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Article

Development of Moisture/Oil-Resistant Biocoatings from Waste Cooking Oil for Packaging Applications: Scientific Upcycling with Circular Economy Potential

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ABSTRACT: The excessive generation and mishandling of waste cooking oil (WCO) necessitate the need for its conversion into specialty resins and polymers for various applications. This work focuses on the development of a biocoating from WCO for paper-based packaging with improved hydrophobicity and moisture barrier properties. WCO was epoxidized and acrylated to form AWCO, subsequently characterized, and modified into a water-based emulsion using a poly(vinyl alcohol) solution. (Aminopropyl)triethoxysilane was grafted in varying proportions to AWCO emulsion to enable cross-linking onto hydroxyl groups of cellulosic paper. The curing process of the coated paper (~40 gsm coating) was carried out at 130 °C for 5−7 min to ensure complete cross-linking. The coated papers exhibited a water contact angle of greater than 90°, a kit rating of 12/12 along with excellent water retention and oil and ice repellency. Water vapor transmission rate reduction up to ~129 g/m^2·day was achieved by depicting the moisture barrier as well. High glass transition temperatures (T_g) and thermal stabilities of the coatings revealed their suitability for hot food packaging. The coatings most importantly depicted high repulping potential and home compostability, making it a commercially feasible replacement for the existing plastic liners in paper packaging.

KEYWORDS: *waste cooking oil (WCO), bioresin, paper coating, hydrophobicity, oleophobicity, repulpability*

INTRODUCTION

The world's consumption of edible oils has increased tremendously in the past years and has resulted in a concomitant increase in the generation of waste cooking oil (WCO), possessing high levels of free radicals and total polar compounds (TPCs), which upon ingestion into the human body causes serious illness. Currently, the net global production of WCO is about 29 million tons/annum, and the WCO market is expected to reach \$8886.7 million by 2026, with a CAGR of 5%.[1] At present, only 3−5 % of it gets converted into value-added products like green solvent, surfactant, biodiesel, etc. (biodiesel as the major conversion), with the rest getting discarded off.[2−5] Unlawful WCO handling and dumping would cause it to either end up in sewage systems, creating havoc in water treatment and thus resulting in water pollution, or get embarked back into the food chain.[2,6] The packaging industry is an essential sector in which one of the most widely used substrates in food and general packaging is paper as a replacement for plastic, due to its light weight and compostable nature. However, paper possesses high hydrophilicity and a low moisture barrier, which necessitates the need for lamination using water-resistant plastics or bioplastics, e.g., low-density polyethylene, poly(lactic acid), etc., which either are nondegradable, have high cost, have lesser properties, or require specific industrial composting conditions to degrade, which are rarely performed in most of the countries.[7−9] Although the addition of nanoparticles of silver and zinc oxide to impart antibacterial and antioxidant properties to these plastics has been reported, they are prone to migration, making them unsuitable for food packaging.[10,11] Also, removal of the plastic liner from paper for recycling is too difficult.[12−15] Hence, the synthesis of home compostable coatings derived from biosources is a niche area for consideration.

Vegetable oils with the presence of multiple reactive double bonds, hydroxyl groups, etc., is a suitable choice for the synthesis of polymers with unique properties.[16−19] Recently, Khan et al. made AESO into a water-based emulsion using poly(vinyl alcohol) (PVA), coated on kraft paper and subsequently cured with a photoinitiator (2-hydroxy-2-methylpropiophenone) under UV light, resulting in a kit rating of 7/12.[20] However, the use of a photoinitiator is not

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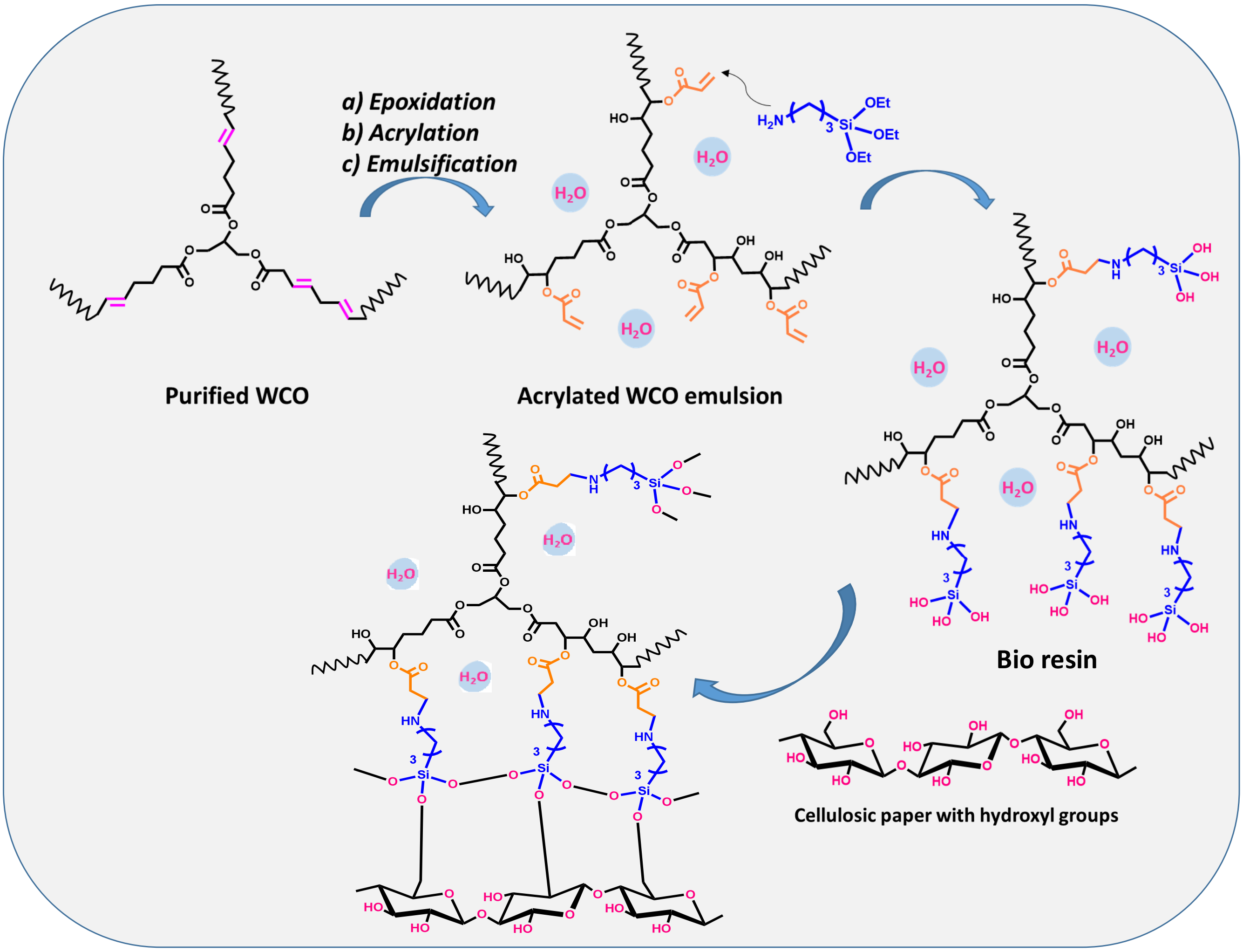

Figure 1. Reaction scheme for the conversion of WCO into the bioresin and its grafting onto the paper surface.

acceptable because it is reported to leach out even from the cured polymer, which raises a concern in its usage for food packaging application.[21,20,22,23] Silanization of epoxidized corn oil and epoxidized castor oil has been carried out using (aminopropyl)triethoxysilane (APTES), which was then coated on filter paper and cured at 100−120 °C for 3 h to obtain good barrier properties with 60−70% reduction in the water vapor transmission rate (WVTR).[13,24] However, most of the reported works in such biobased coatings carried out using the thermal curing technique require a very high curing time (>3 h), which limits its commercialization at the industry level.

The objective of this study was to develop a water-based emulsion coating using WCO, a feat that has not been explored in prior articles. The coatings were synthesized without the use of a photoinitiator and in a very short curing time, overcoming the drawbacks of reported works. Utilizing WCO instead of pure vegetable oils (which are from first-generation biomasses) represents a better approach in the conversion of waste into value-added polymeric products while certifying a circular economy. The coating was aimed at possessing the properties required for food packaging, such as water and oil resistance, along with meeting circular economy criteria, such as repulpability and compostability. In the current work, waste sunflower oil was acrylated and made into a water-based emulsion using PVA, a degradable polymer with good emulsifying properties.[35] Silane groups were grafted onto the acrylate moieties through an aza-Michael reaction using APTES in varying proportions and then coated on kraft paper through condensation. The coatings were characterized for thermal stability, water, oil, and ice resistance, transparency, heat-sealing ability, repulpability, and compostability. Thus, the utilization of WCO for replacing plastic lamination in paper packaging is explored along with the recyclability potential, in accordance with the United Nations Sustainable Development Goals.

■ RESULTS AND DISCUSSION

The detailed methods for purification, epoxidation, acrylation, and emulsification of WCO, along with the grafting of APTES onto AWCO, are explained in the Materials and Methods section of the Supporting information (SI). The reaction scheme for the processes involved is depicted in Figure 1, and a detailed explanation is provided in the SI. The sample codes for the optimized formulations are listed in Table 1

Free Fatty Acid (FFA) Reduction. In this study, the WCO collected was physically purified using filtration to remove solid impurities and then reacted with zeolite to remove FFAs present in it. The generation of FFAs is highly probable during the heating up of oils, which can possibly hinder the functionalization process, and, thus, its removal or

Table 1. Sample Codes for the Developed Coatings[a]

sample code	wt % of AWCO	wt % of silane
AWCO-30S	70	30
AWCO-35S	65	35
AWCO-40S	60	40
AWCO-45S	55	45

[a] The gel and biomass contents of the bioresin formulations are tabulated in Table S1.

reduction becomes necessary.[25] A decrease in the acid value from 0.7 to 0.3 was observed upon reaction with zeolite for 5 h. There was no further decrease in the value with a higher reaction time or zeolite concentration, and, hence, the purified WCO with an acid value of 0.3 was utilized for further reactions.

Functional Group Analysis. EWCO was determined to have an OOC of 5.86 and a percentage epoxidation of 61.75, which is comparable to a percentage epoxidation of 62.9 reported for pure sunflower oil.[26] This shows that the functionalization of WCO was as successful as that for pure vegetable oil.

Fourier transform infrared (FTIR) spectra of WCO, EWCO, and AWCO are depicted in Figure 2a. All of the spectra consist of $-\mathrm{C{=}O}$ stretching peaks in the range of 1730−1750 cm^{-1} arising from the ester group in the triglycerides, doublets in the range of 2800−3000 cm^{-1} from symmetric and asymmetric C−H stretching of methylene groups, and peaks at 1381−1459 cm^{-1} corresponding to −C−H scissoring and rocking vibrations of the long alkane chain. The peak at 719 cm^{-1} in WCO corresponding to the $-\mathrm{C{=}C}-$ bending vibration is seen to have reduced in intensity for EWCO, whereas the peak at 3006 cm^{-1} corresponding to the C−H symmetric stretching vibration of $\mathrm{C{=}C{-}H}$ disappeared in the spectra of EWCO, which shows that the unsaturation in WCO decreased due to some apparent reaction. The appearance of a peak at 836 cm^{-1} in EWCO corresponds to the symmetric C−O−C stretch, confirming the formation of an epoxy ring. A peak at 810 cm^{-1} and that at 1630 cm^{-1} in AWCO corresponding to the vinyl group and $-\mathrm{C{=}C}-$ stretch of the acrylate group showcase successful acrylation of the epoxy groups. A peak at 987 cm^{-1} due to the $\mathrm{C{=}C}$ bend of monosubstituted alkene also confirms the same. Further, the disappearance of the peak corresponding to the epoxy group at 836 cm^{-1} shows the complete conversion of epoxy groups to acrylate groups. A broad peak in the range of 3000−3500 cm^{-1} corresponding to −OH stretching and a peak at 1053 cm^{-1} corresponding C−O stretching of the primary alcohol group are also observed in AWCO due to formation of the −OH group at the opening of the epoxy ring. A peak in the range of 1100−1300 cm^{-1} corresponding to the −C−O stretch in ester is present in both WCO and EWCO. Figure 2b shows the ^{1}H NMR spectra of WCO, EWCO, and AWCO. The peaks in the range 5−5.5 ppm corresponding to protons of unsaturation present in WCO are seen to have decreased in EWCO, whereas new peaks are formed in the range 2.9−3.5 ppm corresponding to epoxy protons. The appearance of acrylate group protons in the range 5.9−6.5 ppm and the complete disappearance of epoxy proton peaks are also observed in AWCO, which confirms the efficient acrylation of WCO.

The percentage of acrylation was calculated to be 71.32% from the NMR spectrum using the formula

$$\%\ \text{acrylation} = \frac{\text{area under the acrylate peak}}{\text{area under (acrylate peak + epoxy peak + peak for unsaturation)}} \times 100$$

FTIR spectra of AWCO and its emulsion (Figure 3a) are devoid of any significant differences except for an increase in the intensity of the hydroxyl peak due to the presence of water, which signifies the absence of any undesirable side reactions.

FTIR spectra of all of the coated samples and those for all of the cured resins are seen to overlap, due to the absence of any additional functional groups or difference in grafting positions among them (Figure S2). Figure 3b shows a comparison of FTIR spectra of uncoated kraft paper and AWCO- and AWCO-35S-coated paper. The intensities for the peaks at 810 and 1630 cm^{-1} attributed to the acrylate groups are seen to reduce after curing, along with the appearance of a weak N−H bending peak at 1576 cm^{-1}, proving the reaction of the amine group in APTES with an acrylate moiety via Michael addition.[27] A broad and intense peak in the range of 1000−1170 cm^{-1} is noticed in AWCO-S, which may be attributed to the overlapped peaks of Si−O−Si and Si−O−C stretching formed by silanol self-condensation and its condensation with cellulosic hydroxyl groups. These analyses confirm the

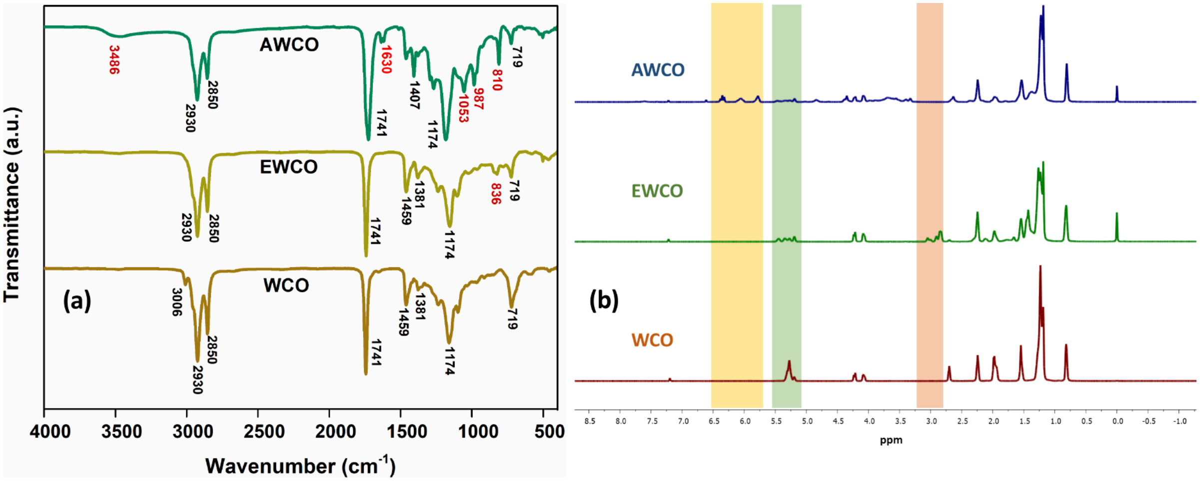

Figure 2. (a) FTIR spectra of WCO, EWCO, and AWCO. (b) ^{1}H NMR spectra of WCO, EWCO, and AWCO.

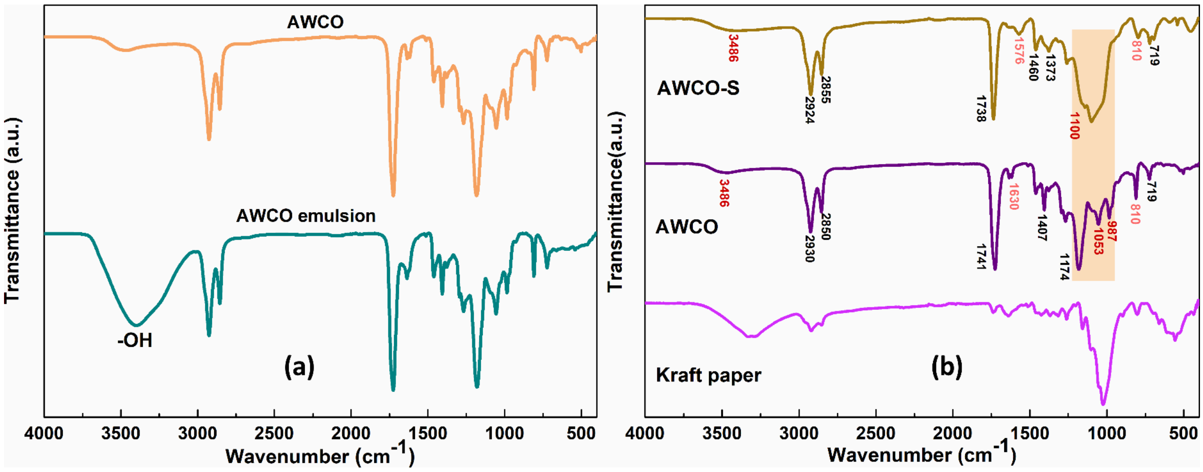

Figure 3. (a) FTIR spectra of AWCO and AWCO emulsion, with the emulsion showing the retention in acrylate groups. (b) FTIR spectra of pristine kraft paper and AWCO- and AWCO-35S-coated paper, showcasing the utilization of acrylate groups and grafting of a silane moeity.

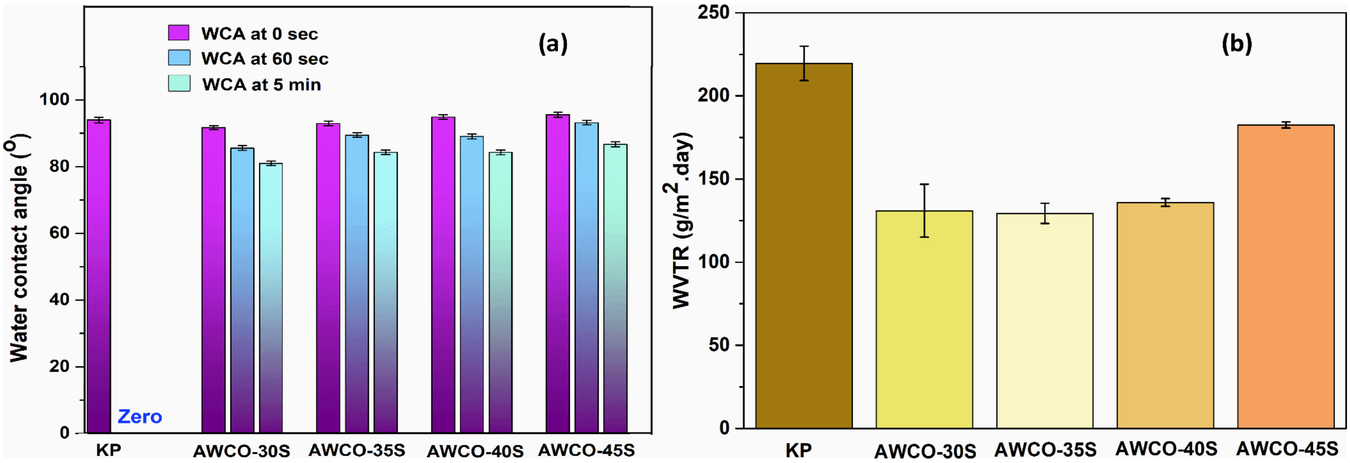

Figure 4. (a) Variation in the WCA with time. A slight reduction was observed for all of the coated specimens, whereas a drastic decrease to zero was observed for the uncoated kraft paper. (b) Plot showing the WVTRs of the samples.

successful grafting of APTES onto AWCO and the subsequent grafting of this onto the paper surface.

Water Resistance. Water resistance or hydrophobicity is a crucial property for any packaging material because the packaged contents have to be made free from water exposure to safeguard its quality. Figure 4a shows the water contact angles (WCAs) for all samples and its variation with time, while the initial contact angles of the samples are shown in Figure S3. A high WCA was observed for the uncoated paper right after dropping the water droplet onto the surface, which reduced drastically to an unmeasurable value in less than a minute. The high WCA observed initially must be due to the surface roughness imparted by the projections of cellulose fiber on pristine paper, whereas the inherent hydrophilicity of the same led to the diffusion of water into the microporous substrate within a few seconds.[28,29] All of the coatings exhibited an initial WCA of >90°, where a gradual increase in the WCA up to 95.6° was observed with an increase in the silane content. In contrast to the uncoated paper, all of the coated samples exhibited significant retention in the contact angle with time and maintained a contact angle above 80° even after 5 min, showcasing their ability to hold water. This retention in the contact angle can be attributed to the micropores on the substrate being covered by the coatings, which allows the water droplet to remain on the surface instead of getting seeped into the fiber matrix.

The WVTRs of the uncoated and coated samples were determined to understand the effect of these coatings on the barrier properties of the paper (Figure 4b). A decrease in water vapor transmission was obtained for all of the coated samples, due to coverage of the porous surface of paper with the resin, which hinders the passage of vapor molecules across the thickness of the substrate.[30] Although this surface coverage is applicable to all of the coated samples, a higher degree of vapor barrier obtained for AWCO-35S and AWCO-40S may be attributed to the higher content of APTES, leading to better grafting of the acrylate moieties with the silane. However, a further increase in APTES (as in AWCO-45S) may cause self-condensation and agglomeration of the silanols, leading to a nonuniform cross-linking within the polymeric matrix, which, in turn, causes a decrease in the vapor barrier.[31]

Oil Resistance. Kit rating was performed to check the resistance of the coatings toward grease, which is a necessary facet for oily or fried food packaging. The uncoated paper showed the lowest value of 0/12, whereas all of the coated samples showed excellent oil resistance with the highest kit value of 12/12, which is comparable to that of the commercial plastic-coated paper.[32,33]

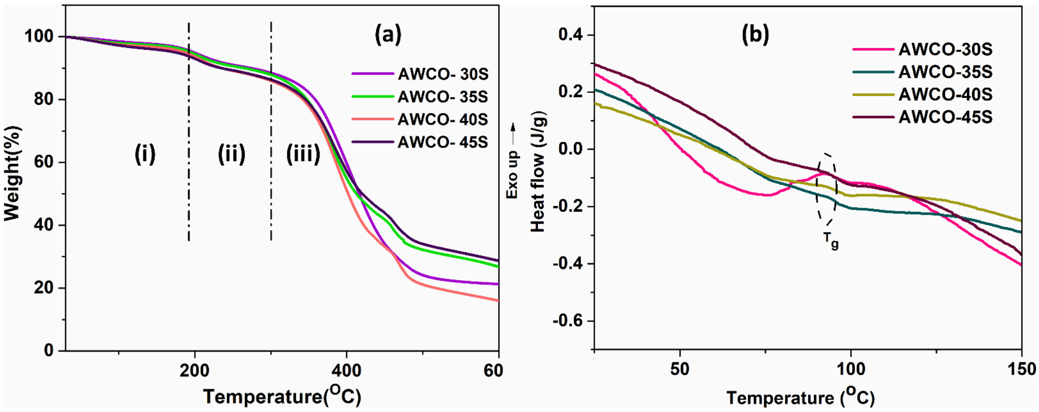

Figure 5. (a) Thermogravimetric analysis: plot of weight loss against temperature. (b) DSC curves of the cured resins showing their respective phase transitions.

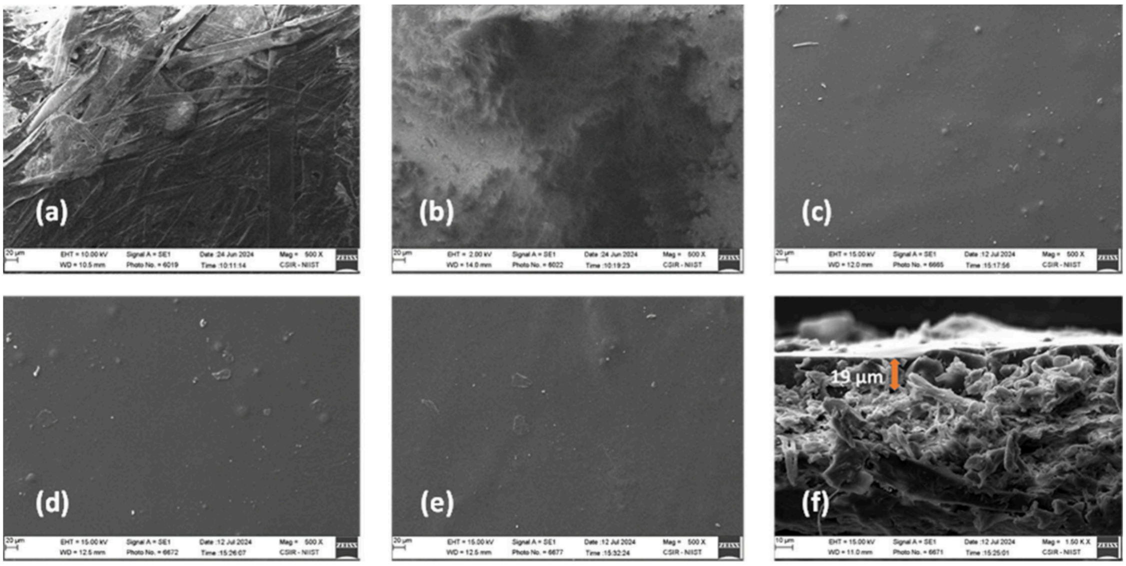

Figure 6. SEM micrographs of the coatings: (a) kraft paper; (b) AWCO-30S; (c) AWCO-35S; (d) AWCO-40S; (e) AWCO-45S. (f) Cross-sectional image of AWCO-35S.

Thermal Analysis. Thermal characteristics of the cured resins are given in Table S2, and the degradation regions are specified as (i), (ii), and (iii) in Figure 5a. T_{onset} of all of the samples is higher than 160 °C, indicating its high thermal stability and suitability in the packaging of hot food. This degradation in region (i) might correspond to the loss of water molecules entrapped within the cured resin. A slight decrease in T_{10} observed with an increase in the silane content may be explained as due to the degradation of self-condensed silanol moieties in the temperature range of 200−300 °C [region (ii)]. The major weight loss occurring in the range 380−410 °C, corresponding to T_{max} [region (iii)], can be ascribed to the degradation of multiple regions in the cured resin, namely, the acrylated oil,[33] and the breakage of Si−O and Si−C bonds resulting in mass fragmentations of the moieties.[34]

Differential scanning calorimetry (DSC) plots of the cured resins (Figure 5b) show the glass transition temperature (T_g) in the range of 93−94 °C, which confirms its application potential in hot food packaging. This high T_g signifies minimal segmental mobility among the polymer chains due to a highly cross-linked network. Less precision in the curve for T_g can also be attributed to the presence of this highly cross-linked network. Further, the absence of any exothermic peak in the plots dismisses the chance of any residual curing, which signifies complete curing of the coatings.

Mechanical Properties. The tensile strength and elongation at break for the coated papers are explained in Figure S4.

Transparency. Transparency of the coatings is an important aspect for specific applications that require aesthetic appeal. The UV−visible absorbance spectra and percentage transmittance are discussed in Figure S5 and Table S3.

Scanning Electron Microscopy (SEM). SEM images of the uncoated and coated papers are depicted in Figure 6 to compare the morphology and surface coverage of the pores. These images reveal the successful surface coverage by the coatings. The pristine kraft paper (Figure 6a) has visible fiber strands over the surface, revealing its coarse surface morphology, which lends an initial water retention effect, as is evident by its initial WCA. The surface coverage of AWCO-30S (Figure 6b) is seen to be partial due to an uneven distribution of the resin on the surface. Because the coating grammage of all of the samples lies in the same range, it can be interpreted that this uneven surface coverage is a result of absorption of the uncured resin due to its lesser solid content. However, the rest of the samples as depicted in Figure 6c−e

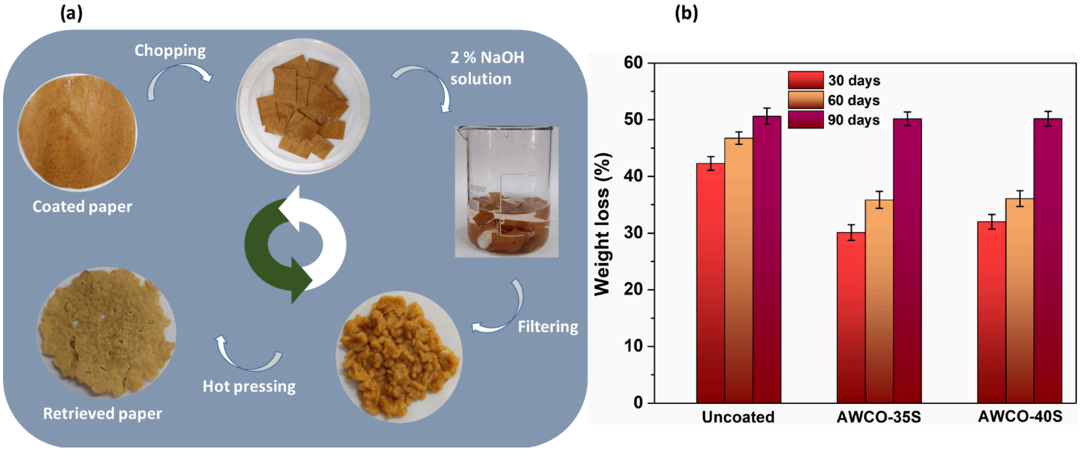

Figure 7. (a) Retrieving pristine paper from the coated paper by reaction with a NaOH solution. (b) Comparison of the weight losses of the samples when buried in a vermicompost.

are found to have uniform surface coverage by the resins, which blocks the protrusion of hydrophilic cellulose fiber strands toward the surface, which, in turn, can be correlated with the hydrophobicity and water retention in these coatings. However, the pristine kraft paper, being devoid of such coverage, lacks water retention and hydrophobicity. The cross-sectional SEM image of AWCO-35S (Figure 6f) shows a coating thickness of around 19 μm, revealing easy processability in tuning the thickness of the coatings. The coating layer is also seen to adhere well to the substrate, with no phase separation visible between the two, confirming strong bonding of the resin with cellulose fibers.

Heat Sealing. In addition to water and oil resistance, sealing of the coated papers is also an important requirement for the manufacture of products like paper cups, pouches, and takeaways. This sealing, if performed without the use of an external glue or an additional plastic layer or heat seal laquer, is always preferable in reducing the production cost and carbon footprint. All of the coated papers could be sealed adequately under a hot press machine at a temperature of 150 °C and a pressure of 20 bar when the coated papers were subjected to UV-ozone treatment for 15 min prior to the sealing (Figure S6). UV-ozone treatment was performed to improve the adhesive performance of the papers via the generation of high energy activated oxygen in the presence of ozone. These oxygen radicals can abstract the hydrogen atoms from the polymer chains, whereby radical atoms are formed on the surface.[35,36] These radical atoms, being highly reactive, promote the bonding of the treated surface to another surface. This results in the bonds between sealed substrates being of chemical nature, aiding in its high strength and stability.[15] The heat seal strength of the bonded papers analyzed by a lap shear test could not yield any value because breakage occurred within the substrates with all of the seal joints remaining intact. This confirms that all of the samples could be bonded with a high sealing strength under the above-mentioned sealing conditions.

Icephobicity. Resistance to ice/cold products is a necessary property for the storage of frozen/cold food. Icephobicity of the coated paper was analyzed and is represented in Figure S7a. It is evident from the results that the uncoated kraft paper started to absorb water (melted from the ice), whereas the coated counterpart remained intact with no visible trace of absorption. This phenomenon is well aligned with the expected outcome because icephobicity is mostly related to hydrophobicity and superhydrophobicity. A surface that is resistant toward water is also expected to be resistant toward ice because they possess similar surface tension.[37] Hence, the developed coatings are suitable for cold storage of food as well.

Hot Water/Oil Resistance. Parts b and c of Figure S7 show the ability of the coated specimen to withstand oil and water in its heated state. The liquids were heated to 80 °C and abruptly dropped on the surface of the coated and uncoated paper substrates, respectively, which was then wiped off using a cotton pad after 10 min of contact time. The uncoated paper is seen to absorb both liquids, with a significant absorption of oil observed in the first few minutes itself, whereas the coated paper withstands both liquids, with no visible sign of absorption. This proves the efficiency of the developed coating in the application of oily and aqueous hot food storage.

Bonding Strength. Figure S8 represents the WCA of AWCO-35S after the tape peel test. It is evident from the figure that there is only a minor decrease in the contact angle even after 50 peeling cycles, showcasing the high bonding strength of the coating with the paper substrate.

Repulping Potential. Repulping of the coated paper for its recycling is a very crucial process to certify the sustainability and circular economy of the coating. Recycling of the coated paper was achieved by complete removal of the coating and recovery of the kraft paper by treatment with an alkali solution. The coated paper was chopped into small pieces and agitated at 200 rpm in a 2% NaOH solution at 80 °C for 1 h. The mixture of loosened paper was allowed to settle, washed with water to remove the excess NaOH solution, and filtered. The recovered pulp was hot-pressed at a temperature of 90 °C for 10 min, and the dried paper thus obtained was characterized using FTIR (Figure S9). The peaks at 1741 and 810 cm^{-1} corresponding to the carbonyl stretching and C═C stretching frequencies, respectively, are found to disappear in the repulped paper, proving the absence of AWCO-S. The spectra for repulped paper are found to overlap substantially with that of pristine kraft paper, indicating the complete recovery of kraft paper. A high recovery yield of ~90 % was also obtained, indicating the practical feasibility of this repulping process (Figure 7a). The removal of coatings can be explained by the breakage of Si−O bonds because these bonds undergo cleavage in an alkali medium.[38] This results in the separation of the silane-grafted AWCO moiety from the cellulose surface and hence leads to complete removal of the coating.

Soil Burial Test. All of the coated papers along with the uncoated kraft paper were tested for their degree of compostability by burying them in a vermicompost and measuring the respective weights at regular intervals of time at a temperature of 25−32 °C and a relative humidity of 79−85%. Figure 7b shows the percentage degradation of each of the samples with time. It can be observed that degradation for the uncoated paper is higher than that for the coated papers during the initial days, which is obvious. However, the rate of degradation increases with time for the coated samples, and all of the coated samples degrade to almost the same extent (50%) as the uncoated paper by the end of 90 days. This persistence in the remaining 50% of uncoated paper can be attributed to the presence of synthetic internal sizing agents like alkylketene dimer and alkenylsuccinic anhydride in the paper, which can hinder its biodegradation.[39] This can be explained by the fact that the coatings exhibit an initial resistance to microbial attack due to its rigid and high barrier nature, which eventually gets overcome by the degradation potential of microbes like *Bacillus* sp., *Pseudomonas* sp., etc., to break the ester linkages present in the coatings.[21] This degradation analysis shows that the coatings almost exhibit complete degradation in vermicompost, revealing its compostable nature.

The major properties of the developed coating are compared with those of the published works in Table 2. As shown in the table, there are no reported works providing all of the major properties required for packaging applications. It is also noteworthy that even though some of the properties are similar to the reported data, the major raw materials used by them are from first-generation biomasses, creating a competition with the food market. On the contrary, this work uses a second-generation biomass, which not only does not pose a threat to the food market but also eliminates the problem related to its disposal and wrong handling.

Table 2. Comparison Chart of Various Biobased Paper Coatings and Their Key Properties

reference	WCA (deg)	kit rating	WVTR	repulpability	compostability
waterborne AESO[20]		7/12		93% recovery	compostable
UV-cured AESO[33]	90−92	12/12			
silanized castor oil[24]	97		200−250	recovered	
AELO-Beeswax[21]	111.3		20.4	recovered	compostable
suberin dispersion[28]	62.6	12/12	18		
current work	93	12/12	129.3	90% recovery	compostable

CONCLUSION

In this work, a biobased and ecofriendly barrier coating is developed as a replacement for single-use plastic lamination on cellulosic substrates. The bioresin for this coating was derived from WCO, a major source of food waste, and coated on paper for food packaging. Acrylated WCO was synthesized and emulsified using PVA, following which it is bonded with cellulosic hydroxyl groups on the paper through condensation. A high contact angle of ∼93−95° could be obtained by increasing the silane content up to 35−40% (of the base emulsion), whereas a WVTR reduction of more than 40% was obtained for the same samples. The coatings are highly oil-resistant with observation of the highest possible kit rating value of 12 and have high bonding strength and thermal stability. Both water and oil resistances are observed in the coated papers in both hot and cold conditions. The coatings are removable in an alkali environment, enabling easy repulpability, and also compostable in vermicompost soil under ambient conditions. The developed coating exhibits heat sealing after UV-ozone treatment, which is useful for making packaging products without the use of an additional sealing layer. The bioresin coating paved a way for the conversion of WCO into a value-added ecofriendly product for food packaging to be ecofriendly and sustainable.

ASSOCIATED CONTENT

Data Availability Statement

Data will be made available on request.

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acssusresmgt.4c00392.

> Experimental details, analytical data, figures, transparency, and mechanical properties of the coated papers (PDF)

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Notes

The authors declare no competing financial interest.

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