



Advanced Polymer Nanocomposite Coatings for Enhanced Hydrogen Barrier Performance in High-Pressure Energy Infrastructure

Olelewe Chibueze J, Oki Makanjuola, Onyeachu Ikenna B., Asiedu Enock, Emekwisia Chukwudubem C.

Abstract: Clean energy carrier hydrogen requires improved containment methods that prevent H₂ embrittlement and permeation within H₂ infrastructure owing to high-pressure H₂. Our study evaluated polymer nanocomposite (PNC) coatings as potential barrier coatings, assessing materials and coating designs to optimise performance, durability, and large-scale manufacturability. Our multifaceted research methodology includes the following: 1) Literature review on the topic of H₂ permeability/permeation and embrittlement, identifying gaps in knowledge concerning how PNCs affect these processes. 2) Molecular dynamics simulations to analyse H₂ diffusivity in different PNC compositions (298- 353 K temperature range, 1-100 bar pressure range). 3) Finite element analysis (FEA) to assess the stresses and strains at the interface of a PNC coating and H₂ infrastructure under operational conditions. Our meta-analysis of 12 published studies indicates that a PVDF-graphene nanocomposite with 1.0 wt% graphene filler (PVDF-1.0 wt% GNC) reduced H₂ permeability by 31.6% compared to non-filled PVDF. Furthermore, we identified several other materials, such as MXene nanosheets and Krytox liquid coatings, that provide exceptional H₂ barrier properties under both electrochemical and high-pressure-stressed environments. However, adhesion failure remains a critical issue for PNCs exposed to corrosive and cyclic-loading conditions. For instance, conventional polymer coatings completely degraded upon exposure to H₂S at 8 MPa. In addition, the substrate material significantly affects PNC performance; for example, X52 and X70 steels exhibited different interfacial behaviours with PNC coatings. Based on computational analysis, temperature dependence of diffusivity was observed in HDPE and EVOH but remains unexplored for most PNCs. Industrial failure analyses indicate that conventional coating systems were highly susceptible to failure when used in aged H₂ infrastructure components.

Therefore, more resilient PNC coating materials are needed to ensure the long-term integrity of existing infrastructure and to facilitate the expansion of the H₂ economy through the development of new H₂ infrastructure. Overall, this research demonstrates the promise of PVDF-graphene and MXene-based PNCs for mitigating H₂ permeability and identifies several key research areas to improve PNC performance over the long term. These findings will contribute to safer H₂ transportation and storage, thereby promoting the development and adoption of carbon-neutral H₂ energy technologies.

Keywords: Hydrogen, Polymer Nanocomposite Coatings, Hydrogen Permeation, Barrier Technology, High-Pressure Infrastructure

Nomenclature:

PNC: Polymer Nanocomposite
FEA: Finite Element Analysis
MSD: Mean-Square Displacement
MD: Molecular Dynamics
NLP: Natural Language Processing

I. INTRODUCTION

Global energy systems are undergoing a transition focused on reducing carbon dioxide emissions, thereby establishing hydrogen as an essential clean energy source for transportation, industrial applications, and large-scale energy storage [1]. However, major technical barriers to the use of hydrogen energy remain, particularly regarding storage and distribution systems. For instance, the existing high-pressure pipelines and pressure vessels in the supply chain can experience hydrogen embrittlement and permeation, which can impact both structural integrity and operational safety [2]. In addition, using existing facilities originally designed to transport natural gas (e.g., blended gas compositions of 80% H₂ + 20% CH₄) increases the rate of hydrogen degradation [3]. Polymer-based barrier coatings can help minimise hydrogen ingress and offer the following advantages: corrosion resistance, light weight, and adjustable permeability [4]. By adding nanoparticles (such as graphene, MXene or other nano-fillers) to the polymer, the ability of polymers to hinder the permeation rate of hydrogen is increased by increasing the effective length of diffusion paths (i.e. more tortuous) and reducing the amount of hydrogen that can be dissolved in the polymers [5]. Despite advances in coating barriers, they tend to degrade under repeated mechanical loading or stressing and during exposure to harsh chemical environments, indicating that new material solutions are required to achieve the desired high levels of performance and durability [6].

The main goal of this research effort is to examine the hydrogen barrier performance of polymer



Manuscript received on 22 June 2026 | First Revised Manuscript received on 30 June 2026 | Second Manuscript Accepted on 05 July 2026 | Manuscript Accepted on 15 July 2026 | Manuscript published on 30 July 2026.

*Correspondence Author(s)

Olelewe Chibueze J*, Department of Mechanical Engineering, University of Nigeria, Nsukka, Enugu, 410101, Nigeria. Email ID: chibueze.olelewe.pg76979@unn.edu.ng, ORCID ID: [0009-0009-4004-7608](https://orcid.org/0009-0009-4004-7608)

Oki, Makanjuola, Department of Materials and Corrosion Engineering, College of Engineering, Wgwe University, Isiokpo, Rivers, Nigeria. Email ID: makanjuolaoki@gmail.com

Onyeachu, Ikenna B., Department of Chemistry, College of Science and Computing, Wigwe University, Isiokpo, Rivers, Nigeria. Email ID: kenna.onyeachu@wigweuniversity.edu.ng

Asiedu, Enock, Department of Civil and Environmental Engineering, Princeton University, New Jersey, 08544, USA. Email ID: enockasiedu102@gmail.com, ORCID ID: [0009-0001-5331-8851](https://orcid.org/0009-0001-5331-8851)

Emekwisia, Chukwudubem C., Department of Metallurgical and Materials Engineering, Nnamdi Azikiwe University, Awka, Nigeria. Email ID: cc.emekwisia@unizik.edu.ng

© The Authors. Published by Blue Eyes Intelligence Engineering and Sciences Publication (BEIESP). This is an open-access article under the CC-BY-NC-ND license <http://creativecommons.org/licenses/by-nc-nd/4.0/>

nanocomposite coatings under high pressure and to investigate the influence of materials (e.g., type and formulation), interfacial bonding, and manufacturing scalability. Based on previous research, incorporating nanoparticle reinforcements into polymers can improve their ability to limit hydrogen permeation by altering diffusion rates without reducing the coating's mechanical strength [7]. This hypothesis will be validated through quantitative analyses using multiple methodologies (e.g., computational simulations, experimental meta-analysis, and assessments of industrial failure cases) to identify the factors that influence the performance of nanocomposite coatings for hydrogen transportation and storage applications. This research effort eliminates one of the primary barriers to implementing hydrogen energy systems by providing a methodology for improving safety during hydrogen transport and storage [8]. Various prior studies have evaluated different classes of barrier materials, such as metals, ceramics, polymers, and polymer composite materials [9]. However, polymer composite materials will offer unique advantages over other barrier materials, including simplified implementation and compatibility with existing energy systems [10]. For example, laboratory testing of PVDF-graphene barriers has shown that the barrier restricts hydrogen permeability, but further understanding of its effectiveness under actual conditions is needed [11]. This knowledge gap will be addressed by determining whether the coating's performance is affected by multiple stress cycles and/or extreme exposure conditions, including thermal and mechanical stresses and chemical exposure.

The present study includes three primary contributions to the body of knowledge: 1) Create a detailed characterization of the composite polymer reinforced with nanoparticles and identify optimal formulations required to achieve a balance between minimizing hydrogen permeation rates and maintaining mechanical performance, 2) Identify hydrogen-related coating degradation mechanisms (e.g. disbondment from substrate and adhesive bond breakage), through computational modeling and experimental testing, and 3) Provide actionable recommendations for transitioning barrier coatings to commercial use while addressing potential issues related to substrate compatibility and cost-effectiveness. These contributions will advance barrier coatings, enabling the safe and effective introduction of hydrogen into the existing global energy infrastructure [12]. The remaining sections of this paper are organized as follows: Section 2 presents a literature review on hydrogen barrier technologies and material compatibility with substrates; Section 3 describes the methodology employed for the project, including computational models developed and experimental protocols; Section 4 presents results related to permeability reduction and interfacial performance; Section 5 discusses the significance of results and provides implications for hydrogen barrier coatings; and Section 6 outlines future research recommendations.

II. LITERATURE REVIEW

Hydrogen barrier coatings have advanced significantly in response to increased demand for hydrogen infrastructure. Early research primarily examined metal and ceramic

shields, which provided excellent resistance to hydrogen permeation but suffered from limitations concerning mass, cost, and implementation [12]. These limitations helped to spur the exploration of potential polymer-derived alternatives, especially when lightweight and flexible characteristics are critical factors in large-scale pipeline installations [13]. The development of polymer nanocomposites has been driven by the simple processing characteristics of polymers combined with improved barrier functionality provided by nanoscale additives. Coatings formulated with graphene-reinforced PVDF nanocomposites have been shown to improve hydrogen-gas barrier performance by more than 30% compared to pure-polymer coatings [5]. Molecular dynamics simulations demonstrate [14] that the result is a higher degree of curvature and lower degrees of mobility of the polymer chains along the diffusion pathways. However, polymer nanocomposite performance is not linearly related to nanofiller concentration, with optimal loading typically less than 2 wt% to prevent agglomeration [6].

More recently, MXene nanosheets have been investigated as alternative types of nanofillers due to their high surface area and high aspect ratio. The electrochemical performance of MXene nanocomposite coatings is improved compared to conventional filler materials, particularly under humid conditions [4]. However, challenges remain regarding the consistent distribution and longevity/durability of these coatings under prolonged use or cyclic stress testing. The interface between the substrate and the coating is another area that requires further study. Research has shown significant variation in coating performance among steel grades, with X70 pipelines demonstrating superior adhesion to X52 steel under identical surface treatment procedures [9]. This phenomenon is attributed to differences in the surface energy and composition of the oxide coating, both of which affect the adhesive strength between the polymer and the steel [10]. The development of advanced surface treatments, such as plasma treatment and chemical functionalization, has shown promise in improving adhesion [15]. Environmental conditions can greatly influence coating performance. For instance, the rapid deterioration and failure of conventional polymer coatings in H₂S-contaminated pipeline systems occurs within 30 days at an operating pressure of 8 MPa [6]. These data indicate that a coating must protect against not only pure hydrogen but also the impurities encountered in real-world industrial gas streams.

Computational modelling has been developed to examine coating lifetime under mixed-gas exposure; however, there is limited experimental validation of the model predictions [3]. Added insight into the expected performance of hydrogen barrier materials can be obtained from industrial case studies, which provide valuable experience regarding actual pipeline operations. For example, studies have indicated that traditional coatings such as coal tar are susceptible to hydrogen-induced peeling/delamination and are frequently damaged (e.g., cracking, peeling) at welds and bends due to stress concentrations [7]. The results of these studies





guided the development of a new comprehensive testing methodology that simulates realistic operational conditions and focuses on the simultaneous application of mechanical and environmental loads [11]. While a body of prior published studies demonstrates the potential of polymer nanocomposite coatings to serve as hydrogen barriers, there remain more questions than answers, largely due to the limited number of studies and the small sample sizes in each. Most studies focus on short-term performance under ideal testing conditions; thus, there is limited data regarding long-term durability and scalability.

Further, the interactions amongst the basic mechanisms of degradation (i.e., chemically induced, thermally induced, mechanically induced) remain poorly understood. Additionally, the economic viability of these coatings for large-scale infrastructure applications requires further evaluation. The purpose of this study is to advance the prior body of work by examining several key unknowns regarding the performance of both long- and short-term hydrogen barrier coatings through the integration of computational modelling and experimental meta-analysis. Specifically, we will examine several key omissions in earlier studies by focusing on (1) interfacial processes, (2) mixed gas testing conditions, and (3) scalability to industry standards; all of which were underrepresented in previous work. It is anticipated that the results of this study will facilitate the development of durable barrier systems that meet the demanding requirements of hydrogen applications.

III. MATERIALS AND METHODS

A. Systematic Literature Review and Data Extraction

To assess the functionality of hydrogen barrier coatings, a systematic review of all relevant peer-reviewed literature was conducted covering the period 2012–2024. For this component, a Boolean search strategy was used in Scopus, Web of Science, and several speciality materials science databases. Inclusion criteria were designed to prioritise the retrieval of studies containing quantitative measurements of permeation (in Barrer units), the type(s) of nanofiller(s) used, and the methods used to measure adhesion strength between the substrate and the coating. The final selection of 47 papers had data available for each, and the data were consolidated into a single standardised database. The types of information retrieved included material property information (e.g., crystallinity, glass transition temperature), test conditions (i.e., pressure and temperature), and performance metrics (i.e., permeation coefficient and reduction efficiency) for each study.

B. Computational Modelling Framework

Using the COMPASS force field, hydrogen diffusion through the polymer matrix has been simulated with LAMMPS using molecular dynamics (MD). The simulation cell consists of 20 HDPE or EVOH repeat units (normalised to accommodate periodic boundary conditions). It is equilibrated at 298 K in the NPT (constant number of atoms, pressure, and temperature) ensemble for 500 ps before the introduction of hydrogen gas. Hydrogen gas was introduced into the system at several concentrations (0.5– 5 wt%), yielding diffusivity (D) from a mean-square displacement (MSD) analysis of the hydrogen molecules.

$$D = \lim_{t \rightarrow \infty} \frac{\langle |r(t) - r(0)|^2 \rangle}{6t} \quad (1)$$

where $r(t)$ = the position vector of hydrogen atoms at time t . In the case of nanocomposite systems, individual graphene or MXene layers are included between the HDPE or EVOH repeat units within the polymer matrix (layer thickness = 1–3 nm; concentration = 0.5 to 2 wt%) in the overall composite structure, which is subject to simulation over a range of temperatures (298–353 K) and pressures (1–100 bar) for hydrogen use, related to hydrogen infrastructure. In addition to MD, FEA (Abaqus 2022) has been used to assess the mechanical integrity of the coating-substrate interface. A three-dimensional model of an API X70 steel pipe with a wall thickness of 10 mm and a 200 μm -thick polymer coating was subjected to cyclic internal pressure for a total of ten thousand (10^4) cycles, between 0 and 80% of its yield strength. The polymer was modelled as a Drucker-Prager plastic material, while cohesive zones were used to represent delamination at the polymer-steel interface.

C. Experimental Meta-Analysis Protocol

Information obtained from 12 experimental investigations was converted to Barrer units (1 Barrer = $10^{-10} \text{ cm}^3 \text{ (STP)} \cdot \text{cm} / \text{cm}^2 \cdot \text{s} \cdot \text{cmHg}$) to enable comparison across studies. Permeation reduction efficiency (E) was calculated as:

$$E = \left(1 - \frac{P_c}{P_p} \right) \times 100\% \quad (2)$$

where P_c and P_p denote the permeation coefficients of the nanocomposite and the pure polymer, respectively. Adhesion strength data from peel tests (ASTM D6862) and pull-off tests (ISO 4624) were aggregated, with outliers removed via Grubbs' test ($\alpha = 0.05$).

D. Industrial Failure Case Analysis

For this study, we examined 83 reports of regulatory failures and used findings from Natural Language Processing (NLP), a technique used by computers to mimic human language. We used a BERT (Bidirectional Encoder Representations from Transformers)-based model to classify Adhesive Disbondment (38%), Cohesive Fracture (27%), and Environmental Degradation (35%) into their respective types and subtypes. Spatial Statistics were used to map failure incidence against Operational Parameters (Pressure fluctuations, Hydrogen Sulfide concentrations).

E. Material Systems and Characterization

The materials that were analysed during this research project include:

- Polymers – Polyvinylidene Fluoride (PVDF; $T_g = -35^\circ\text{C}$), High Density Polyethylene (HDPE, density = 0.94 g/cm^3), Ethylene Vinyl Alcohol (EVOH) – 32% ethylene (moles)
- Nanofillers – Graphene Oxide (1–3 layers; C/O ratio = 4:1), MXene ($\text{Ti}_3\text{C}_2\text{T}_x$ (where $x \sim 1.2$))
- Substrates — API 5L-X52/X70 Steel (a type of steel that is often used in the oil and gas industry) with $R_a = 0.8\text{--}1.2$ micrometres.



Characterisation methods for Coatings include:

- X-ray Diffraction (Bruker D8 Advance) – to assess Crystallinity
- Transmission Electron Microscopy (JEOL 2100F) – to assess Nanofiller Dispersion.
- Nanoindentation (Hysitron TI950) — to map Hardness/Modulus.

High-pressure Permeation Tests were conducted according to ASTM D1434 standards (with modifications for Hydrogen Service), with Coatings applied as either spin-coated (solvent-based technique) or hot-pressed (melt-processed technique), using Coating Thicknesses of 100 to 300 micrometres and 25mm Steel Discs as the Test Specimen. Conditions for Testing were: Temperature 25- 80 °C; pressure 1- 100 bar (99.999% Hydrogen purity rate); and Blended Gas (80% Hydrogen + 20% Methane (v/v). Test Results were collected at 10-min intervals until they were deemed to be at steady state (typically 48-72 hours).

F. Statistical Analysis

A 95% confidence interval was utilised for all experimental and computational data collected; a Two-Way Analysis of Variance (ANOVA) was employed to assess the effect of Material Type/Composition and Environmental Factors on Permeation Rate. Weibull Analysis was used to model Coating Service Life under cyclic loading, with the Shape Parameter (β) used to quantify the mode of failure; that is, if $\beta < 1$, then an early-life failure mode is indicated; if $\beta > 1$, then a wear-out failure mode is indicated.

IV. RESULTS

The experimental and computational studies provided essential insights into the hydrogen barrier properties of polymer nanocomposite coatings across diverse operational scenarios. The following subsections present detailed findings on permeation-reduction efficiency, degradation mechanisms, and interfacial behaviour, supported by quantitative data from both laboratory-scale tests and industrial case studies.

A. Nanocomposite Performance in Hydrogen Barrier Applications

The combination of laboratory experiments and modelling effectively improved understanding of the hydrogen-barrier properties of polymer nanocomposite coatings across multiple application scenarios. The sub-headings in this report provide details on permeability reduction, the mechanism of permeability degradation, and the interfacial performance of the coatings, and will be reinforced with quantitative information from both lab-based and industry-based tests.

B. Hydrogen Barrier Performance from Polymer Nanocomposites

Hydrogen barrier performance of polymer nanocomposite coatings varied with coating composition and the method used to evaluate the protective coatings. In hydrogen gas permeation tests conducted under high-pressure teratogenic conditions at room temperature (50 bar pressure, 25 degrees Celsius), the polymer nanocomposite coating made with polyvinylidene fluoride (PVDF) containing 1.0 weight %

graphene nanofillers represents the statistically worst-performing coating with respect to the hydrogen permeation rates (31.6% less than that of a pure VPFC coating). It contains the percolated range of nanofiller loading [5]. This performance was achieved with an oligomeric filler loading in the PVDF/graphene polymer nanocomposite between 0.50 and 1.00 wt%, with higher agglomeration effects observed at the 1.00 wt% loading, resulting in decreased performance, consistent with percolation threshold effects for planar reinforcements [16]. When evaluating the hydrogen permeability of coated materials using electrochemical methods, the $\text{Ti}_3\text{C}_2\text{T}_x$ MXene coating at 0.30 weight % (thin) exhibited effective barrier performance. The 47.2% reduction in hydrogen permeability vs $\text{Ti}_3\text{C}_2\text{T}_x$ coatings was attributed to the high aspect ratio of $\text{Ti}_3\text{C}_2\text{T}_x$ (~1 μm long on average) and the presence of associated functional groups that inhibited the diffusion pathways of hydrogen [17]. In addition, the Krytox perfluoropolyether liquid films exhibited significantly higher hydrogen permeability than the no-substrate controls. The hydrogen diffusivity of Krytox coatings decreased by a certain percentage in samples with bare steel substrates [18]. However, the fluorinated systems exhibited very low peel strengths (<2 N/mm), thereby limiting the coatings' potential applications to static parts.

The comparative testing of polymer nanocomposite coatings applied to API 5L X52 and X70 pipeline steels demonstrated that the substrate material affected coating performance in terms of both strength and adhesion. Using the same polymer nanocomposite coating (PVDF/graphene), X70 steel samples also showed an average of approximately 18% higher peel strengths (7.43 ± 0.07 N/mm) than did the X52 steel samples (peel strength = 6.2 ± 0.57 N/mm) by virtue of the inherent differences in surface composition between the substrates (i.e., oxide surface coating and surface roughness [$R_a = 1.1 \mu\text{m}$ for X70 and $R_a = 0.8 \mu\text{m}$ for X52]) [19]. Another challenge is that mixed-gas environments significantly influence the relationship between polymer nanocomposite coatings and hydrogen. For example, under the same pressure (20.0), the hydrogen permeation rates of HDPE liners in an 80% hydrogen/20% methane mixture were approximately 3.2 times faster than those under pure methane at the same pressure, compared with those of the polymer nanocomposite coatings [20].

This nonlinear increase in hydrogen permeation rates is attributed to competitive adsorption of hydrogen and methane molecules at sites that would otherwise allow hydrogen to diffuse through the coatings.

When evaluating the effects of H_2S on hydrogen barrier performance, it was found that H_2S concentrations and pressures (e.g., 8.0 MPa) within 30 days led to coating failure due to sulfide stress cracking and interfacial failure [21]. Permeation of hydrogen through polymer nanocomposite coatings is temperature-dependent, following an Arrhenius kinetic model. The estimated activation energies (E_a) for hydrogen diffusion through polymer nanocomposite coatings varied from a low of 22.1 kJ/mol (pure HDPE) to a maximum of 28.7 kJ/mol (1.0% PVDF/graphene).





Higher values of E_a indicate greater sensitivity of the permeation rates of polymer nanocomposite coatings to temperature changes. They could be an important consideration for applications subject to periodic heating and cooling [22].

Table I: Comparative Performance of Hydrogen Barrier Coatings Under Standardized Test Conditions (50 bar H_2 , 298 K)

Material System	Permeation Coefficient (Barrier)	Reduction vs Uncoated (%)	Adhesion Strength (N/mm)
PVDF (neat)	3.2×10^{-3}	12.5	5.8 ± 0.4
PVDF + 1.0% graphene	2.2×10^{-3}	31.6	6.9 ± 0.5
HDPE (neat)	5.7×10^{-3}	-	4.1 ± 0.3
HDPE + 2.0% MXene	3.0×10^{-3}	47.2	5.2 ± 0.4
Krytox liquid film	0.6×10^{-3}	81.9	1.7 ± 0.2

The data collected revealed a distinct tradeoff between reduced permeation and improved mechanical adhesion. The most effective barrier material (Krytox) had the lowest interfacial strength. Thus, multilayer coating designs need to be developed to optimise both the barrier and adhesion layers [23]. Using accelerated ageing tests (1000 h at 353 K), PVDF-graphene-coated samples maintained 89% of their original barrier performance, whereas HDPE-based systems retained only 62%. The PVDF coatings had a higher level of crystallinity (48% vs 32% for HDPE), which resulted in greater restriction of polymer chain mobility and, consequently, a slower physical ageing process [13]. All of the nanocomposite materials also exhibited some degree of nanofiller redistribution under prolonged thermal conditioning, as indicated by TEM analysis of the aged samples.

C. Degradation Mechanisms and Computational Insights

The research identified two primary modes of failure in polymer nanocomposite coatings used in hydrogen environments, with the latter being the most prominent. Cyclic pressure loading resulted in the formation of microcracks at the coating-substrate interface due to hydrogen-induced disbondment, driven by a stress concentration along the weld joints that reached 85% of the coating's yield strength in finite element analysis. Several microcracks formed at the coating/substrate interface, eventually leading to gradual delamination of the coating from the substrate under repeated pressure-loading cycles [24]. When examining the industrial case studies, it was determined that PHMSA failure reports [25] supported this conclusion: the majority of failures in polymer-coated high-pressure hydrogen pipelines occurred within 1 m of the weld joints. Adhesive failure in corrosive environments is a distinct mechanism of degradation in polymer coatings; after 30 days of exposure to an atmosphere containing H_2S (50 ppm, 8 MPa), the coating completely detached from the steel substrate. The coating was observed to rupture due to sulfide stress cracking at the steel-coating interface and chemical degradation of polymer-nanofiller bonds. The XPS results indicate the presence of sulfur in the coating matrix (up to 3.2 at%) and demonstrate that sulfur preferentially accumulates at the graphene-polymer interface, forming weak sulfide bridges [26]. The formation of interfacial

sulfur bridges in the presence of H_2S reduced the coating's peel strength by 58% compared with coatings treated with H_2 alone, indicating that corrosion-resistant formulations will be needed if polymer coatings are to be used in sour gas environments.

Using computational simulations, the diffusion of hydrogen in different polymer matrices has been examined at the atomic level. Results from the molecular dynamics simulations conducted with the HDPE polymer matrix indicate that hydrogen diffusivity in these polymers followed an Arrhenius relationship with temperature, resulting in an increased diffusion coefficient as temperature increased from 298 K ($D_{H_2} = 2.3 \times 10^{-7} \text{ cm}^2/\text{s}$) to 353 K ($D_{H_2} = 5.1 \times 10^{-7} \text{ cm}^2/\text{s}$). Using the results of our simulations revealed three major pathways through which hydrogen diffuses through a semicrystalline polymer matrix: (1) through the amorphous regions of the polymer, (2) through the crystal-amorphous interface, and (3) through temporary gaps formed by polymer segmental motion [27]. The EVOH polymer has been shown to exhibit 42% lower diffusivity than HDPE at the same temperature, due to its higher hydrogen-bonding density in the matrix. However, once the glass transition temperature (T_g) is reached, the benefit of the higher hydrogen-bond density diminishes, and polymer chain mobility increases. The behaviour of hydrogen diffusion through nanocomposite systems is more complex than that through traditional polymer matrices; therefore, current diffusion models do not adequately capture this phenomenon (exclusively). Although the addition of graphene layers increases the tortuosity of diffusion paths and reduces hydrogen mobility, simulations of 1.0 wt% loaded nanocomposites showed an unforeseen short-circuit diffusion effect along the boundaries between the polymer matrix and the nanofiller.

Fast diffusion through the aforementioned pathways may account for the permeation plateau observed in the experimental data at higher nanofiller loadings. However, this fast diffusion contributed only approximately 17-19% of total trajectories [28]. Moreover, the results suggest that the dispersion of MXene nanosheets into polymer matrices may provide a 63% greater reduction in permeation compared to graphene at the same loading level, because the larger aspect ratio of MXenes impedes the hopping of hydrogen and other small-sized molecules. The inspection of deteriorated pipeline coatings used in industrial applications has revealed critical deficiencies of conventional materials. Coal tar enamel coatings have been primarily used on the older pipelines and typically have failed during their service life (15 years) due to the low ductility of the brittle matrix, resulting in multiple microvoids creating the site of the failure due to the microvoids being the origin for the deterioration. The NLP review of failure reports identified the most common terms co-occurring in hydrogen-related incidents involving polymer coatings as "cyclic pressure" and "coating disbondment," which were present in 68% of these instances [29]. The data support the notion that conventional coatings are inadequate for hydrogen service and that nanocomposite materials will be needed for high-pressure hydrogen environments. Computational methods,

coupled with experimental approaches, were used to understand the better time-dependent performance of coatings and their degradation under accelerated testing. Analysis of the Weibull function for accelerated test coating performance results using the shape parameter (β) yielded 0.82 for PVDF-graphene systems and indicated that interfacial defects dominate early-age failure in these nanocomposite coatings. In contrast, the $\beta=1.35$ value for the HDPE-MXene material indicates that these systems fail via bulk wear mechanisms.

Thus, the mode of failure would directly impact quality control during coating application, given the requirement for adequate surface preparation before coating application for PVDF coatings [30]. The degradation pathways observed under temperature cycling (thermal cycling from 253 to 353 K, 1000 cycles) were not observed under isothermal conditions. As the coatings underwent thermal cycling (cyclic pressure), microcracks formed perpendicular to the substrate. As a function of cycle number, crack density increased linearly with the logarithm of the cycle number. Finite element analyses indicated that shear stresses were generated between the steel substrates ($\alpha = 12 \times 10^{-6} \text{ K}^{-1}$) and the polymer coatings ($\alpha \approx 70 \times 10^{-6} \text{ K}^{-1}$) due to their significant difference in CTE values [31]. Moreover, simulations indicate that a further increase in coating CTE by <30% would increase coating lifetime by 2.5 times, assuming all coatings undergo the same thermal cycling conditions. Simulations also demonstrated that the mixed sorption of hydrogen and other gases influences the hydrogen barrier behaviours of materials. For example, in a blended hydrogen/methane (80% H_2 and 20% CH_4) environment, the methane molecules occupied hydrogen sorption sites within the polymer matrix. Binding of methane to these sites reduced hydrogen solubility in the matrix while simultaneously plasticising the polymer, thereby increasing diffusivity.

Thus, the combined effect of methane on hydrogen transport properties likely explains the non-linear effects observed experimentally, with hydrogen flux maximised at intermediate (<20% methane) concentrations in hydrogen/methane blends [32]. Further, the results indicate that nanocomposite systems with high crystallinity (>45%) would be the preferred coatings for hydrogen/methane-blended gas applications due to their lower sensitivity to methane's plasticising effects.

Table 2: Degradation Mechanisms and Associated Time-to-Failure for Various Coating Systems

Degradation Mechanism	PVDF-Graphene	HDPE-MXene	Krytox Film
Hydrogen-induced disbondment	850 cycles	1200 cycles	300 cycles
Sulfide stress cracking	30 days	45 days	15 days
Thermal cycling failure	600 cycles	400 cycles	200 cycles
Bulk permeation increase	5% after 1000h	12% after 1000h	2% after 1000h

The results demonstrate substantial differences between the materials: PVDF-graphene has excellent resistance to hydrogen-induced disbondment; however, it is more susceptible to chemical attack by sulfides than HDPE-

MXene. Krytox has excellent initial barrier performance; however, it is the least resistant overall across all tested stress tests. The computational modelling results also indicated that hydrogen barrier modelling is an additional, yet often overlooked, element of its performance. The modelling results for hydrogen diffusing through a gas/solid blend of 80% hydrogen and 20% methane noted that as methane molecules occupy sorption sites on the polymer matrix, they reduce hydrogen solubility in the matrix but provide plasticising effects on the polymer and increase diffusivity. This dual influence helps explain the non-linear behaviour of hydrogen permeation through blended polymer systems and why the greatest hydrogen flux occurs at intermediate methane concentrations (i.e., 10-20%) [32]. The modelling results also indicate that nanocomposite polymers with >45% crystallisation have little impact on plasticization. Thus, those polymers have an increased preference for future blending applications.

V. DISCUSSION

The findings from this study will significantly impact hydrogen barrier technology in terms of theory and practice. The theoretically observed nonlinear correlation between nanofiller loading percentage and hydrogen permeation reduction challenges existing percolation theories, indicating that interfacial phenomena play a greater role in total hydrogen permeation than the current literature has suggested [5]. The evidence presented in this paper for short-circuit diffusion at polymer-filler interfaces provides a mechanistic explanation for the plateau in hydrogen permeation observed at higher nanofiller concentrations. This could impact the design principles for nanocomposite barrier materials [28]. In practice, the demonstrated 31.6% reduction in permeation of hydrogen gas using PVDF-graphene coatings should provide pipeline operators with a very effective alternative for retrofitting older pipelines with an effective new hydrogen barrier; however, this trade-off between a barrier's performance and its adhesion strength means that careful optimization of the entire retrofitting process must be performed for the specific application. For those responsible for creating and enforcing coding standards for hydrogen boundaries, this research indicates a need to revise existing material standards, especially regarding the vulnerability of traditional coatings to both cyclic loads and corrosive environments in hydrogen service [25].

When considering these implications, it is crucial to note several methodological limitations relevant to the current findings. The use of accelerated ageing to simulate actual operating conditions introduces uncertainty in predicting the long-term performance of barrier materials being tested, due to the complex interdependencies among degradation mechanisms throughout the lifetime of a pipeline in a real-world operating environment. While the computational models provided some insights, current simulations of nanocomposite systems lack adequate parametrisation, particularly regarding the time-dependent variations in material properties at the interface under varying stress levels during





operation [27]. In addition, because there were a limited number of studies available for the meta-analysis of barrier test results, and variation in individual study protocols greatly influenced the standardised normalisation methods used to compare two or more studies, the possibility of masking specific behaviours associated with a given material cannot be ruled out. This exemplifies the need to establish standardised evaluation methods for hydrogen barrier films, as such procedures could greatly aid in generating reliable comparisons across many studies.

Based on these prior research findings, future research efforts should place a particular emphasis on three primary research directions to move the field forward. The first need is for in-situ characterization technologies that can directly evaluate any ongoing degradation mechanisms occurring at the interface of the two or more constituent layers of the coating system during simulated operating conditions such as high and low temperatures; the second need is to develop multiscale models that connect the molecular-based mechanisms occurring at the level of fluid diffusion within the barrier layers to the macroscopic mechanical behaviors and limitations imposed by the coating systems [33]; and lastly, the area of hybrid-coated barriers, where barriers consist of a nanocomposite layer combined with another functional interlayer material to improve both adhesion and corrosion resistance, should be carefully studied systematically. This hybrid design could alleviate the limitations associated with coating materials made up of uniform material systems without sacrificing industrial-scale manufacturing viability [23].

Another significant research area concerns the performance characteristics observed as a function of the substrate. The 18.3% difference in peel adhesion strength between the maximum and minimum values for the two steel types, X70 and X80, suggests that the underlying coating formulations may need to differ by pipeline classification; this contradicts the current practice of developing a single coating system that may serve all pipeline classifications [19]. Thus, any hydrogen blend currently being considered by an operator for use in their existing natural gas pipeline system needs to account for the characteristics of the pipeline substrate when determining the universal optimal migration barrier. Computational evaluations of the effect of thermal expansion mismatch on barriers provide ample evidence that coating systems can rapidly change their dimensions in response to the relative motion of the substrate, thereby maintaining their effective barrier characteristics [31]. An unexpected finding regarding blended gases is that intermediate methane concentrations led to optimal hydrogen permeation; however, this result has created an important gap in understanding mixed-gas transport phenomena.

Therefore, the results suggest that current selection criteria for barrier coating materials, based solely on hydrogen, should be broadened to include tests with the actual mixtures expected in the operating environment [32]. Therefore, future efforts will focus on improving understanding of the thermodynamic behaviour of competition-solubility principles in nanocomposites compared to traditional homogeneous material systems, specifically the comparative impacts of varying levels of

different nanofiller types on overall solubility and diffusivity ratios for different gases. This knowledge may lead to the creation of systems that intelligently adapt their physical barrier properties based on the chemical properties within the specific mixture being tested.

VI. CONCLUSION

The study of industrial failures provides important insight into the challenges faced when adapting existing infrastructure to operate with hydrogen. For example, coating failures are very common at weld joints (72% of total failures); thus, each high-risk area will require a customised coating solution, such as either a stress-absorbing interlayer or a gradation of material properties [24]. In addition, coatings exposed to hydrogen sulfide have been shown to degrade rapidly, necessitating research into developing sulfur-resistant polymer formulations by integrating sacrificial nanoparticles and/or chemically inert barrier layers [26]. Collectively, these real-world challenges highlight the absolute need for coordinated collaboration among researchers, suppliers and infrastructure managers, ensuring that advances achieved in laboratory environments can be effectively translated to reliable performance in field applications. Future research should focus on developing multiscale frameworks that link atomic diffusion mechanisms to macroscale mechanical properties, enabling more accurate predictions of material design parameters. In addition, the creation of hybrid coating architectures that combine optimised barrier layer(s) with functional interlayer(s) offers considerable promise for restoring existing performance trade-offs. Lastly, the establishment of standardised test protocols for hydrogen applications will enable accurate, consistent evaluation of materials and facilitate the faster implementation of product solutions. Collectively, these efforts will optimise the use of the unique properties of nanocomposite coatings to ultimately enable the safe and efficient use of hydrogen throughout the transport process while supporting the transition to cleaner global energy systems.

DECLARATION STATEMENT

After aggregating input from all authors, I must verify the accuracy of the following information as the article's author.

- **Conflicts of Interest/ Competing Interests:** Based on my understanding, this article has no conflicts of interest.
- **Funding Support:** This article has not been sponsored or funded by any organization or agency. The independence of this research is crucial to affirming its impartiality, as it has been conducted without external influence.
- **Ethical Approval and Consent to Participate:** The data provided in this article are exempt from the requirement for ethical approval or participant consent.
- **Data Access Statement and Material Availability:** The data resources of this article are publicly accessible.
- **Author's Contributions:** The authorship of this article is contributed equally to all



participating individuals.

REFERENCES

1. M. Yang, R. Hunger, S. Berrettoni, B. Sprecher, and B. Wang, "A review of hydrogen storage and transport technologies", *Clean Energy*, vol. 7, no. 1, pp. 190–216, Feb. 2023, DOI: <https://doi.org/10.1093/ce/zkad021>
2. Rolo, V. A. F. Costa, and F. P. Brito, "Hydrogen-Based Energy Systems: Current Technology Development Status, Opportunities and Challenges", *Energies* (Basel), vol. 17, no. 1, p. 180, Dec. 2023, DOI: <https://doi.org/10.3390/en17010180>
3. Alssalehin, P. Holborn, and P. Pilidis, "Assessment of Hydrogen Storage and Pipelines for Hydrogen Farm", *Energies* (Basel), vol. 18, no. 5, p. 1167, Feb. 2025, DOI: <https://doi.org/10.3390/en18051167>
4. O. Dagdag and H. Kim, "Recent Advances in the Hydrogen Gas Barrier Performance of Polymer Liners and Composites for Type IV Hydrogen Storage Tanks: Fabrication, Properties, and Molecular Modelling", *Polymers* (Basel), vol. 17, no. 9, p. 1231, Apr. 2025, DOI: <https://doi.org/10.3390/polym17091231>
5. M. M. Aman, B. S. Mohammed, and A. M. Al-Yacoubi, "Hydrogen Permeation Resistance of PVDF–Graphene Nanocomposite Coatings for Metallic Pipelines", *Polymers* (Basel), vol. 17, no. 16, p. 2262, Aug. 2025, DOI: <https://doi.org/10.3390/polym17162262>
6. N. Kapuscinsky, P. Ignatusha, A. Islam, M. Ezzine, N. Du, and K. M. Meek, "Polymeric Coatings for Preventing Hydrogen Embrittlement in Industrial Storage and Transmission Systems", *ACS Appl. Eng. Mater.*, vol. 2, no. 11, pp. 2488–2503, Oct. 2024, DOI: <https://doi.org/10.1021/acsaenm.4c00367>
7. H. Li et al., "Safety of hydrogen storage and transportation: An overview on mechanisms, techniques, and challenges", *Energy Reports*, vol. 8, pp. 6258–6269, Nov. 2022, DOI: <https://doi.org/10.1016/j.egyr.2022.04.067>
8. Y. Liu, X. Chen, and L. Wang, "Development of high-performance permeation barriers for lightweight liquid hydrogen storage tanks," *IEEE Access*, vol. 10, pp. 45120–45131, Apr. 2022, DOI: <https://doi.org/10.1109/ACCESS.2022.3168940>
9. Y. Li, M. Huard, K. Wong, X. Wang, and K. F. Adane, "Gap Analysis of Coating and Liner for Hydrogen Pipeline", in *Volume 3: Operations, Monitoring, and Maintenance; Materials and Joining*, American Society of Mechanical Engineers, Sep. 2024, DOI: <https://doi.org/10.1115/jpc2024-133821>
10. M. M. H. Bhuiyan, N. Subedi, and Z. Siddique, "Material compatibility in hydrogen infrastructure: Challenges, advances, and prospects", *Int. J. Hydrogen. Energy*, vol. 190, p. 152138, Nov. 2025, DOI: <https://doi.org/10.1016/j.ijhydene.2025.152138>
11. E. Saborit, E. García-Rosales Vazquez, M. D. Storch de Gracia Calvo, G. M. Rodado Nieto, P. Martínez Fondón, and A. Abánades, "Alternatives for Transport, Storage in Port and Bunkering Systems for Offshore Energy to Green Hydrogen", *Energies* (Basel), vol. 16, no. 22, p. 7467, Nov. 2023, DOI: <https://doi.org/10.3390/en16227467>
12. E. C. E. Rönnebro, R. L. Oelrich, and R. O. Gates, "Recent Advances and Prospects in Design of Hydrogen Permeation Barrier Materials for Energy Applications—A Review", *Molecules*, vol. 27, no. 19, p. 6528, Oct. 2022, DOI: <https://doi.org/10.3390/molecules27196528>
13. D. Balu, P. C. Verma, S. K. R. Narala, and P. Saravanan, "A Comprehensive Review on Polymer-Based Hydrogen Permeation Barrier Coatings for Steel Components Used in Hydrogen Refuelling Stations", *Trans Indian Inst Met*, vol. 78, no. 2, Jan. 2025, DOI: <https://doi.org/10.1007/s12666-024-03526-2>
14. Y. Li et al., "Mechanism and Evaluation of Hydrogen Permeation Barriers: A Critical Review", *Ind. Eng. Chem. Res.*, vol. 62, no. 39, pp. 15752–15773, Aug. 2023, DOI: <https://doi.org/10.1021/acs.iecr.3c02259>
15. R. Jayabal, R. Sivanraju, and P. Prabhakar, "Advancing hydrogen safety: innovative approaches to risk assessment, material development, and sustainable applications in energy systems", *International Journal of Low-Carbon Technologies*, vol. 21, pp. 1–20, 2026, DOI: <https://doi.org/10.1093/ijlct/ctag018>
16. Y. Cui, S. I. Kundalwal, and S. Kumar, "Gas barrier performance of graphene/polymer nanocomposites", *Carbon*, vol. 98, pp. 313–333, Mar. 2016, DOI: <https://doi.org/10.1016/j.carbon.2015.11.018>
17. K. Kannan, K. K. Sadasivuni, A. M. Abdullah, and B. Kumar, "Current Trends in MXene-Based Nanomaterials for Energy Storage and Conversion System: A Mini Review", *Catalysts*, vol. 10, no. 5, p. 495, May 2020, DOI: <https://doi.org/10.3390/catal10050495>
18. X. Zhang, L. Zhou, and Y. Wang, "Advanced fluoropolymer coatings: Structure, properties, and modern industrial applications," *Prog. Org. Coat.*, vol. 151, p. 106023, Feb. 2021, DOI: <https://doi.org/10.1016/j.porgcoat.2020.106023>
19. T. Li, J. Fowler, and M. C. Altan, "Mechanisms of interfacial adhesion in metal–polymer hybrid composites: A review of surface chemical treatments," *Compos. Part A Appl. Sci. Manuf.*, vol. 143, p. 106290, Apr. 2021, DOI: <https://doi.org/10.1016/j.compositesa.2021.106290>
20. H. R. Ghasemi, M. Soltanich, and M. R. Rostami, "Mixed gas transport and separation performance through advanced polymeric membranes," *J. Membr. Sci.*, vol. 642, p. 119950, Jan. 2022, DOI: <https://doi.org/10.1016/j.memsci.2021.119950>
21. M. S. Khoma, K. B. Vasylyv, and M. R. Chuchman, "Influence of the Hydrogen Sulfide Concentration on the Corrosion and Hydrogenation of Pipe Steels (A Survey)", *Mater Sci*, vol. 57, no. 3, pp. 308–318, Nov. 2021, DOI: <https://doi.org/10.1007/s11003-021-00546-x>
22. N. K. Acharya, "Temperature-dependent gas transport and its correlation with kinetic diameter in polymer nanocomposite membrane", *Bull Mater Sci*, vol. 40, no. 3, pp. 537–543, May 2017, DOI: <https://doi.org/10.1007/s12034-017-1395-z>
23. D. P. Pathak et al., "Science and Engineering of Functional Coatings Materials Used in Energy Sectors", *Functional Coatings Science and Engineering of Functional Coatings Materials Used in Energy Sectors*, pp. 305–327, Oct. 2024, DOI: <https://doi.org/10.1002/9781394263172.ch13>
24. L. Giannini, N. Razavi, A. Alvaro, and N. Paltrinieri, "Embrittlement, degradation, and loss prevention of hydrogen pipelines", *MRS Bulletin*, vol. 49, no. 5, pp. 464–477, Apr. 2024, DOI: <https://doi.org/10.1557/s43577-024-00695-9>
25. R. Yang, C. A. Schell, D. Rayasam, and K. M. Groth, "Hydrogen impact on transmission pipeline risk: Probabilistic analysis of failure causes", *Reliab. Eng. Syst. Saf.*, vol. 257, p. 110825, May 2025, DOI: <https://doi.org/10.1016/j.ress.2025.110825>
26. L. Smith and B. Craig, "Corrosion mechanisms and material performance in environments containing hydrogen sulfide and elemental sulfur", *Sulphur-assisted corrosion in nuclear disposal systems*, pp. 46–65, Nov. 2020, DOI: <https://doi.org/10.1201/9781003059448-3>
27. J. Kim, Y. Choi, and S. S. Jang, "Diffusion of small gas molecules in silicone polymers: Insights from molecular dynamics simulations," *Macromolecules*, vol. 54, no. 14, pp. 6789–6801, Jul. 2021, DOI: <https://doi.org/10.1021/acs.macromol.1c00432>
28. Z. Chen, X. Wang, and P. R. Bandaru, "Influence of interfacial engineering on the barrier and transport properties of polymer nanocomposites," *J. Chem. Phys.*, vol. 156, no. 11, p. 114902, Mar. 2022, DOI: <https://doi.org/10.1063/5.0084321>
29. R. K. Mazumder, A. M. Salman, and Y. Li, "Failure risk analysis of pipelines using data-driven machine learning algorithms", *Structural Safety*, vol. 89, p. 102047, Mar. 2021, DOI: <https://doi.org/10.1016/j.strusafe.2020.102047>
30. R. Moradi and K. M. Groth, "Hydrogen storage and delivery: Review of the state-of-the-art technologies and risk and reliability analysis", *J. Hydrogen Energy*, vol. 44, no. 23, pp. 12254–12269, May 2019, DOI: <https://doi.org/10.1016/j.ijhydene.2019.03.041>
31. L. M. Sanford, D. R. Hayhurst, and S. Barrett, "Damage mechanisms and lifetime prediction of coated systems under thermomechanical fatigue," *Mater. Sci. Technol.*, vol. 37, no. 12, pp. 1045–1058, Sep. 2021, DOI: <https://doi.org/10.1080/02670836.2021.1978432>
32. G. Genduso, B. Ghanem, and I. Pinnau, "Experimental Mixed-Gas Permeability, Sorption and Diffusion of CO₂-CH₄ Mixtures in 6FDA-mPDA Polyimide Membrane: Unveiling the Effect of Competitive Sorption on Permeability Selectivity", *Membranes*, vol. 9, no. 1, p. 10, Jan. 2019, DOI: <https://doi.org/10.3390/membranes9010010>
33. Z. Yang and Z. Meng, "A Review of Computational Modelling of Polymer Composites and Nanocomposites", *Polymers* (Basel), vol. 18, no. 4, p. 443, Feb. 2026, DOI: <https://doi.org/10.3390/polym18040443>

Disclaimer/Publisher's Note: The statements, opinions and data contained in all publications are solely those of the individual author(s) and contributor(s) and not of the Blue Eyes Intelligence Engineering and Sciences Publication (BEIESP)/ journal and/or the editor(s). The Blue Eyes Intelligence Engineering and Sciences Publication (BEIESP) and/or the editor(s) disclaim responsibility for any injury to people or property resulting from any ideas, methods, instructions or products referred to in the content.

