



Development of an Emulsion Paint from the Copolymer Composite of Monomethylol Urea and Expanded Polystyrene Waste

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ABSTRACT

This study aimed to develop an environmentally friendly, waterborne emulsion paint by incorporating recycled polymeric materials, offering a sustainable alternative to traditional petroleum solvent-based paints. The methodology involved synthesizing a copolymer composite of monomethylol urea (MMU) and expanded polystyrene (EPS) waste, followed by physicochemical characterization and performance evaluation of the resulting paint formulations. Key findings indicated that the addition of EPS significantly enhanced binder performance, with the MMU/EPS copolymer exhibiting higher viscosity (100.3 mPa·s vs. 12.2 mPa·s for MMU), increased elongation at break (112% vs. 32%), and reduced formaldehyde emissions (0.0102 ppm vs. 0.033 ppm). Paint formulations containing MMU/EPS demonstrated superior physical properties, including flexibility, adhesion, hardness, blistering resistance, and drying time, meeting SON standards and performing comparably to commercial PVA paints. Chemical resistance tests revealed that MMU/EPS films withstand acidic, alkaline, and saline environments, whereas pure MMU blisters in alkaline solutions. Blending MMU with EPS effectively improves mechanical, chemical, and environmental properties, yielding a durable, elastic, and sustainable emulsion paint. The study recommends MMU/EPS formulations as a viable industrial coating solution and a strategy for valorizing plastic waste.

Keywords: Emulsion paint, copolymer composite, monomethylol urea, expanded polystyrene waste, sustainable materials.

INTRODUCTION

The growing need to balance industrialization and make the environment sustainable has increased the interest in finding alternative materials that would minimize ecological footprints without

compromising functional performance. In the coatings industry, the traditional paint formulations, which incorporate mostly petroleum-based binders and systems that are based on solvents, raise concerns due to their role in the creation of plastic waste, the release of volatile organic compounds (VOC) into the air, and health hazards [1]. These have renewed interest in waterborne finishes and waste-based polymers as potential solutions to petroleum-based binder paints [2].

The decline in the use of solvent-based polymer solutions is a major force in the creation of water-based latex paints. Water-based paints are relatively less damaging to the environment and human health as compared to petroleum solvent-based systems, thereby explaining the increasing trend towards their use. Even though petroleum solvent-based paints tend to be more successful in their performance because of the plasticizing effect of organic solvents that facilitate the formation of a film, water-based systems is an alternative as long as the latex properties are carefully formulated and optimized [3].

The advantage of emulsion paints is due to their low VOC levels, convenience, and application in interior conditions when long-term human manifestations of air pollutants are a frequent occurrence [4]. Latex systems have low viscosity to permit the heat transfer of polymerization, high flowability, and spreading over covered surfaces. Latex particles are coalesced and then form a continuous polymer film after being applied, and then the water rapidly evaporates. In this regard, a suitable formulation is needed to regulate the size of particles, increase the stability of the emulsions, enhance flexibility, and decrease the production costs. Also, flexibility is one of the characteristics of emulsion polymerization processes. The formulation facilitates the manipulation of the property of latex by using a range of polymers, processing procedures, and additives to customize the properties of latex, like particle size, molecular weight, morphology, and chemical functionality. Polymer chains can also be functionalized, and a mixture of dissimilar latexes can be developed with or without copolymerization to satisfy a defined performance. These benefits make emulsion polymerization desirable in terms of the development of advanced and sustainable paint binders.

The properties of emulsion paints are highly dependent on the character of binder systems that determine the formation of the film, its adhesion, stability, and ability to resist the effects of environmental factors [3]. The creation of alternative binders based on renewable and recycled

materials is a significant move towards less reliance on petrochemical resources and improved environmental performance.

Among the most commonly produced polymeric substances, expanded polystyrene boasts of the following characteristics: lightweight, thermal insulation, chemical resistance, flexibility, and dimensional stability [5]. Its low cost has resulted in its wide use in packaging, insulation, and residential practices. Nonetheless, its very slow degradation rate, which can be hundreds of years, and a significant amount of post-consumer waste make EPS an outstanding danger to the environment [6, 7]. EPS is light and bulky and hence consumes a lot of landfill space, thereby adding up to waste-management costs and logistical difficulties. Its production and disposal are also associated with emissions that contribute to the increase of its environmental burden [8].

Recent research has shown that EPS waste can be converted into value-added products through physical, chemical, and composite-based recycling pathways, which can achieve the goals of the circular economy [9, 10]. Polystyrene is especially appealing in applications that involve emulsions because of its high stability and low glass transition temperature, which only allows preparing films and other solid-like binders to be flexible [11]. Polystyrene has demonstrated an increase in mechanical integrity, moisture resistance, and durability when used in composite systems, particularly in building-related materials [12, 13].

Also, urea resins have been known to have high binding, minimal cost, and compatibility with aqueous systems. Organized urea-formaldehyde chemistry provide better reactivity and crosslinking capabilities and allow better control of formaldehyde emissions with improved synthesis conditions [14-16]. Such urea-based systems, together with recycled polymers, may be functional hydrophilic matrices, which promote adhesion and structural cohesion without affecting the environmental safety.

The introduction of expanded polystyrene waste into urea-derived polymer systems is a promising solution in designing sustainable paint binders to solve the waste management problem and the material performance. This idea helps in integrating the most hydrophobic and flexible polystyrene to an already very hydrophilic urea-based network and hence creating a synergistic copolymer composite. Existing research regarding polymer-modified construction materials suggests that the interaction of these synergies can have a profound beneficial effect on mechanical strength, the ability to withstand environmental stressors, and duration [6, 17].

The strategy would fit the circular economy concept since a continuous plastic waste stream is transformed into a useful part of a high-value product. Expanded polystyrene is also an easy raw material to consider since it has a large potential of the resources being recovered [6]. Recycling EPS waste as paint binders will not only remove waste in landfills but also use fewer petrochemical feedstocks, which helps to conserve resources and minimise emissions [9].

EPS-based copolymer binders have feasible benefits for the coatings industry. Waterborne systems developed using such binders should have better adhesion, washability, and longevity and low VOC emission, which are required to be used in residential and commercial use [2, 3]. Moreover, the use of recycled materials is associated with the growing regulatory and consumer pressure in favor of sustainable building material to encourage healthier indoor environments[4]. This study aims to transform plastic waste into useful and high-performance coatings by synthesizing and integrating this new binder into a waterborne paint formulation and testing its performance properties, which supports the idea of innovative polymer chemistry as an alternative in developing sustainable materials and construction.

MATERIALS AND METHODS

The chemicals and reagents used in this study were urea (Sigma-Aldrich, USA), formaldehyde solution (37 wt.% in water, Merck, Germany), sodium dihydrogen phosphate (NaH_2PO_4 , Sigma-Aldrich, USA), sodium hydroxide (NaOH, 0.5M, BDH, UK), sulfuric acid (H_2SO_4 , 0.1M, Merck, Germany), expanded polystyrene (EPS) waste, toluene,

Resin Synthesis

MMU was prepared using the one step process (OSP) as reported by Archibong *et al* [18] with some modifications. One mole of urea (6.0 g) was made to react with one mole of formaldehyde (8.11mL) 37-41% (w/v), using 0.02 g of sodium dihydrogen phosphate as catalyst. The pH of the solution was adjusted to 7.30 by using 0.1M H_2SO_4 and 0.5M NaOH solutions. The solution was heated in a thermostatically controlled water bath at 50 °C. The reaction was allowed to proceed for 60 min after which the resin was removed and kept at room temperature (30 °C).

Determination of Density, Turbidity, Melting Point and Refractive Index

The density of the resins was determined by taking the weight of a known volume of resin inside a density bottle using Pioneer (Model PA64) weighing balance. Three readings were taken for

each sample and average value calculated. The turbidity of the samples was determined by using Supertek digital turbidity meter (Model 033G). To determine the effect of melting point on monomethylol urea, a melting point differential macrophase separation technique was developed. In this technique, MMU was introduced into a porcelain dish. The dish with its content was transferred into an oven set at 120 °C for curing. The mixture was removed periodically from the oven and stirred until the mixture gelled and finally solidified. The temperature was then raised to 150 °C and left for 5 min after which the sample was removed and cooled for observation. The experiment was repeated three times

Determination of Gel Time and Viscosity

Gel Time: This was measured using a gel-time meter. The apparatus temperature was set to 121 °C, and the container was filled with liquid paraffin to the brim. The heating system and stirrer were switched on. Ten grams of the sample were weighed and placed in a test tube, which was then positioned vertically in the hot paraffin bath using a wooden holder. The resin level in the tube was adjusted to remain well within the hot bath. A glass rod was inserted into the test tube and connected to the spindle drive via magnetic coupling. As the spindle rotated, the bottom fin and glass rod rotated together. When the resin began solidifying, resistance to the glass rod rotation stopped the bottom fin, while the upper fin continued rotating until it contacted the stationary fin. The gel time was recorded from the stopwatch at this point [19].

Viscosity: This was determined using a Brookfield viscometer. The appropriate spindle was selected, and the motor speed was set according to standard procedures [19]. The resin temperature was measured with a temperature probe. The spring cap was removed, the spindle fixed, and immersed to the designated mark in the resin. The motor was switched on, and spindle rotation produced shear, providing the viscosity reading. Measurements were carried out at 25 °C.

Determination of Moisture Uptake

The moisture uptake of the resin films was determined gravimetrically, according to method described by Archibong [20]. Known weight of the samples was introduced into desiccators containing a saturated solution of sodium chloride. The increase in weight (wet weight) of the sample was monitored until a constant weight was obtained. The difference between the wet

weight and dry weight of the sample was recorded as the moisture uptake by the resin. Triplicate determinations was made for each sample and the average value recorded.

Determination of Elongation at Break

The elongation at break was determined using Inston Tensile Testing Machine (Model 1026). Resin films of dimension 50 mm long, 10 mm wide and 0.15 mm thick was brought to rupture at a clamp rate of 20 mm/min and a full load of 20 kg. Three runs were carried for each sample and the average elongation evaluated and expressed as the percentage increase in length.

Determination of Formaldehyde Emission Using UV-Spectrophotometer

To determine any possible absorbance by formaldehyde, deionized water was used as the blank. The cuvette was rinsed several times with tap water followed by deionized water, it was then filled with deionized water, and placed in the holder, and the spectrophotometer was blanked at 563 nm. The sample was then put into another cuvette and the absorbance was noted at the same wavelength of 563 nm, with concentration recorded

Determination of Water Solubility

The solubility of MMU was determined by mixing 1mL of the resin with 5 mL of distilled water at room temperature (30 °C). The degree of solubility was evaluated visually and the result recorded.

Paint Formulation

The method described by Karakas et al [21] was adopted with some modifications. The method splits the production process into three main stages as illustrated in Table 1. The mixture in each stage was stirred for 15 min using a mechanical stirrer.

First Stage

At this stage, additives such as, dispersants, defoamer, thickener, anti-skin, drier, wetting agents, stabilizer, pH adjuster and preservatives were added. The basic purpose of this stage is to provide a favourable environment for wetting and dispersion of particles. A volume of 185 mL of distilled water was introduced into a mixing tank and the overhead stirrer switched on after the addition of 12.7 g of the additives as shown in Table 1. This mixture was stirred using a high speed stirrer for 15 min.

Second Stage

In the second stage, also known as “Millbase”, pigments and extenders were dispersed in the mills. Immediately after dispersion, the stirrer speed in the millbase stage was increased to a very high speed and the mixture was stirred for another 15 min. In millbase, binder was not added to avoid its structural deformation under the high mechanical forces.

Third Stage

Finally, binder plus the rest of the additives used in the first stage were mixed with 15 ml of water. This stage is called “Letdown”. At this stage, the mixture was stirred at moderate speed for another 15 min. Energy losses in the mill base are minimized by adding thickeners before the dispersion stage of the production process. The above processes were repeated using pure MMU as binder.

Table 1: Recipe for the formulation of an emulsion paint

Stage	Material	Quantity (g)
First	Water	100.00
	Anti-form	0.20
	Drier	0.02
	Calgon	1.16
	Genepour	1.16
	Bermocoll	2.50
	Troystan	1.14
	Dispersant	0.20
	Butanol	5.00
	Ammonia	0.54
Second [Millbase]	TiO ₂	50.00
	Al ₂ SiO ₃	11.20
	NaCO ₃	30.58
	Kaolin	2.52
	CaCO ₃	73.00
Third [Letdown]	Binder	100.00
	Water	50.00

Dispersant	0.20
Nicofoam	0.20
Anti-Skinning agent	0.20

Tests for the Paint Samples

Paint samples were analyzed as described in Niger Cedar laboratory hand book of paint formulation recipe and standard organization of Nigeria methods [22].

Viscosity

Viscosity of paint sample was determined by using an I.C.I. Rotothinner viscometer. The 500 mL tin sample container of the instrument was filled with paint sample under test to 0.3 mm of the top rim of the depth gauge. The sample was stirred with a thermometer and the temperature maintained at 27 °C. The container with the paint sample was placed in the processing ring on the turntable and the lever pulled down to switch on the motor automatically. The disc was then allowed to run until a steady state was reached (5 minutes). At the end of the required time, the viscosity was recorded in poises. Triplicate determinations were made for each sample and mean value recorded.

pH Measurement and Opacity Test

The pH of the paint sample was determined using a standard laboratory pH meter. The paint sample was scooped with a palette knife and evenly spread across the width of a Mohest chart (black and white striped paper) approximately 4 cm from the edge. The paint was then applied down the length of the paper using a K-bar (stainless steel applicator) and allowed to dry. A second coat was applied in the same manner, leaving a 5 cm gap from the edge of the first coat. Opacity was assessed based on the extent to which the black and white stripes were covered by the paint. Three replicates were performed for each sample, and the mean value was recorded for quality assessment.

Drying Time and Flexibility

Dry time was evaluated by applying the paint sample on a glass panel with the aid of bar applicator, and allowed to dry. Dry to touch was taken when the paint film was no longer sticking to the finger and dry to hard was taken when the film resisted finger print. Triplicate evaluations were made for each sample and mean value assessment recorded. For flexibility test, paint sample was applied on a freshly degreased and chromate aluminium with the aid of paint applicator. The

film was allowed to air dry under room temperature (27-30 °C) for 7 days. The painted panel was carefully bent through 180° in a smooth motion, taking approximately 1–2 seconds. After bending, the panel was removed and visually examined for any cracking or loss of adhesion. The presence of cracks or detachment indicated inflexibility or brittleness of the paint film. Each sample was tested in triplicate at a temperature of 27–30 °C to ensure reliability and consistency of the results.

Chemical Resistance

The chemical resistance of the paint films was carried out thus; three flexible aluminium panels (150 mm x 0.3 mm) were used as the test panels. A coat of paint with paint applicator was applied on the panels. One litter glass beaker was filled with 0.1 M NaOH solution to a depth of 150 mm and the test pieces was immersed for 48 hours to the depth of approximately 120 mm. The test piece was removed, washed with running water and stand to dry for 2 hours. The above procedure was repeated using 0.1M HCl and 0.1M NaCl respectively. Poor chemical resistance indicated by the presence of any surface defects such as cracking, blistering, peeling or changes in color.

Tackiness

This was carried out qualitatively on the dried film by hand feeling to find out if the paint film is sticky or not. Stickiness of a dried paint film is an indication that the film is tacky. Triplicate samples were used for each determination and the average quality assessment recorded.

Adhesion Property

This was carried out by applying a coat of paint film with film applicator on a degreased metal panel and allowed to dry for 48 hours. Two sets of lines, one crossing perpendicularly over the other were drawn with a crosshatch tester on the paint film. An adhesive tape was pressed firmly with the thumb covering all the intersections of the perpendicular line. The adhesive tape was held at its loose end and forcibly removed from the panel. Removal of more than 50% of square lines of the paint film indicates a poor adhesion. Triplicate determination were made

Resistance to Blistering

To determine Resistance to blistering, undiluted paint sample was applied to a glass panel with an applicator to give a wet film thickness of about 120 µm, which was allowed to dry for 24 hours. At the end of this period 4 ml of distilled water in the form of circular drop was placed on the film. The presence of blistering, wrinkling, swelling or cracking within a period of 30 minutes indicates

poor water resistance. Triplicate determinations of each sample were made and quality assessment recorded as mean.

Stability Test

The paint sample was fully sealed in a container and allowed to stay at room temperature (27 °C) for 12 months. At the end of this incubation period, the sample was re-examined for any change in viscosity or coagulation of the emulsion paint. Absence of coagulation or any change in viscosity is regarded as pass. Triplicate samples were used for each determination and average recorded

RESULTS AND DISCUSSION

Table 2 summarizes the physicochemical properties of MMU and MMU/EPS, showing that the EPS-modified copolymer exhibits improved performance while meeting or exceeding standard industry requirements.

Table 2: Physicochemical properties of MMU, MMU/EPS and the comparison with accepted levels in the coating industry

Properties	MMU	MMU/EPS	Accepted Level in the coating industry
Refractive Index	1.4187	1.4309	1.4000(minimum)
Viscosity(mpa.s)	12.20	100.30	3.11-38.00
Elongation at break (%)	32	112	124
Formaldehyde Emission (ppm)	0.0330	0.0102	0.08 (maximum)
Moisture Uptake (%)	2.5	0.82	3.00 (maximum)
Density (g/cm ³)	1.183	1.16	1.07 (Minimum)
Melting Point (°C)	226.20	215.24	200 (Minimum)
Solubility	Soluble	Slightly Soluble	-
Turbidity (NTU)	326	684	-
Gel Time(h)	264	76	-
Appearance	Clear	Clear	-

Physicochemical data in Table 2 indicate that the addition of expanded polystyrene into monomethylol urea resin has a considerable positive effect on the performance of the binder without compromising the requirements of the coating industry. The refractive index of the MMU/EPS copolymer is a little bit greater than that of pure MMU, which shows that the structure of the polymer matrix has been altered without affecting the optical acceptability of the polymer in the coating use.

The viscosity of the MMU/EPS binder enhances significantly, which is indicative of elevated molecular weight and enhanced interaction between the molecules due to the formation of copolymer. Such an upsurge will be useful in the film formation and application control since adequate viscosity will help form a uniform thickness of the coating and minimize surface flaws.

Likewise, the elongation at break of MMU/EPS is significantly greater than that of pure MMU, indicating increased flexibility and toughness. It is owed to the enhanced ductility provided by the flexible EPS segments in the polymer that enable the material to sustain higher deformation rates before breaking.

The MMU/EPS binder records a considerable decrease in the emission of formaldehyde. The reason behind this decrease is the reduced availability of reactive formaldehyde positions by the interaction between MMU and EPS that successfully scavenges the free formaldehyde in the polymer structure [23]. This is valuable, especially considering that formaldehyde is a carcinogen and that the use of this chemical is being regulated more and more [20, 21]. The decreased emissions also help to increase the quality of indoor air and safety in the workplace [22, 23].

The absorption of moisture is less in MMU/EPS than pure MMU due to the high level of hydrophobicity induced by EPS and the ability to resist water intrusion. The diminishing density and melting point are also signs of larger free space and free movement of the molecules in the copolymer matrix that allow easier processing. Moreover, the higher the turbidity and the lower the gel time, the more interchain interactions and faster curing behavior.

In all, MMU/EPS copolymer has better mechanical, environmental, and processing properties, which prove its applicability as a sustainable binder in emulsion paint formulations.

The physical performance of paints formulated using MMU, MMU/EPS, and PVA binders clearly demonstrates the critical role of binder chemistry in determining coating behavior, durability, and service performance. As presented in Table 3, the pH values of all formulations fall within or very close to the acceptable range specified by the Standard Organization of Nigeria (SON), indicating good chemical stability and suitability for practical application.

Table 3: Physical Parameters of Paint Synthesis from MMU, MMU/EPS, and PVA Binder

Parameters	MMU	MMU/EPS	PVA	SON Standard
Ph	8.30	8.62	8.82	7-8.5
Flexibility	Fail	Pass	Pass	Pass
Viscosity (poise)	8.8	15.8	15.1	6-15
Opacity	Pass	Pass	Pass	Pass
Adhesion	Fail	Pass	Pass	Pass
Hardness test	Fail	Pass	Pass	Pass
Tackiness	Pass	Pass	Pass	Pass
Blistering	Fail	Pass	Pass	Pass
Drying Time				
Touch	50	35	67	20
Hard	73	61	77	120
Storage stability (6 months)	Pass	Pass	Pass	Pass

The slightly higher pH recorded for the MMU/EPS paint compared to the pure MMU formulation reflects the reduced proportion of MMU within the copolymer composite and the moderating influence of EPS. Such mildly alkaline conditions are advantageous, as they limit microbial proliferation and contribute to improved storage stability, which is corroborated by the six-month stability results. All the paint samples satisfied the opacity requirement, confirming effective pigment dispersion and adequate hiding power. However, notable differences emerged in mechanical performance. The failure of the pure MMU paint in flexibility, adhesion, hardness, and blistering tests highlights inherent deficiencies associated with its hydrophilic character and lower cohesive strength. These limitations are consistent with polymer systems characterized by lower molecular weight and reduced crosslink density, which restrict their ability to withstand mechanical stress and moisture exposure. In contrast, the MMU/EPS paint exhibited marked

improvements in flexibility and adhesion, achieving performance comparable to the commercial PVA paint. This enhancement can be attributed to the introduction of EPS segments, which impart elastic behavior and improve stress distribution within the polymer network. The superior mechanical performance of the PVA paint is similarly linked to its higher molecular weight and more extensive crosslinked structure, which promote robust film formation and durability [3].

The resistance to blistering observed for the MMU/EPS paint further indicates improved water resistance and reduced permeability. The hydrophobic nature of EPS, combined with the formation of a more compact and interconnected polymer network, limits moisture ingress and solvent diffusion through the coating film. Conversely, the failure of the pure MMU paint in this test underscores its vulnerability to moisture-induced degradation.

Drying time results show that the MMU/EPS paint achieved both touch-dry and hard-dry states more rapidly than the MMU formulation. This behavior reflects enhanced film formation efficiency and higher crosslink density, which facilitate faster water evaporation and particle coalescence. The increased viscosity observed for the MMU/EPS paint, relative to MMU, further supports this interpretation, as it reflects increased molecular weight and structural complexity of the copolymer binder. While elevated viscosity improves film build and reduces sagging, it remains within a workable range suitable for application.

Although fillers were not explicitly incorporated in these formulations, their inclusion has been shown to further influence coating performance by modifying film formation, mechanical reinforcement, and barrier properties [24]. Bio-based fillers such as lignins and microfibrillated cellulose can enhance adhesion strength and durability through improved mechanical interlocking within the coating matrix [24]. The integration of such lignocellulosic materials aligns with sustainable material development goals and has been demonstrated to improve strength, stiffness, and resistance to environmental degradation in composite coatings [25, 26].

The MMU/EPS binder significantly enhances the physical performance of the paint, effectively overcoming the limitations of pure MMU and producing properties comparable to those of conventional PVA-based paints. These findings confirm that copolymerization with EPS is a viable strategy for developing robust, flexible, and water-resistant coatings while supporting sustainable materials innovation.

Table 4 shows the different effects of binder composition on the chemical stability of paint films in acidic, alkaline, and saline media.

Table 4: Comparison of Chemical Resistance of MMU, MMU/EPS and PVA Paint Film

Sample	Media		
	0.1M HCl	0.1M NaOH	0.1M NaCl
MMU	No effect	Blistering	No effect
MMU/EPS	No effect	No effect	No effect
PVA	No effect	No effect	No effect

Chemical resistance is a primary performance criterion of coating materials since it dictates their capability to withstand hostile environments without integrity or protective performance failures. The sodium chloride medium was found to be stable in all paint films (MMU, MMU/EPS, and PVA), meaning that they had sufficient neutral salt resistance. This experiment indicates that under these conditions, all the coating matrices are quite resistant to ionic penetration, and the film continuity and surface cohesion are high enough to withstand degradation by salt [27]

Significant changes were observed on alkaline exposure. In sodium hydroxide solution, the pure MMU paint film exhibited blistering, which means that it is susceptible to alkaline attack, a typical failure in alkyd and other resin systems that have hydrolyzable ester groups [27] According to this reaction, it may be explained by the hydrophilic character of the MMU matrix that facilitates the intake of the solvent and interaction between the matrix and the alkaline species. These interactions destroy intermolecular bonding in the film, which causes localized swelling and the creation of surface defects [27]. The fact that no such defects can be found in acidic conditions indicates that the MMU matrix is relatively more stable in low pH conditions, where hydrolytic interactions are less hostile to the polymer backbone. Moreover, there are ester groups in some resin formulations, including the ones in alkyd and poly(ester amide) resins, which are more susceptible to the alkaline hydrolysis, and the free Na^+ and OH^- ions of NaOH will begin to break down the ester groups, which will create sodium carboxylates and cause the breakage of the film [27].

The MMU/EPS paint film, on the contrary, was found to be entirely resistant to all test media, and the surface did not show signs of degradation in acidic, alkaline, or salty conditions. This increase in chemical stability is indicative of structural advantages brought about by the introduction of EPS, in which the introduction of hydrophobic polystyrene segments into the

copolymer structure decreases accessibility of solvents to the film and chemical diffusion into the film. Compact size and the formation of interrelations between polymer structure also inhibit the movement of the molecules, thus preventing swelling and hindering chemical diffusion in adverse environments. Likewise, the PVA paint demonstrated good chemical resistance in all the environments tested, also in agreement with a binder system that has a strong intermolecular bond and homogeneous film-forming properties, which can resist chemical intrusion

These findings prove that chemical resistance (alkaline attack, which is frequently observed in service conditions) is much enhanced by copolymerizing MMU with EPS. The MMU/EPS paint film is seen to have better protective performance than pure MMU, thereby making it a longer-lasting and chemically stable coating that can be used in demanding environments.

The correlation between gel time and viscosity of the MMU resin (Figure 2) follows the known curing patterns, according to which longer periods of reaction provide more opportunities to extend the polymer chain and crosslink [28].

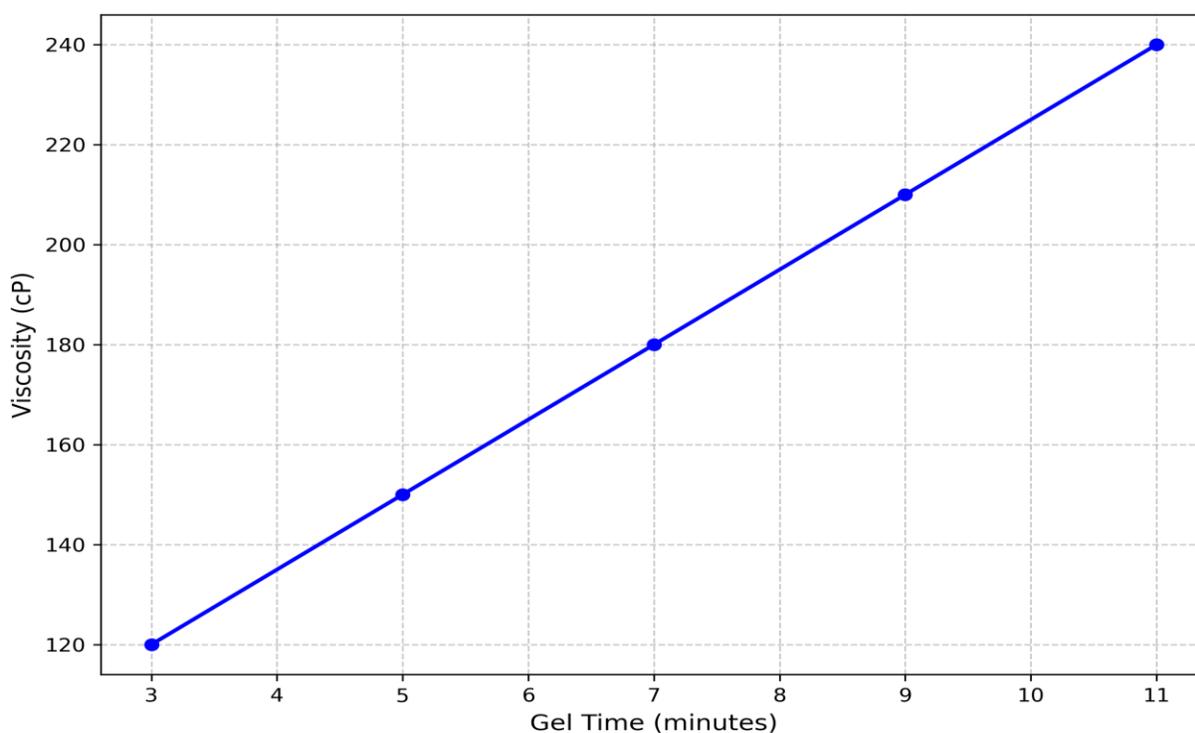


Figure 2. Gel Time vs. Viscosity of Monomethylol Urea (MMU) Resin.

According to the rheological studies, pot life and gel time are paramount factors that define processability because gel time is the beginning of the network building due to polyfunctional monomers cross-linking [28]. The more the degree of crosslinking is achieved, the greater the network density will affect the glass-transition temperature and mechanical characteristics of the cured material [28, 29]. Reactive diluents like dimethyl itaconate and methyl methacrylate have been proven to be involved in regulating the viscosity of the resin; the optimum ratio of the mixture is key to the high gel content and the good thermomechanical behavior [30]. The trend in the viscosity value with an increasing gel time in this study has shown that the network is formed properly, thus proving that the MMU resin observed is effective as a binder in the application of emulsion paint.

CONCLUSIONS

The current research proves the fact that MMU/EPS-based emulsion paint is a sustainable emulsion coating. A significant improvement in the physicochemical properties, such as viscosity, flexibility, elongation at break, and moisture resistance, and a significant decrease in formaldehyde emissions are achieved by the incorporation of EPS into the MMU binder. The resulting paint exhibited excellent mechanical performance, flexibility, adhesion, hardness, and resistance to blistering, with an equal performance to commercial PVA paints. The chemical resistance tests ensured that the entire MMU/EPS film was completely stable in acidic, alkaline, and saline solutions and hence its applicability in harsh service conditions. The MMU copolymerized with EPS does not only enhance the performance of the paint but also contributes to environmental sustainability through the use of post-consumer plastic waste as an alternative to the traditional solvent-based coated paints, which have a negative impact on the environment, and offers a viable, eco-friendly substitute to the conventional solvent-based coated paints in the paint business.

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REFERENCES

1. Demchuk, Z., Mora, A., Choudhary, S., Caillol, S. & Voronov, A. (2021). Biobased latexes from natural oil derivatives. *Industrial Crops and Products*, 162, 113237. <https://doi.org/10.1016/j.indcrop.2021.113237>
2. Pieters, K. & Mekonnen, T. H. (2024). Progress in waterborne polymer dispersions for coating applications: Commercialized systems and new trends. *RSC Sustainability*. <https://doi.org/10.1039/d4su00267a>
3. Ibrahim, B., Helwani, Z., Wiranata, A., Fadhilah, I., Miharyono, J. & Nasruddin, N. (2022). Properties of emulsion paints with binders based on natural latex grafting styrene and methyl methacrylate. *Applied Sciences*, 12(24), 12802. <https://doi.org/10.3390/app122412802>
4. Mermer, N. K., Ugur, N., Kuzgun, F., Bakar, B., İnceoğlu, F. & Pınar, E. Ü. (2023). Evolution of coalescent agent-free ultra-low VOC paint with formaldehyde capturing properties. *Atmospheric Pollution Research*, 14(8), 101812. <https://doi.org/10.1016/j.apr.2023.101812>
5. Yang, T. C. & Xing, Y. (2025). Polystyrene upcycling via photocatalytic and non-photocatalytic degradation. *Molecules*, 30(15), 3165. <https://doi.org/10.3390/molecules30153165>
6. Gautam, B., Tsai, T.-H. & Chen, J. (2024). Towards sustainable solutions: A review of polystyrene upcycling and degradation techniques. *Polymer Degradation and Stability*, 225, 110779. <https://doi.org/10.1016/j.polyimdegradstab.2024.110779>
7. Yan, J., Wu, H., Huang, P., Wang, Y., Shu, B., Li, X., Ding, D., Ya, S., Wang, C., Wu, J. & Sun, L. (2022). Investigation on the controllable synthesis of colorized and magnetic polystyrene beads with millimeter size via in situ suspension polymerization. *Frontiers in Chemistry*, 10. <https://doi.org/10.3389/fchem.2022.891582>
8. Hashish, S. A., Kishar, E., Ahmed, D., Ragei, S. M. & Ebrahim, A. A. M. (2025). Sustainable slag-based geopolymers incorporating styrofoam and Moringa leaves ash for mechanical enhancement and crystal violet adsorption. *Scientific Reports*, 15(1). <https://doi.org/10.1038/s41598-025-20119-1>
9. Capricho, J. C., Prasad, K., Hameed, N., Nikzad, M. & Salim, N. V. (2022). Upcycling polystyrene. *Polymers*, 14(22), 5010. <https://doi.org/10.3390/polym14225010>

10. Pokorný, J., Zárbybnická, L., Ševčík, R. & Podolka, L. (2024). Utilization of waste-expanded thermoplastic as a sustainable filler for cement-based composites for greener construction. *Buildings*, 14(4), 990. <https://doi.org/10.3390/buildings14040990>
11. Save, M., Hellaye, M. L., Villedon, V. de, Adoumaz, I., Pillet, M., Atanase, L. I., Lahcini, M., Deniau, E., Khoukh, A., Pellerin, V., Ly, I., Dulong, V. & Schmitt, V. (2022). Biosourced polymeric emulsifiers for miniemulsion copolymerization of myrcene and styrene: Toward biobased waterborne latex as Pickering emulsion stabilizer. *Biomacromolecules*, 23(6), 2536. <https://doi.org/10.1021/acs.biomac.2c00257>
12. Ahmed, S. A., Ebrahim, E. & El-Feky, M. S. (2024). Achieving sustainable performance: Synergistic effects of nano-silica and recycled expanded polystyrene in lightweight structural concrete. *Scientific Reports*, 14(1), 26648. <https://doi.org/10.1038/s41598-024-77029-x>
13. Ferrández, D., Zaragoza-Benzal, A., Barrak, É., Santos, P. & Rodrigues, C. (2024). Upcycling expanded polyethylene waste for novel composite materials: Physico-mechanical, hygrothermal and life cycle assessment. *Results in Engineering*, 24, 103535. <https://doi.org/10.1016/j.rineng.2024.103535>
14. Al-Mokhalelati, K., Karabet, F., Allaf, A. W., Naddaf, M. & Lafi, A. G. A. (2023). Spectroscopic investigations to reveal synergy between polystyrene waste and paraffin wax in super-hydrophobic sand. *Scientific Reports*, 13(1). <https://doi.org/10.1038/s41598-023-36987-4>
15. Horma, O., Charai, M., Hassani, S. E., Hammouti, A. E., Moussaoui, M. A. & Mezrhab, A. (2022). Thermal performance study of a cement-based mortar incorporating EPS beads. *Frontiers in Built Environment*, 8. <https://doi.org/10.3389/fbuil.2022.882942>
16. Yang, H., Wang, H., Du, G., Ni, K., Wu, Y., Su, H., Gao, W., Tan, X., Yang, Z., Yang, L. & Ran, X. (2023). Ureido hyperbranched polymer modified urea-formaldehyde resin as high-performance particleboard adhesive. *Materials*, 16(11), 4021. <https://doi.org/10.3390/ma16114021>
17. Alhazmi, H., Shah, S. A. R., Anwar, M. K., Raza, A., Ullah, M. K. & Iqbal, F. (2021). Utilization of polymer concrete composites for a circular economy: A comparative review for assessment of recycling and waste utilization. *Polymers*, 13(13), 2135. <https://doi.org/10.3390/polym13132135>
18. Archibong, C. S., Fadawa, F.W and Bintu Kime (2025). Optimization of the processing parameters of HPKO/MMU composite, as copolymer binder for a water resistant paint. *Nigerian Journal Research Journal of Chemical Sciences* 13(1),237-257

19. Ashori, A. & Kuzmin, A. M. (2024). Effect of chitosan-epoxy ratio in bio-based adhesive on physical and mechanical properties of medium density fiberboards from mixed hardwood fibers. *Scientific Reports*, 14(1). <https://doi.org/10.1038/s41598-024-55796-x>
20. Archibong Christopher Sunday (2023). Comparison of some physiochemical parameters of emulsion paint formulated from Monomethylol urea resin, and that from di, and Trimethylol urea resins. *Global Scientific Journal*, 11(6): 3102-3113
21. Karakas, F., Pyrgiotakis, G., Celik, M. S. & Moudgil, B. M. (2011). Na-bentonite and MgO mixture as thickening agent for water-based paints. *KONA Powder and Particle Journal*, 29, 96–106. <https://doi.org/10.14356/kona.2011011>
22. Ganeshram, V. & Achudhan, M. (2013). Synthesis and characterization of phenol formaldehyde resin as a binder used for coated abrasives. *Indian Journal of Science and Technology*, 6(6), 4814-4823.
23. Porfyrus, A. D., Vafeiadis, A., Gkountela, C. I., Politidis, C., Messaritakis, G., Orfanoudakis, E., ... & Vouyiouka, S. N. (2024). Flame-Retarded and Heat-Resistant PP Compounds for Halogen-Free Low-Smoke Cable Protection Pipes (HFLS Conduits). *Polymers*, 16(9), 1298.
24. Calvez, I., Garcia, R. Á., Koubaa, A., Landry, V. & Cloutier, A. (2024). Recent advances in bio-based adhesives and formaldehyde-free technologies for wood-based panel manufacturing. *Current Forestry Reports*, 10(5), 386. <https://doi.org/10.1007/s40725-024-00227-3>
25. Benhamou, A. A., Abid, L., Calvez, I., Cloutier, A., Nejad, M., Stevanović, T. & Landry, V. (2025). Advances in lignin chemistry, bonding performance, and formaldehyde emission reduction in lignin-based urea-formaldehyde adhesives: A review. *ChemSusChem*. <https://doi.org/10.1002/cssc.202500491>
26. Buyondo, A. K., Kasedde, H., Kirabira, J. B. & Yusuf, A. A. (2025). Integration of fillers in paint formulation: Comprehensive insights into methods, properties, and performance. *Results in Engineering*, 26, 105543. <https://doi.org/10.1016/j.rineng.2025.105543>
27. Abdelkawy, M. A., Shoker, A., Badr, B. I. & Mahmoud, Y. A. G. (2026). Eco-sustainable chitosan–PVA biocomposites reinforced with recycled alum: preparation, characterization, and antimicrobial assessment. *RSC advances*, 16(6), 5471-5489.

28. Seychal, G., Ocando, C., Bonnaud, L., Winter, J. D., Grignard, B., Detrembleur, C., Sardón, H., Aramburu, N. & Raquez, J. (2023). Emerging Polyhydroxyurethanes as Sustainable Thermosets: A Structure–Property Relationship. *ACS Applied Polymer Materials*, 5(7), 5567. <https://doi.org/10.1021/acsapm.3c00879>
29. Choi, J. W., Jang, S. H. & Jang, K. (2025). Interactions and Curing Dynamics Between UV-Triggered Epoxy Acrylate Binder, Curing Agents and Photoinitiators. *Polymers*, 17(9), 1252. <https://doi.org/10.3390/polym17091252>
30. Spasojević, P., Šešlija, S. I., Marković, M. D., Pantić, O., Antić, K. M. & Spasojević, M. (2021). Optimization of Reactive Diluent for Bio-Based Unsaturated Polyester Resin: A Rheological and Thermomechanical Study. *Polymers*, 13(16), 2667. <https://doi.org/10.3390/polym13162667>
31. Gabrielli, S., Pastore, G., Stella, F., Marcantoni, E., Sarasini, F., Tirillò, J. & Santulli, C. (2021). Chemical and mechanical characterization of licorice root and palm leaf waste incorporated into poly(urethane-acrylate) (PUA). *Molecules*, 26(24), 7682. <https://doi.org/10.3390/molecules26247682>
32. Rahmani, A., Abdulkhali, A., Ashori, A. & Hosseinzadeh, J. (2024). Development of high-performance biocomposites through lignin modification and fiber reinforcement. *Scientific Reports*, 14(1). <https://doi.org/10.1038/s41598-024-80256-x>
33. DeRosa, M. E., Baker, L. S., Melock, T. L. & Yang, B. (2021). Ultraviolet cure kinetics of a low Tg polyurethane acrylate network under varying light intensity and exposure time. *Progress in Organic Coatings*, 158, 106353. <https://doi.org/10.1016/j.porgcoat.2021.106353>
34. Ganeshram, V. & Achudhan, M. (2013). Synthesis and characterization of phenol formaldehyde resin as a binder used for coated abrasives. *Indian Journal of Science and Technology*, 6(6), 4814-4823.