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Effects of Sub-Zero Temperatures on the Viscoelastic Properties of Magnetorheological Elastomer

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Abstract

Magnetorheological elastomers (MREs) are smart materials capable of tuneable stiffness and damping under applied magnetic fields. This renders the material as a promising candidate for adaptive vibration control and semi-active engineering applications. While their behaviour at elevated temperatures has been widely explored, limited attention has been given to their performance under sub-zero conditions, which are critical for cold-environment applications. As this has become an important concern, the study investigates the influence of sub-zero temperatures on the viscoelastic properties of silicone-based MREs, containing 70 wt.% of carbonyl iron particles. For the rheological study under dynamic shear loading, the MRE samples were fabricated under isotropic condition and subjected to controlled sub-zero conditioning at −15°C, −20°C and −25°C for five days each. Rheological characterization was conducted using oscillatory shear tests under both off-state and on-state magnetic field conditions. Key parameters including initial storage modulus (G'₀), storage modulus (G') and magnetorheological (MR) effect, were evaluated. Hardness test was then carried out using Shore A hardness, while morphological analysis using scanning electron microscopy (SEM) was performed to correlate microstructural changes with the properties change. The results show a significant increase in stiffness with decreasing temperatures, as indicated by higher storage modulus and hardness values. This finding is attributed to restricted polymer chain mobility and enhanced particle–matrix interactions. However, the relative MR effect decreases at lower temperatures, suggesting reduced field-induced tunability capability due to limited particle interactions within the stiffer matrix. Microstructural observations reveal wrinkles, interfacial debonding and small holes caused by thermal mismatch, contributing to increased rigidity and reduced energy dissipation. Overall, the findings highlight a trade-off between stiffness enhancement and MR performance under sub-zero conditions, providing valuable insights for the design of MRE-based systems for low-temperature applications.

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INTRODUCTION

Magnetorheological elastomers (MREs) are a class of smart composite materials consisting of

magnetizable particles dispersed within an elastomeric matrix, enabling rapid and reversible modulation of mechanical properties under an external magnetic field. This field-responsive behaviour, known as the magnetorheological (MR) effect, allows MREs to exhibit tuneable stiffness and damping characteristics, making them highly attractive for applications such as vibration isolation, adaptive structures and semi-active control systems[1]. The performance of MREs is typically evaluated through viscoelastic parameters, including G′, loss modulus and damping factor, which govern their ability to store and dissipate energy under dynamic loading conditions. Importantly, these properties are strongly influenced by the microstructural configuration of the material, including particle distribution, particle–matrix interaction and matrix composition. For instance, the fraction and arrangement of carbonyl iron particles (CIPs) have been shown to significantly affect the G′ and linear viscoelastic behaviour of MREs, where higher particle content generally increases stiffness but may reduce durability due to structural instability [2–3].

Despite their promising adaptive capabilities, the long-term durability of MREs under real operating environments remains a critical concern that directly impacts their reliability and service life [4]. In contrast to controlled laboratory conditions, MREs in real applications are exposed to complex environmental stressors such as temperature variations, humidity, ultraviolet radiation and mechanical fatigue, all of which can induce degradation over time. Environmental aging can lead to changes in the elastomeric matrix, including chain scission, crosslinking and oxidation, which ultimately alter the mechanical and rheological behaviour of the material. For example, accelerated weathering, which combine heat, humidity, and UV exposure, have shown that prolonged environmental stress results in surface roughening, increased void formation and changes in phase shift angle, indicating modifications in viscoelastic behaviour and internal structure [5–6]. Furthermore, aging-induced changes in interfacial adhesion between magnetic particles and the polymer matrix can lead to embrittlement and reduced durability of MRE systems. The interaction between magnetic particles and the elastomer matrix is particularly critical, as it governs stress transfer efficiency and magnetic responsiveness under external fields[7]. Poor interfacial bonding can introduce defects, voids and weak regions within the composite, leading to reduced mechanical integrity and compromised MR performance.

Among various environmental factors, temperature is one of the most dominant parameters influencing the viscoelastic and durability behaviour of MREs, primarily due to the temperature-sensitive nature of polymer matrices. Temperature variations directly affect molecular mobility, crosslink density and interfacial interactions, leading to significant changes in mechanical and rheological properties. Increasing temperatures fade the molecular chains of the elastomer, substantially weakening the interfacial adhesion and bonding strength between the polymer matrix and the embedded magnetic filler [8–9]. Therefore, a comprehensive understanding of temperature-dependent behaviour is crucial for predicting the performance of MREs in real-world applications. In contrast to high-temperature studies, the behaviour of elastomeric materials under low and sub-zero temperatures presents a different set of challenges, primarily associated with reduced molecular mobility and increased stiffness. At low temperatures, polymer chains experience restricted motion, leading to increased modulus, reduced flexibility and potential embrittlement. Additionally, phenomena such as cold crystallization may occur in certain elastomers, where ordered structures form within the amorphous matrix, resulting in a significant increase in G′ and altered dynamic mechanical behaviour [10]. These structural transformations can drastically affect the energy dissipation capability and overall mechanical response of elastomer-based systems. Moreover, prolonged exposure to extremely cold environments has been shown to induce microcracks, pore formation and progressive surface degradation in rubber materials, further compromising their mechanical integrity [11]. Such findings emphasize that low-temperature environments introduce complex physicochemical changes that are fundamentally different from those observed at elevated temperatures.

Although significant progress has been made in understanding MRE behaviour under various conditions, a critical research gap remains in the systematic investigation of viscoelastic properties of MREs under sub-zero temperature conditions [11–12]. Most existing studies have focused on

improving MR performance, enhancing interfacial bonding or evaluating thermal aging at elevated temperatures [13], while relatively limited attention has been given to low-temperature performance. Furthermore, existing low-temperature studies on elastomers are primarily cantered on mechanical properties rather than dynamic viscoelastic behaviour, which is crucial for applications involving vibration control and energy dissipation [8]. The combined effects of matrix stiffening, particle–matrix interaction changes and potential crystallization phenomena under sub-zero conditions remain poorly understood in MRE systems. This lack of understanding limits the ability to design MREs for applications in cold environments such as aerospace, systems in cold regions and cryogenic engineering. Therefore, this study aims to systematically investigate the effect of sub-zero temperature on the viscoelastic properties of MREs, with particular emphasis on G′ and loss modulus behaviour under varying thermal conditions. By subjecting MRE samples to controlled sub-zero temperature environments, this work seeks to elucidate the underlying mechanisms governing stiffness evolution, energy dissipation and structural changes at low temperatures. The study also aims to correlate macroscopic rheological behaviour with microstructural and molecular-level phenomena, providing a comprehensive understanding of temperature-induced effects. Ultimately, the findings are expected to contribute to the development of more durable and reliable MRE systems capable of operating in extremely cold environments. This research not only addresses a significant gap in current literature but also provides valuable insights into the design and optimization of smart materials for next-generation engineering applications

MATERIAL AND METHODOLOGY

This section details the materials and experimental procedures employed to evaluate the performance of magnetorheological elastomers (MREs) under extreme cold environments. First, it outlines the selection of the silicone rubber matrix and carbonyl iron particles (CIPs), as well as the fabrication process for the isotropic MRE samples. Next, it explains the controlled sub-zero temperature conditioning protocol used to simulate cold-region applications. Finally, it describes the comprehensive characterization techniques utilized in this study, which include Shore-A hardness testing, rheological evaluation of the material's viscoelastic properties, morphological observation via scanning electron microscopy (SEM), and thermal transition analysis using Differential Scanning Calorimetry (DSC)

Materials

MRE samples were fabricated using a silicone rubber (SR) matrix, supplied by Nippon Steel Co, Japan, due to its excellent flexibility, thermal stability and suitability for dynamic mechanical applications. The SR was selected as the base elastomer because of its low glass transition temperature and stable viscoelastic response over a wide temperature range, making it appropriate for investigating sub-zero temperature effects. CIPs with high magnetic permeability and low remanence, purchased from BASF, Germany (OM grade) were used as the magnetic filler to impart MR functionality to the composite. The average particle size of the CIPs was in the 3.9 to 5.2 μm range, ensuring effective magnetic responsiveness and uniform dispersion within the matrix. The composition of MRE was fixed at a specific weight fraction of CIPs (70 wt.%), which has been widely reported as an optimal concentration for achieving a balance between mechanical strength and MR effect [2–14]. A curing agent compatible with the silicone rubber system was used to facilitate crosslinking during the fabrication process. All materials were used as received without further chemical modification. The selection of materials and composition was based on previous studies demonstrating that particle content and matrix properties significantly influence the viscoelastic and durability behaviour of MREs [2–15].

Fabrication of MRE Samples

The MRE samples were prepared through a conventional mixing and curing process to ensure homogeneous dispersion of magnetic particles within the elastomer matrix as shown in Figure 1. Initially, the required amount of CIPs was gradually added into the silicone rubber matrix and mechanically mixed for a predetermined duration to achieve uniform distribution. The mixture was then degassed under vacuum conditions to eliminate entrapped air bubbles, which could otherwise introduce defects and affect mechanical performance.

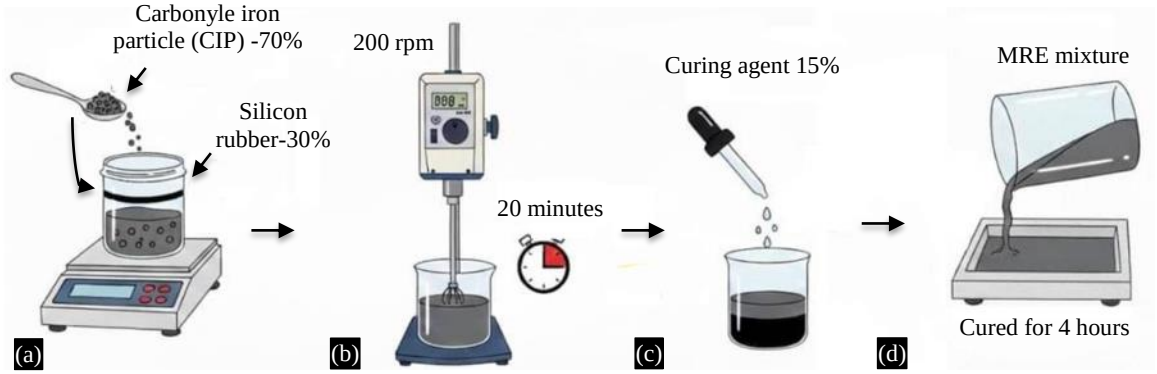


Figure 1. Schematic illustration of the fabrication and experimental procedure of MRE samples: (a) mixing of SR matrix with CIPs, (b) both components were stir using mechanical stirrer (c), adding curing agent to the homogenous mixture, and (d) final mixture then was poured into mould.

Subsequently, the curing agent was added to the mixture and stirred thoroughly to initiate crosslinking. The prepared mixture was poured into a mould with specified dimensions and subjected to curing at room temperature for a fixed duration. In this study, the samples were fabricated under isotropic condition ensuring consistent microstructural configuration across all specimens. After curing, the samples were demoulded and trimmed to remove access material. The sheet was then cut into strips of 35mm in width and 70 mm in length prior to low temperature exposure process.

Sub-Zero Temperature Conditioning

To investigate the effect of low temperature on viscoelastic behaviour, the MRE samples were subjected to controlled sub-zero temperature conditioning. The samples were placed in a low-temperature chamber capable of maintaining stable temperatures within the range of -15 °C, -20ºC to -25 °C, as shown in Figure 2. This selected temperature range represents typical sub-zero environmental conditions encountered in cold-region applications.

The samples were conditioned at the designated temperatures for five days per temperature to ensure that thermal equilibrium was achieved throughout the material. This prolonged exposure was intended to simulate realistic environmental conditions and to provide sufficient time for temperature-induced microstructural and molecular changed to develop within the elastomer matrix. After temperature conditioning, the specimens were allowed to equilibrate at room temperature prior to testing. In accordance with ASTM D832-07, a minimum of 16 hours was maintained between curing and testing to ensure consistent samples conditioning. This experimental procedure enables the assessment of temperature-induced changes in the viscoelastic behavior of the MREs, including variations in stiffness, damping characteristics and potential microstructural alterations resulting from the thermal exposure.

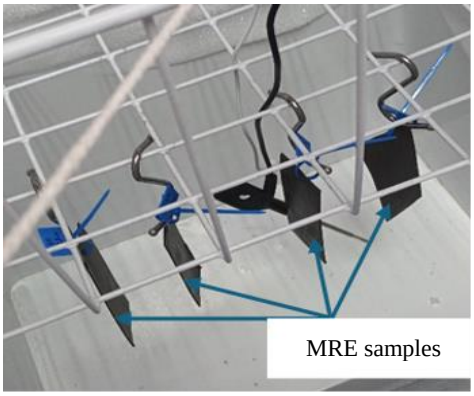


Figure 2. MRE samples placed in a low-temperature chamber for sub-zero conditioning.

Hardness on Shore-A durometer

Hardness is a fundamental physical characteristic of elastomeric materials and magnetorheological elastomers (MREs) that essentially serves as a measure of the material's modulus and overall stiffness [5]. Typically evaluated using a Shore A durometer in accordance with testing standards such as ASTM D2240, this metric provides a direct indication of a material's flexibility[5–11]. A lower hardness value signifies a softer material with superior elasticity, whereas a higher value indicates a more rigid and less flexible structure. As environmental temperatures drop to sub-zero levels below the elastomer's glass transition temperature, the thermal motion of the oscillating polymer chains is severely restricted, causing the material to transition into a brittle, glassy state that exhibits significantly increased hardness and strength[16].

Rheological Characterization

The viscoelastic properties of the MRE samples were characterized using a rotational rheometer (Physica MCR 302, Anton Paar, Graz, Austria) under oscillatory shear mode. The measurements were conducted using a parallel-plate geometry with a constant gap setting appropriate for the sample thickness. A strain sweep test was performed to evaluate the G′ of the MRE samples as a function of shear strain. The strain was varied over a defined range to identify the linear viscoelastic (LVE) region and to observe the strain-dependent behaviour of the material. The G′₀ was determined from the plateau region within the LVE range at very low strain, representing the intrinsic stiffness of the material prior to structural breakdown.

All rheological measurements were conducted at 25°C, using the rheometer's temperature control system. This allowed direct evaluation of temperature-dependent viscoelastic behaviour. To investigate the MR effect, measurements were carried out under both off-state (without magnetic field) and on-state (with magnetic field) conditions. For the on-state measurements, an external magnetic field was applied using an integrated current supply system, with the applied current varied from 0, 1, 2, 3, 4 and 5A, equivalent to magnetic field under discrete magnetic field conditions of 0, 0.18, 0.38, 0.57, 0.72 44 and 0.83 T. The influence of magnetic excitation on G′ was analysed by comparing the response under different current levels. The obtained data were used to assess the effects of temperature and magnetic field on the stiffness and field-responsive behaviour of the MRE samples, with particular emphasis on the variation of G′ under different exposed low temperature of -15ºC, -20ºC and -25ºC conditions.

Morphological Characterization

To support the interpretation of rheological results, the microstructure of the MRE samples was examined using a low vacuum scanning electron microscope (LVSEM), model JEOL IT300LV, Akishima, Tokyo, Japan. Scanning electron microscopy (SEM) was employed to observe the surface morphologies of the samples before and after exposure to low temperature. The samples were cut and coated with a thin conductive layer prior to observation. The SEM analysis was used to evaluate the dispersion of magnetic particles, interfacial bonding, and any microstructural changes induced by sub-zero temperature exposure. This analysis provides insights into the relationship between microstructural features and viscoelastic behaviour, particularly in terms of particle distribution, void formation, and interfacial integrity.

Differential Scanning Calorimetry (DSC)

To measure how a material's physical properties change as a function of temperature and time, the powerful thermal analysis technic using Differential Scanning Calorimetry (DSC), model NETZSCH DSC 214 Polyma, Selb, Bavaria, Germany. To evaluate the thermal properties, the samples were heated from -150°C up to ambient temperature at a steady rate of 10 °C/min. Equipped with an oval Arena furnace, a high sensitivity Corona sensor, and integrated liquid nitrogen cooling, it provides the low thermal inertia and rapid temperature control necessary to accurately capture the sub-zero transitions of silicone, starting from its exceptionally low glass transition temperature, up to its complete melting point. When testing a dense, heterogeneous MRE sample containing a high weight percentage of magnetic carbonyl iron particles (SIPs), the system's specialized Concavus aluminum pans ensure optimal flat thermal contact. This analysis provides the map for material's thermal transition to define its mechanical and magnetic operating limits

RESULTS AND DISCUSSIONS

This section presents the experimental findings and discusses the impact of sub-zero temperature conditioning on the silicone-based magnetorheological elastomers (MREs). First, the macroscopic physical changes of the MRE samples are evaluated through Shore A hardness testing. This is followed by a detailed rheological analysis focusing on the storage modulus (G′) and the magnetorheological (MR) effect under varying temperatures and magnetic fields. To correlate these macroscopic behaviors with internal structural changes, microstructural observations via scanning electron microscopy (SEM) are discussed, highlighting the interfacial effects and defects caused by thermal mismatch. Finally, the thermal transitions and overall thermal stability of the MREs under extreme cold are analyzed using Differential Scanning Calorimetry (DSC)

Effect of Sub-Zero Temperatures on Hardness Values

Table 1 presents the variation in hardness of the MRE samples, measured using the Shore A durometer, at different temperatures ranging from 25°C to −25°C. This table illustrates the response of the material's surface resistance to progressively decreasing temperatures, representing extreme environmental conditions. Using the Shore A hardness scale, the data captures the change in surface rigidity as the material transitions from room temperature to sub-zero conditions. At 25°C, the material exhibits its lowest hardness value 32, indicating a relatively more compliant and pliable state. As the temperature decreases to −25°C, the hardness increases to 37, reflecting a stiffer material behaviour.

The results demonstrate a clear relationship between temperature reduction and increased hardness, suggesting that the material becomes progressively more rigid as thermal energy within the elastomer matrix decreases. This behaviour can be attributed to restricted polymer chain mobility and enhanced particle-matrix interactions at lower temperatures, which contribute to higher resistance against indentation [13]. The increase in hardness is not strictly linear across the entire temperature range. When the temperature decreases from 25°C to −15°C (a 40°C reduction), the hardness increases by 3 units (from 32 to 35). In contrast, within the lower temperature range from −15°C to −25°C, the hardness increases more consistently, with an increment of approximately 1 unit for every 5°C decrease (from 35 to 36 at −20°C, and to 37 at −25°C). This indicates a more pronounced sensitivity of the material to temperature changes at lower temperatures [11].

Effect of Sub-Zero Temperature on Storage Modulus G′

The G′ represents the elastic response of MREs and serves as a key indicator of material stiffness under oscillatory loading[15–17]. Figure 3 illustrates the variation of G′ as a function of shear strains after exposed to different temperature conditions, including room temperature (25°C) and sub-zero temperatures (-15°C to -25°C).

As shown in Figure 3, the G′ increases noticeably with decreasing temperature, indicating enhanced stiffness of the MRE under sub-zero conditions. At room temperature, the material exhibits a relatively lower G′, reflecting greater molecular mobility and a more compliant viscoelastic response. This is due to residual stress caused by prolonged cold exposure. In contrast, at -15 °C and -25 °C, the modulus increases significantly across the entire strain range, demonstrating a transition toward a stiffer and more elastic-dominant behaviour. This behaviour can be attributed to the restriction of polymer chain mobility within the elastomer matrix at lower temperatures [18]. The reduction in thermal energy limits segmental motion, thereby increasing resistance to deformation. In addition, the embedded magnetic particles act as rigid inclusions within the matrix, enhancing load transfer and contributing to the overall increase in G′ The combined effect of reduced chain flexibility and strengthened particle–matrix interactions lead to a more pronounced elastic response at sub-zero temperatures.

Table 1. Variation of shore a hardness of MRE samples under sub-zero temperature conditions.

	Hardness			
Temperature	25ºC	-15ºC	-20ºC	-25ºC
Shore A (Average)	32	35	36	37
SD ±	0.45	0.15	0.53	0.63

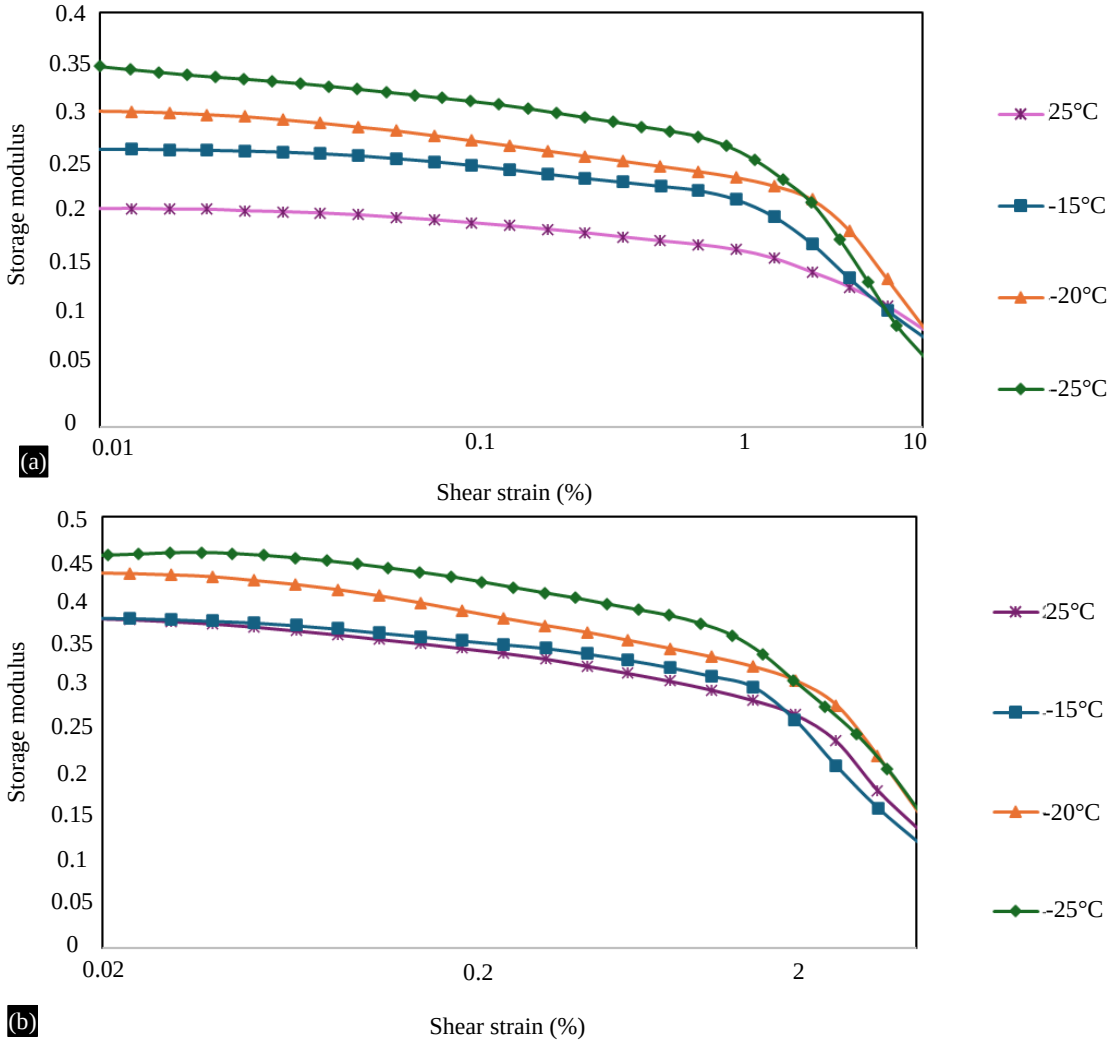
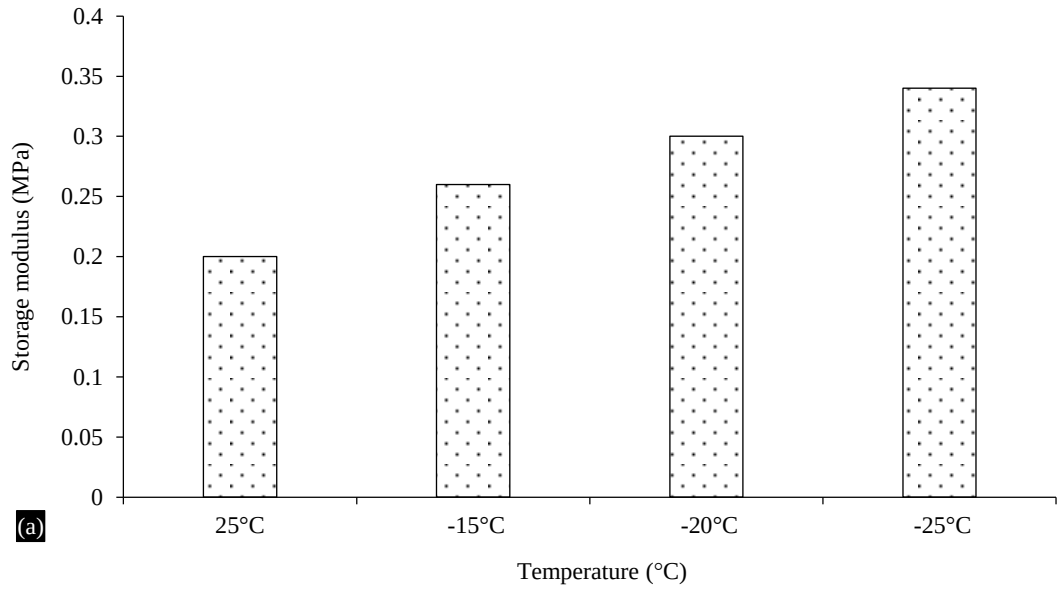
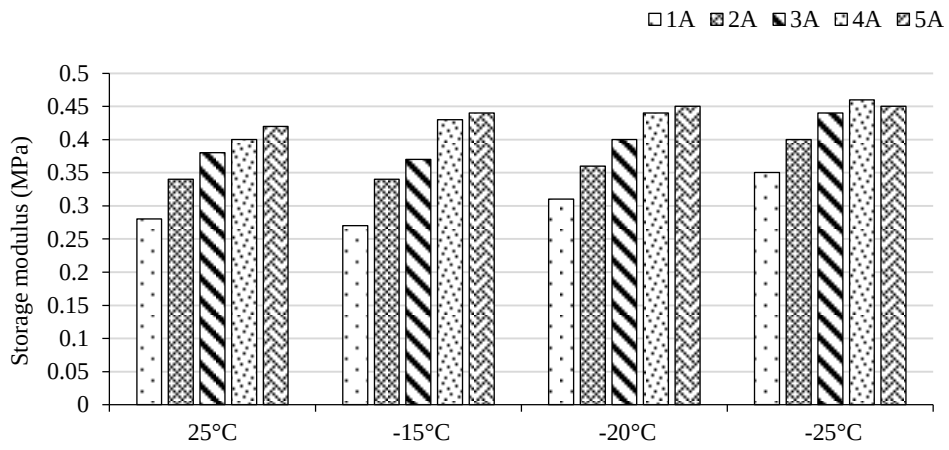


Figure 3. G′ of MRE samples as a function of shear strain after exposed different temperature conditions. (a) off state G′ versus shear strain (b) on state G′ versus shear strain.





(b)

Figure 4. G′₀ of MRE samples as a function of temperature under (a) off-state and (b) on-state conditions at different applied currents.

From the strain-dependent perspective, G′ generally decreases with increasing shear strain, indicating the typical softening behaviour associated with the breakdown of internal microstructures, such as particle chains and filler networks. This reduction is more evident at higher strain levels, where the internal structure of the MRE is progressively disrupted. However, even at elevated strains, the samples tested at lower temperatures maintain higher modulus values compared to those at room temperature, confirming the dominant influence of temperature on stiffness enhancement. The observed trend highlights the sensitivity of MRE stiffness to thermal conditions, particularly under sub-zero environments, where increased rigidity may be beneficial for load-bearing applications but could influence flexibility and adaptability under large deformation.

Building on the strain-dependent behaviour discussed in Figure 3, further insight can be obtained from the G′₀ extracted from the strain sweep curves under both off-state and on-state conditions, as presented in Figure 4. From the strain sweep results, the G′₀ taken within the LVE region at very low strain, was determined for each temperature condition. This parameter represents the inherent stiffness of the material prior to any structural breakdown and provides a direct comparison of the material's elastic response under different thermal environments[19]

The data clearly illustrate that the G′₀ increases as the temperature decreases from 25 °C to -25 °C, which is consistent with the trends observed in Figure 3. At lower temperatures, the MRE exhibits higher stiffness even at minimal deformation, indicating a stronger resistance to elastic deformation. This behaviour reflects the reduced mobility of the elastomer matrix and enhanced effectiveness of stress transfer between the matrix and embedded magnetic particles[20]. In addition, the comparison between off- and on-state conditions highlights the influence of the applied magnetic field on the material response. Under the on-state condition, the G′₀, is consistently higher than that of the off-state across all temperatures. This increase is attributed to the formation of particle chain structures aligned with the magnetic field, which reinforce the internal microstructure and enhance stiffness [21].

The effect of magnetic field strength is further demonstrated through the variation of applied current from 1 to 5 A. As the current increases, the G′₀ shows a progressive increase, indicating a stronger MR effect. This trend becomes more pronounced at lower temperatures, where the combined influence of reduced matrix flexibility and strengthened particle interactions results in a more significant enhancement in stiffness. These observations confirm that both temperature and magnetic excitation play a critical role in governing the initial elastic response of MREs, with sub-zero conditions amplifying the stiffness and the effectiveness of magnetic field-induced structuring.

Effect of Sub -Zero on Magnetic Flux

Following the analysis of G′ under varying temperature and magnetic field conditions in the previous section, further evaluation was carried out to quantify the MR effect of the MRE samples. Figure 5 presents the variation of G′ with magnetic field under different temperature conditions, while Table 2 summarizes the corresponding absolute and relative MR effect values.

The data provide a quantitative assessment of how temperature influences the magnetically induced stiffness enhancement of the MRE system. The MR effect, derived from the difference between on-state and off-state G′, reflects the material's ability to respond to an excitation magnetic field through changes in its internal structure and mechanical properties. As shown in Table 2, both absolute and relative MR effects exhibit a general decreasing trend as the temperature is reduced from 25 °C to -20 °C. The absolute MR effect decreases from 0.18 at 25 °C to 0.22 at -15 °C, and further to its minimum value of 0.21 at -20 °C. A slight decrease is observed at -25 °C, where the value rises to 0.20. A similar trend is observed for the relative MR effect, which decreases from 202% at 25 °C to 90% at -20 °C, followed by a modest recovery to 89.84% at −25 °C.

This behaviour indicates that although the overall stiffness of the material increases at lower temperatures (as discussed in Section 3.2), the relative contribution of the magnetic fields to stiffness enhancement becomes less pronounced. At reduced temperatures, the elastomer matrix becomes inherently stiffer, limiting the additional structuring effect induced by magnetized CIPs. Although the particles remain magnetically responsive, the constrained particle–matrix interactions limit the material's ability to undergo field-induced microstructural deformation, resulting in a smaller change in stiffness and consequently, reduced the MR effect. The particles movement and reorientation under strong magnetic excitation may partially compensate for the reduced matrix flexibility [22]. This interplay between matrix stiffness and magnetic particle interaction governs the overall MR performance under the sub-zero conditions. These findings demonstrate that the temperatures not only affects the baseline rheological properties of MREs but also influences their field-responsive behaviour, which is critical for applications requiring reliable performance under varying environmental and operational conditions.

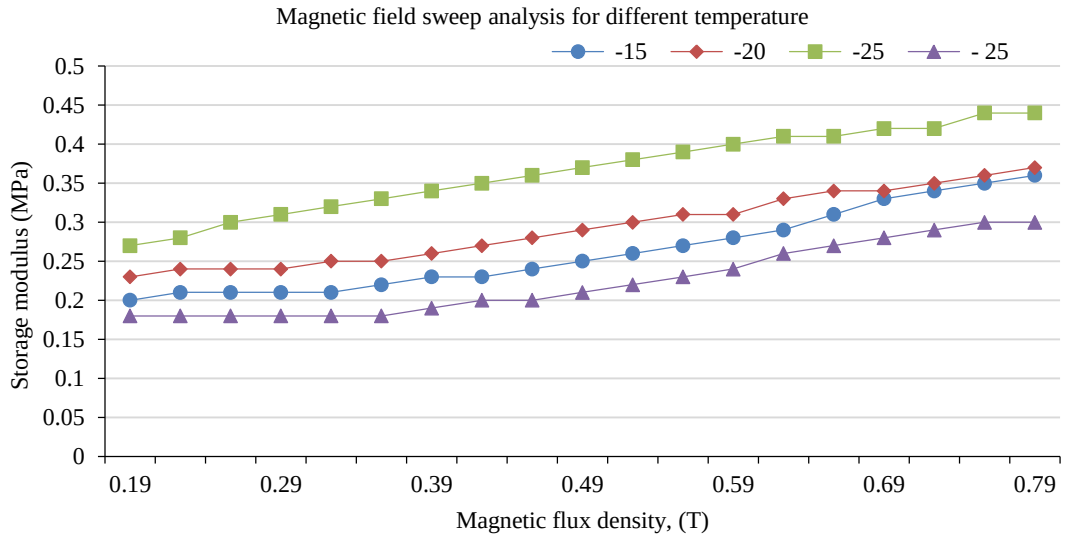


Figure 5. G′ of MRE samples as a function of magnetic field under different temperature conditions.

Table 2. Absolute and relative MR effect of MRE samples at different temperature conditions.

	Temperature			
	25ºC	-15ºC	-20ºC	-25ºC
Absolute MR effect	0.18	0.22	0.21	0.20
Relative MR Effect	202	104	90	84

Microstructural Interpretation of Sub-Zero Effects

To further elucidate the mechanisms underlying the observed rheological behaviour, microstructural analysis was conducted on the MRE samples before and after sub-zero temperature exposure. The SEM observations, as shown in Figure 6, provide important insights into the evolution of particle–matrix interactions and structural integrity under extreme thermal conditions.

At room temperature, the MRE samples exhibit a relatively uniform distribution of CIPs embedded within the silicone rubber matrix. The particles appear well dispersed with minimal agglomeration, indicating effective mixing and good interfacial adhesion between the filler and matrix. This uniform microstructure facilitates efficient stress transfer under mechanical loading and contributes to the stable viscoelastic response as observed for MRE sample at 25°C. Following exposure to sub-zero temperatures (-15°C to -25°C), noticeable microstructural changes are observed. The formation of wrinkles and small holes becomes evident, particularly at lower temperatures [11]. These features are indicative of thermally induced internal stresses arising from the mismatch in thermal contraction between the rigid magnetic particles and the comparatively flexible elastomer matrix. As the temperature decreases, the elastomer undergoes volumetric contraction, while the embedded particles remain relatively dimensionally stable, resulting in interfacial stress concentration [23]. In addition, the reduced molecular mobility of the polymer chains at sub-zero temperatures limits the material's ability to relax these induced stresses. Consequently, stress accumulation at the particle–matrix interface promotes debonding and small holes initiation. The presence of wrinkles and surface irregularities further suggests localized deformation and structural instability within the matrix. These microstructural defects collectively contribute to the observed increase in stiffness and hardness, as the material transitions into a more rigid and constrained state.

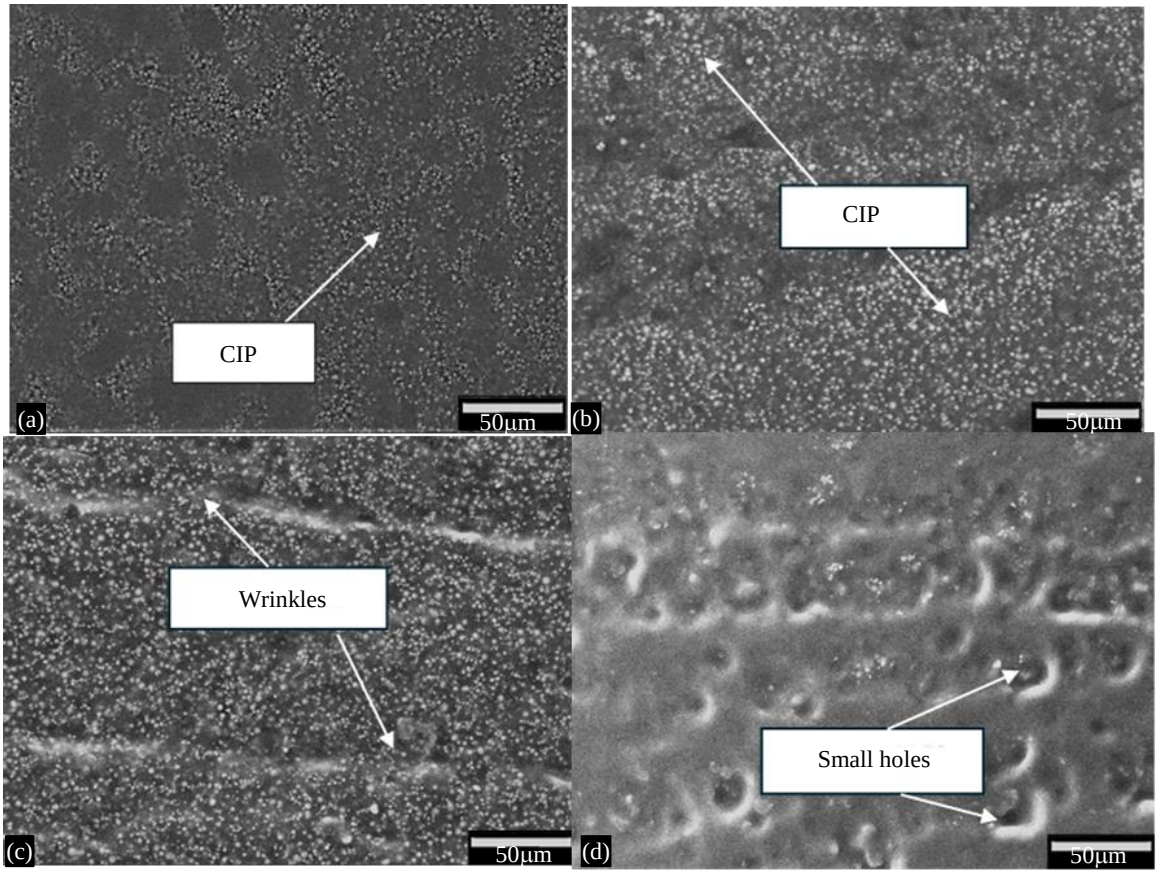
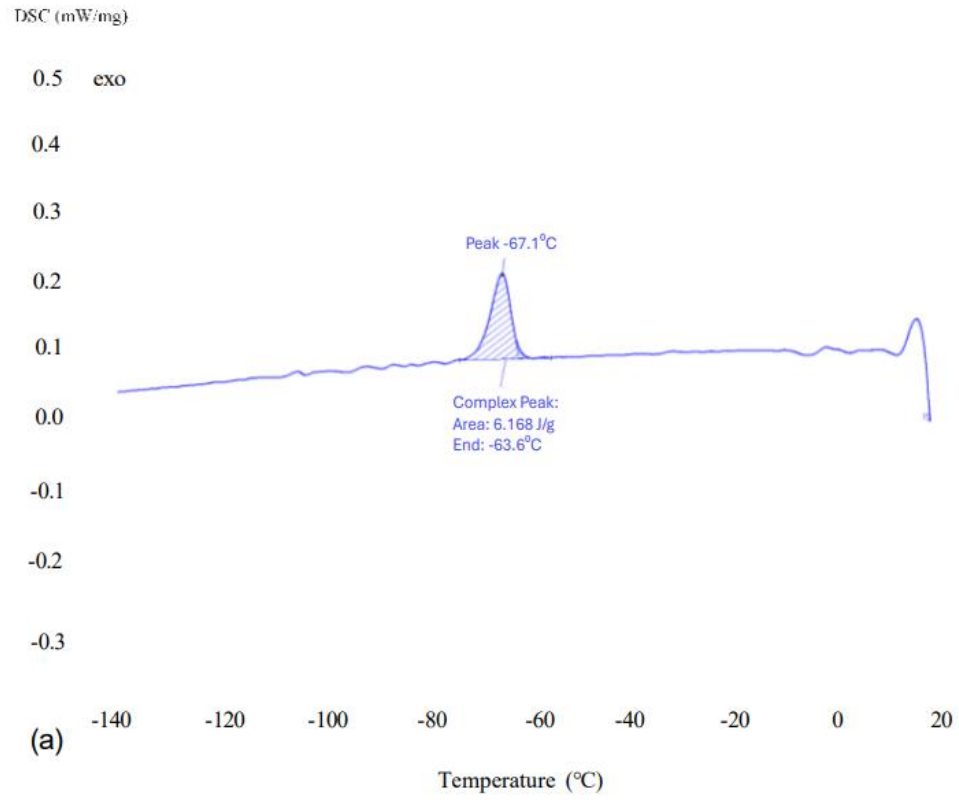
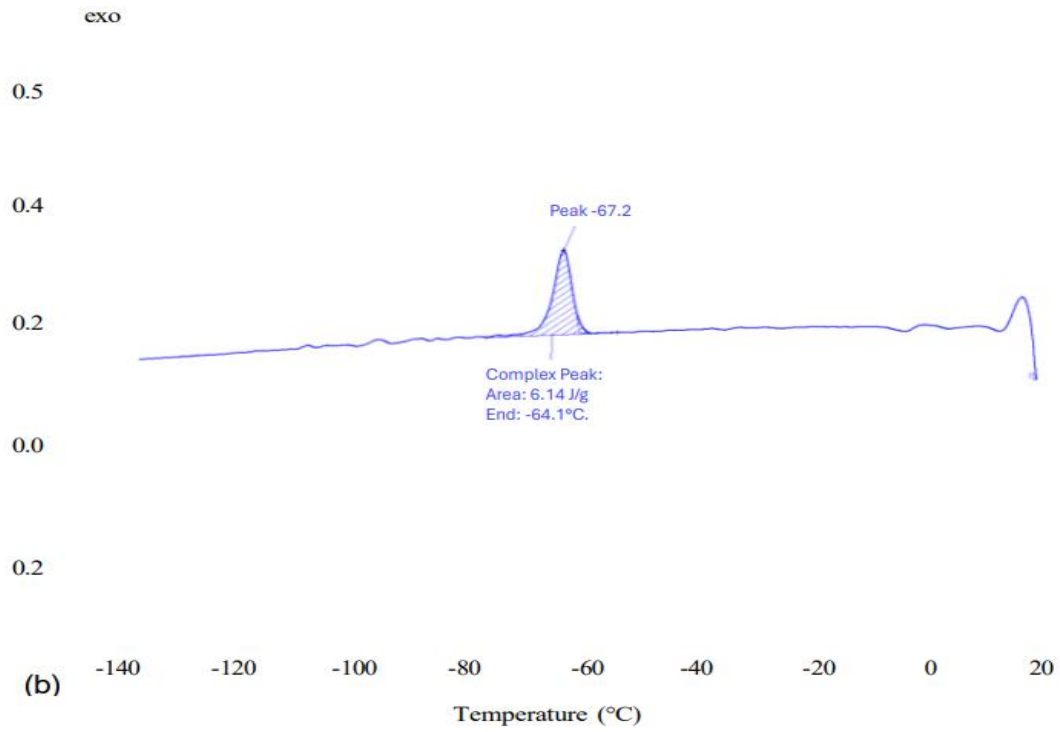


Figure 6. Microstructural characteristics of MRE samples under different temperature conditions; (a) 25°C (b) -15°C (c) -20°C (d) -25°C.





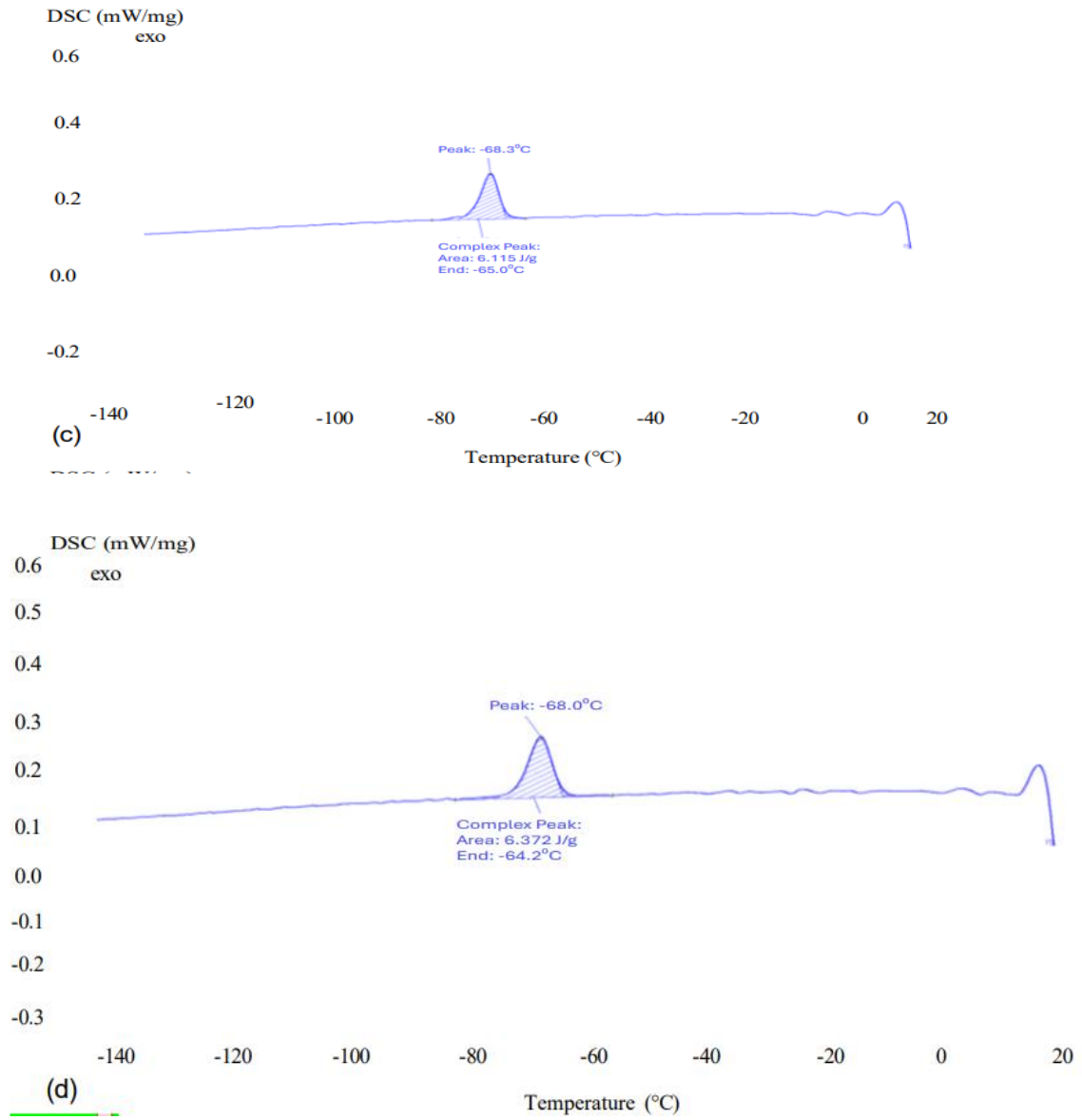


Figure 7. DSC profiles of MRE samples under different temperature conditions; (a) -25°C (b) -20°C (c) -15°C (d) -25°C.

The development of wrinkles and small holes also has significant implications for the viscoelastic behaviour of the MRE. The restriction of polymer chain mobility, combined with interfacial debonding, reduces the material's ability to dissipate energy effectively. This supports the rheological findings, where an increase in G′ is accompanied by a reduction in the relative magnetorheological (MR) effect at lower temperatures. The stiffer matrix limits the reorientation and chain formation of magnetic particles under an applied field, thereby diminishing the field-induced enhancement in modulus. This suggests a competing mechanism between matrix stiffening and particle interaction that governs the overall MR performance under extreme cold conditions. The microstructural observations provide strong evidence that sub-zero temperature exposure induces interfacial degradation, wrinkles formation and structural heterogeneity within the MRE system. These changes directly influence the macroscopic mechanical and rheological behaviour, establishing a clear correlation between microstructural evolution and performance. The findings highlight the critical role of interfacial integrity and matrix flexibility in maintaining the functional reliability of MREs in cold environments.

Table 3. Cold crystallization peak and area of complex peak of MRE samples at different temperature conditions.

	Temperature			
	-25ºC	-20ºC	-15ºC	25ºC
Cold Crystallization Peak	-67.1°C	-67.2°C	-68.3°C	-68.0°C
Area of Complex Peak	6.168J/g	6.14J/g	6.115J/g	6.372J/g

Effect of Sub Zero on Thermal Transitions

Based on Differential Scanning Calorimetry (DSC) graph shown in Figure 7, it is important to note the direction of heat flow: the y-axis has an upward arrow labeled "↑ exo", meaning that exothermic processes (which release heat) will appear as upward peaks, while endothermic processes (which absorb heat) will appear as downward peaks. The Glass Transition Temperature (Tg) is not typically a peak, but rather a step-like shift in the baseline representing a change in heat capacity. On this graph, there is a slight baseline shift on the left side, starting between approximately -120 °C and -110 °C. Crystalline Melting Peak is an endothermic process, which will point downward on this graph. At the far-right edge of the plot, right around +20 °C, the curve takes a sharp dip downward. This endothermic dip indicates the beginning of the crystalline melting peak, though the test appears to end before the full melting peak can be drawn.

The provided Differential Scanning Calorimetry (DSC) data illustrates the cold crystallization process across various tested conditions. Because crystallization is an exothermic event that releases heat, it is represented on the graphs by the prominent, blue shaded upward peaks. As summarized in Table 3, the peak temperatures for this cold crystallization occur at -67.1°C, -67.2°C, -68.3°C, and -68.0°C. Specifically, the data details the enthalpy, or the area under the peak, and the end temperature for each condition: for the -25 °C condition, the enthalpy is 6.168 J/g with the event concluding at -63.6 °C; for the -20 °C condition, the enthalpy is 6.14 J/g, ending at -64.1 °C; for the -15 °C condition, the enthalpy is 6.115 J/g, concluding at -65.0 °C; and finally, for the 25 °C condition, the enthalpy is 6.372 J/g, and the event concludes at -64.2 °C.

Based on the data, the inclusion of carbonyl iron particles (CIP) helps maintain the flexibility and high stability of the amorphous polymers across different temperature exposures. Magnetorheological elastomer (MRE) samples demonstrate exceptional stability at low temperatures and do not exhibit strong crystallization behavior. Furthermore, the samples show nearly identical thermal transitions, with their glass transition remaining completely unchanged. This indicates that sub-zero aging has no significant effect on the material's thermal properties, ultimately proving that the MRE possesses good overall thermal stability without undergoing any significant degradation or phase changes during thermal exposure

CONCLUSION

This study systematically investigated the effects of sub-zero temperature on the viscoelastic behaviour, MR performance and microstructural characteristics of silicone-based MREs. The findings clearly demonstrate that temperature plays a critical role in governing both the rheological response and field-dependent properties of MRE systems. A significant increase in stiffness was observed as the temperature decreased from 25°C to -25°C, as evidenced by higher hardness values and G′. This behaviour is primarily attributed to the restriction of polymer chain mobility and the enhanced load transfer between CIP and the elastomer matrix under low-temperature conditions. The G′₀ further confirmed that the intrinsic stiffness of the material increases substantially at sub-zero temperatures. Despite this enhancement in baseline stiffness, the MR effect showed a decreasing trend 202% at 25°C to 84% at -25°C. The reduced MR effect indicates that the ability of magnetic particles to move and reorient under an applied magnetic field is constrained by the increased rigidity of the Si matrix. This highlights a key trade-off between stiffness and field-induced tunability in cold environments. Microstructural analysis revealed the formation of wrinkles, interfacial debonding and localized small

holes after sub-zero exposure, which are attributed to thermal mismatch between the elastomer matrix and embedded particles. These defects contribute to increased rigidity while potentially compromising durability and energy dissipation capability. Overall, this study provides important insights into the thermal, rheological and microstructural behaviour of MREs under sub-zero conditions. The results emphasize the need for optimized material design, particularly in enhancing interfacial bonding and matrix flexibility, to maintain MR performance in cold environments. These findings are valuable for the development of reliable MRE-based devices in applications such as automotive systems, aerospace components and cold-region engineering.

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