in the storage modulus curve of SR signifies its inherent flexibility and ability to deform under stress. Conversely, the variations in storage modulus observed in immersed SR and SR-MRE samples reflect the influence of external factors such as water absorption and magnetic field application.

The rheological properties of unimmersed and immersed SR-MRE were evaluated in Figure 6. In Figure 6a, the initial storage modulus of unimmersed SR-MRE increased from 0.25 to 0.38 MPa under 0 and 5 A, respectively, corresponding to approximately 0 T and 0.85 T. Immersed SR-MRE at 0 A exhibited a lower initial storage modulus due to weak CIP interactions without a magnetic field. Seawater immersion further decreased the storage modulus, which can be attributed to water "plastification", where water molecules interact with the polymer chains, making them more flexible and reducing stiffness. Additionally, hydrolysis, the chemical

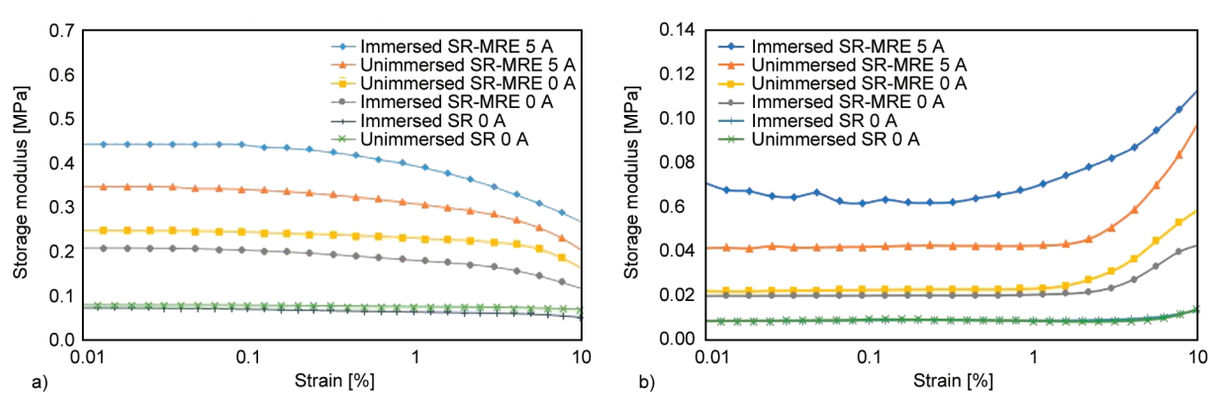

Figure 6. Comparison of a) storage modulus and b) loss modulus of unimmersed and immersed SR and SR-MRE samples under both 0 and 5 A.

breakdown of polymer chains by water molecules, may also contribute to the reduction in storage modulus. However, under a magnetic field (5 A), immersed SR-MRE showed a higher storage modulus, indicating enhanced elastic energy storage. Another observation in Figure 6a, both unimmersed and immersed SR-MRE samples displayed a decreasing trend in storage modulus with strain, with 5 A current narrowing the linear viscoelastic (LVE) region due to increased elastic energy storage from magnetic field influence. At 0 A, a broad LVE region was observed, signifying linear changes in elastic and viscous properties. In contrast, 5 A current narrowed the LVE region due to the high sensitivity of the particle chains to deformation under the magnetic field. The distance-dependent nature of dipole-dipole interactions results in the breaking of connections between the CIPs, leading to a decrease in the storage modulus. This is called the Payne effect, which causes microstructural damage. The contrasting behavior of the storage modulus after immersion in seawater at off-state and on-state conditions for 1 month suggests different underlying mechanisms at play. When immersed in seawater at off-state conditions, the storage modulus decreases, which is expected due to the degradation effects of seawater on the material. However, in the case of immersion at on-state conditions, where the material is subjected to a magnetic field (5 A current), the storage modulus does not decrease as expected due to swelling. This discrepancy may be attributed to the dominant influence of the magnetic field (current effect) over the swelling effect caused by immersion in seawater. The magnetic field's influence on the particle chains: when an external magnetic field is applied to the SR-MRE material, the CIPs will begin to vibrate and produce magnetic moments between the particles, which would then tend to form a chain-like structure, which may lead to a higher storage modulus, counteracting the potential decrease caused by swelling. Therefore, the current effect appears to outweigh the swelling effect, resulting in an overall increase in storage modulus under on-state conditions [37].

Meanwhile, it is well-known that the alignment of the CIPs in SR-MRE under the influence of magnetic field strength can affect the loss modulus of the material, too. Figure 6b illustrates the loss modulus of unimmersed and immersed SR-MRE samples at 0 and 5 A conditions. Unimmersed SR-MRE at 0 A exhibits an initial loss modulus of 0.03 MPa, reaching a maximum of 0.05 MPa at 10% strain, lower than that at 5 A. Immersed samples show slightly lower values, suggesting a mild impact of seawater immersion. At 5 A, unimmersed and immersed samples displayed an increased loss modulus (0.04 MPa minimum, 0.10 MPa maximum), and (0.07 MPa minimum, 0.11 MPa maximum), respectively, indicating limited effect of seawater immersion. When an external magnetic field is applied to the SR-MRE, the CIPs would begin to vibrate and produce magnetic moments between the particles, which would then tend to form a chain-like structure. [38]. A reduction in the distance between the CIPs within the matrix will enhance the interaction between CIPs and the matrix, thereby improving the rheological response of the SR-MRE.

3.5. Seawater element interaction mechanism in SR-MRE

Figure 7 depicts an illustrated phenomenon that could be attributed to changes in particles interaction in SR-MRE in seawater. Previous studies [12, 37] revealed that the decrement of the storage modulus might be due to the variations in outcomes of seawater such as chemical composition, salinity, temperature, or other factors.

As depicted in Figure 7a, the CIP and polymer chain of the unimmersed SR-MRE sample are uniformly dispersed in the matrix. Void formation in an unimmersed SR-MRE sample, which can be seen in Figure 3a, can lead to decreased rheological properties, particularly the modulus. Meanwhile, as for the immersed sample in Figure 7b, when the SR-MRE is exposed to seawater, the water and ions can penetrate into the material through its voids or other openings, which can be seen in the surface analysis in Figure 3b, creating a pathway for the diffusion of seawater into SR-MRE. Besides, it is possible for other elements from seawater contents to diffuse into the SR-MRE during the immersion process. The presence of ions such as C, O, Si, Fe, Na, Cl, and Mg ions within the SR-MRE can cause the material to swell due to the attraction of water, thus penetrating the material. This swelling might create additional stress within the material, leading to the development of cracks and voids, which can then coalesce into larger macro-cracks over time. The development of cracks and voids in the material can also contribute to the decrement of storage modulus [39]. However, under on-state conditions, it is observed

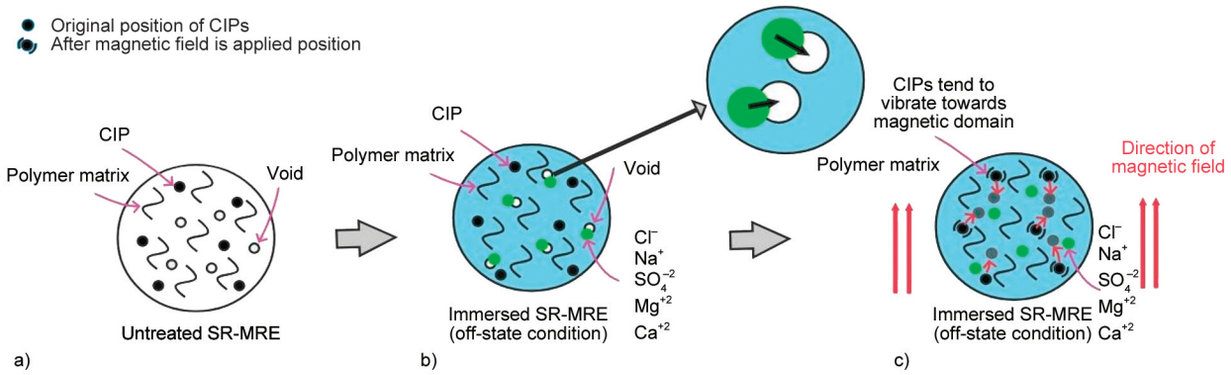

Figure 7. Illustration of seawater immersion phenomenon during immersion of a) unimmersed b) immersed at off-state condition, and c) immersed at on-state condition SR-MRE samples.

that the CIPs in SR-MRE tend to exhibit vibrational tendencies and move in response to the magnetic domain, rather than undergoing direct alignment with the magnetic field. This behavior may create the impression that the CIPs are aligning along the direction of the magnetic field [40]. This movement and vibration induced by the magnetic domain alter the behavior of SR-MRE, leading to increased modulus and modified mechanical properties. As the magnetic particles within the material tend to move and vibrate in response to the external magnetic field, this reorientation results in a structural change within the material [41]. The movement and vibration of the particles contribute to the formation of chains or networks along the field lines, reinforcing the material's internal structure and making it stiffer. Consequently, the movement and vibration of the magnetic particles restrict the material's flexibility and its ability to deform, influencing its response to external forces and thereby affecting its mechanical behavior.

Previously, it has also been reported that seawater was more corrosive than salt water due to their chloride content owing to the biological activity in the presence of microorganisms which may accelerates material degradation [42, 43]. In chloride-rich environments such as seawater, microorganism may contribute to the degradation by accelerating corrosion mechanism. Microorganism in seawater tend to form biofilms on the surface of materials and these biofilms act as a protective layer for the microorganisms but can also trap moisture and corrosive agents close to the material's surface. This prolonged contact can resulting the corrosion process, facilitating and propagating the cracks within the material.

4. Conclusions

This study highlights the impact of seawater exposure on the microstructure, mechanical, and rheological properties of SR-MREs. Immersion in seawater led to a reduction in voids and agglomerations in SR-MRE samples due to swelling, resulting in a reduction in hardness of about 6% depletion due to water absorption and plasticization effects. Minimal water uptake was observed in the immersed SR-MREs, showcasing resistance to water absorption crucial for marine applications. The increase in elongation at break and decrease in Young's modulus after immersion of SR-MRE in seawater suggest a shift towards enhanced flexibility and reduced stiffness attributed to interactions with seawater ions. Additionally, the rheological properties of unimmersed and immersed SR-MRE were assessed, with a decrease in storage modulus observed after seawater immersion attributed to water-induced plastification and hydrolysis. However, under a magnetic field, immersed SR-MRE displayed increased energy storage, suggesting the dominance of the current effect over swelling, resulting in a higher storage modulus under on-state conditions. These findings underscore the importance of understanding SR-MRE behavior in harsh environments, urging further exploration to enhance material durability for seawater applications. These findings underscore the significance of studying SR-MRE behavior under harsh conditions for seawater applications, urging further exploration, including the development of mathematical models for different microparticle fractions and the exploration of new materials to enhance SR-MRE durability.

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