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RESEARCH ARTICLE

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Compositional analysis of multilayered plastic constituents and constituent mixtures using benchtop ^{1}H NMR spectroscopy

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Abstract

Multilayered plastics are widely used in food packaging and other commercial applications due to their tailored functional properties. By layering different polymers, the multilayered composite material can have enhanced mechanical, thermal, and barrier properties compared to a single plastic. However, there is a significant need to recycle these multilayer plastics, but their complex structure offers significant challenges to their successful recycling. Ultimately, the use and recycling of these complex materials requires the ability to characterize the composition and purity as a means of quality control for both production and recycling processes. New advances and availability of low-field benchtop ^{1}H NMR spectrometers have led to increasing interest in its use for characterization of multicomponent polymers and polymer mixtures. Here, we demonstrate the capability of low-field benchtop ^{1}H NMR spectroscopy for characterization of three common polymers associated with multilayered packaging systems (low-density polyethylene [LDPE], ethylene vinyl alcohol [EVOH], and Nylon) as well as their blends. Calibration curves are obtained for determining the unknown composition of EVOH and Nylon in multilayered packaging plastics using both the EVOH hydroxyl peak area and an observed peak shift, both yielding results in good agreement with the prepared sample compositions. Additionally, comparison of results extracted for the same samples characterized by our benchtop spectrometer and a 500-MHz spectrometer found results to be consistent and within 2 wt% on average. Overall, low-field benchtop ^{1}H NMR spectroscopy is a reliable and accessible tool for characterization of these polymer systems.

KEYWORDS

benchtop NMR, multilayer film, polymer blend

1 | INTRODUCTION

Plastics are everywhere, and their ubiquity has led to growing concerns over the sustainability of these popular materials.[1] Multilayer plastic packaging materials exacerbate these concerns as the constituent polymers are often derived from nonrenewable resources, while their complex and varied composition makes them difficult to

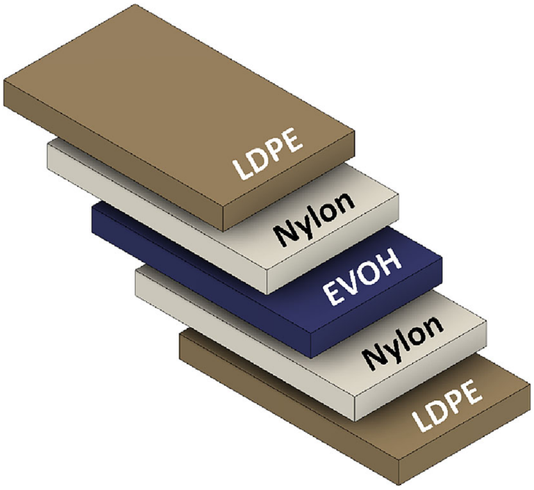

FIGURE 1 Five-layered packaging material with adhesive layers between the individual polymers.

recycle.[2] Packaging plastics are important as they help to preserve the safety and quality of the material enclosed in it including many foods.[3] The multilayered structure of these materials allows the composite materials to have superior properties, such as being tough and flexible but with reduced air and water transport across the film to improve shelf life of the packaged material.[4,5] Multilayer plastics come in many architectures—such as the common five-layered architecture shown in Figure 1—with the different plastic layers bonded together with tie layers. Typical polymers found in multilayer food packaging are[6,7]:

i. Ethylene vinyl alcohol (EVOH) serves as an oxygen barrier to avoid oxidation of the packaged material (e.g., food)
ii. Polyamides (PA) improve the mechanical strength of the packaging material and is resistant to oils and greases.
iii. Low-density polyethylene (LDPE) is an excellent moisture barrier and has significant toughness.

Unlike single material systems, multi-material recycling is difficult and requires separating each of the constituent plastics.[8] Furthermore, these multilayered packaging plastics are bound together with adhesives and tie layers and other additives that would affect the recycling process. Successful recycling of multilayer plastic packaging remains elusive at the commercial scale, though there has been progress toward demonstrating feasibility of different processes.

Solvent-based extraction systems have gained interest for commercial recycling of different mixed polymer streams,[2,9–12] but these methods are intensive and require additional antisolvents and cosolvents to recover the extracted plastic. The solvent-targeted recovery and precipitation (STRAP) strategy uses different compatible solvents to deconstruct the multilayer films and a series of selective solvent extractions to recover the pure plastics. Essential to the STRAP process is the determination of compatible solvents and antisolvents, which can be a very intensive process that is aided by the use of solubility parameter analysis. The Hildebrand solubility parameter estimates the solubility of a polymer based on the cohesive energy density of that polymer.[13,14] A solvent that has a similar cohesive energy density exhibits similar intermolecular bonding and is more likely to dissolve that polymer. Essentially, solubility parameters are based on the principle that “like dissolves like,” meaning that solvents with similar solubility parameters are more likely to be compatible and dissolve each other. While useful, these solubility parameters are only an approximation and require experimental validation for choosing a solvent for a given process.

The solubility of a polymer in a particular solvent is insufficient for effective separation; the solvent must also be selective toward that given polymer to successfully remove it from the multilayer film. Both solubility and purity can be assessed using ^{1}H nuclear magnetic resonance (NMR) spectroscopy.[15–18] ^{1}H NMR spectroscopy is a useful and standard technique for characterizing polymer chemical structures or performing compositional analysis on copolymers or polymer mixtures due to the presence of distinct protons along the polymeric repeat units.[13,16] Recent advances have paved the way for the development of low-field or benchtop NMR spectrometers. Benchtop NMR spectrometers offer numerous advantages with the primary ones being they (1) do not require cryogenic cooling and super conducting magnets to generate the magnetic field and (2) do not require the use of deuterated solvents to obtain spectra due to the application of software algorithms to address spectral drift. Taken together, these two advantages make low-field spectrometers cheaper to purchase with lower maintenance costs compared to high-field instruments. This has correspondingly seen increased interest and use materials characterization.[16,19–23]

For instance, benchtop NMR spectroscopy has received attention in the pharmaceutical sector for quality control and contamination detection of drug products and other applications like quality control in the food sector and in monitoring chemical reaction systems as well.[24–28] Benchtop NMR spectroscopy has also been applied to polymer systems for compositional analysis of copolymers and polymer blends. For instance, the experimental accuracy of benchtop NMR spectroscopy (60 MHz) for characterizing the extracted 1,4-polyisoprene was determined to be with 1% of that obtained using a higher field strength (400 MHz) NMR spectrometer based on the number of scans required for analysis. Benchtop ^{1}H NMR spectroscopy serves as a compact yet reliable tool to characterize polymeric structures and compositional analysis in binary as well as multicomponent systems.[16]

However, there are corresponding disadvantages to using low-field NMR spectrometers that one must take into consideration, with the primary disadvantage being the inherently greater spectral dispersion due to the lower magnetic field that results in greater overlap of peaks in the NMR spectra.[19,22–24] This is a long-standing issue with the characterization of polymers with NMR spectroscopy in general (even for high-field NMR spectrometers) due to the slight differences in the proton environments on the long polymer chains compared to those for discrete smaller molecule analogs. This greater peak width for polymers in general has long precluded the quantitative analysis of polymers or polymer mixtures whose NMR spectra have peaks in close proximity. This issue is exacerbated by the additional peak broadening for low-field NMR spectrometers that is essentially a result of the difference in relative frequency space for which the protons are detected. Essentially, both high-field and low-field spectrometers collect the same information; however, this information is collected across different ranges of frequency space. When these data are then converted into the same range of ppm space for analysis, the lower frequency range results in broader peaks with less peak definition. This disadvantage has led many to discount the use of low-field NMR spectroscopy for polymer materials. However, one must simply do their due diligence to validate the use of these instruments for their systems of interest. This is analogous to what one must do even for the use of high-field NMR spectrometers for polymer systems with similar overlap issues due to the abovementioned inherently broader NMR spectra peaks for polymers.

Here, we demonstrate the use of benchtop NMR spectroscopy for the characterization of polymers and polymer blends relevant for multilayer film plastics recycling. The polymers of interest—LDPE, ethylene vinyl alcohol (EVOH), and Nylon (a polyamide)—were selected due to their common use in multilayer packaging, and a series of solvents are chosen based on their potential for use in solvent-based recycling processes. Benchtop NMR spectroscopy was then used to develop a calibration standard for determining mixture composition that could be used in the quality control of the recycling process of multilayer films made using these polymers.

2 | MATERIALS AND METHODS

Formic acid (88%) was purchased from BDH Chemicals, benzene (98%) was purchased from Beantown Chemicals, and toluene (99.5%) were purchased from VWR Chemicals BDH. Tetramethylsilane (TMS) was purchased from Acros Organics Chemicals (99.9%), deuterated DMSO was purchased from VWR, and the NMR spectroscopy tubes were purchased from Wilmad-Labglass.

Commercial polymers pellets (LDPE, EVOH, Nylon) were used. The LDPE obtained has a density of 0.925 g/cc and melt index of 0.8 g/min at 190°C/2.16 kg. EVOH

TABLE 1 Hildebrand solubility parameters for series of solvents.[13,14]

	$\delta_{solvent} - \delta_{polymer}$ (MPa$^{(1/2)}$)			
			Nylon	
Solvent	**LDPE**	**EVOH**	**Nylon 6**	**Nylon 6,6**
Tetrahydrofuran	1.46	6.65	0.86	6.35
Toluene	**0.17**	7.95	2.16	7.64
Benzene	**0.51**	7.60	1.81	7.30
Ethylene glycol	14.96	6.84	12.63	7.15
Trichlorobenzene	3.32	4.80	0.99	4.50
Acetone	1.94	6.18	0.39	5.87
Dimethylformamide	6.87	1.25	4.54	0.95
Formic acid	6.94	**1.18**	4.61	**0.88**
Phenol	6.10	2.01	3.78	1.71
Acetic acid	3.37	4.75	1.05	4.44

Note: The bold values highlight the solvents used in this study.

(32/68 mol% polyethylene/polyvinyl alcohol copolymer) had a density of 1.19 g/cc with a melt index of 3.2 g/min at 190°C/2.16 kg. Nylon 6/6,6 had a density of 1.12 g/cc with a melt index of 3.3 g/min at 190°C/2.16 kg.

2.1 | Sample preparation for NMR spectroscopy

EVOH and Nylon characterization samples were prepared by dissolving 2 g each in 10 ml formic acid in separate scintillation vials, heated to 50°C, and were stirred at 600 rpm until complete dissolution. The obtained spectra were referenced to formic acid's proton attached to the carboxylic acid group at 8.30 ppm.

The blends of EVOH and Nylon were prepared by mixing known weight fractions dissolved in 10 ml formic acid. For LDPE, the dissolved solution was prepared by dissolving 1.5 g in 100 ml benzene (at 70°C) or toluene (at 105°C) as solvent, stirred at 600 rpm. Three to four drops of TMS were added as reference for LDPE spectra analysis. The EVOH and nylon in benzene samples were prepared by dissolving 2 g each in 50 ml benzene (at 70°C).

For high-field ^{1}H NMR spectroscopy, a coaxial insert tube system was used where deuterated DMSO was used in the outer tube (5 mm ID) as reference standard and the polymeric solution to be analyzed was present in the inner tube (3 mm ID).

2.2 | ^{1}H NMR spectroscopy

^{1}H NMR spectra were acquired using Oxford Instruments Pulsar 60 MHz (1.4 T) spectrometer. The dissolved polymers were added with three to four drops of TMS (reference). The solutions were analyzed by doing a ^{1}H FID process on Spinflow 2.3.0. Each experiment used a filter file of 5208, 32 scans, and a recycle delay of 120 s with typical acquisition times of 1 h and 10 min. The spectra were analyzed using MestReNova software, and each spectrum was baselined prior to analysis. The high-field NMR spectra were obtained using Bruker 500-MHz spectrometer.

3 | RESULTS AND DISCUSSION

3.1 | Characterization of LDPE, EVOH, and Nylon

Dissolved virgin polymers in their respective compatible solvents were analyzed using the benchtop NMR spectrometer. A comparison of solubility parameters,

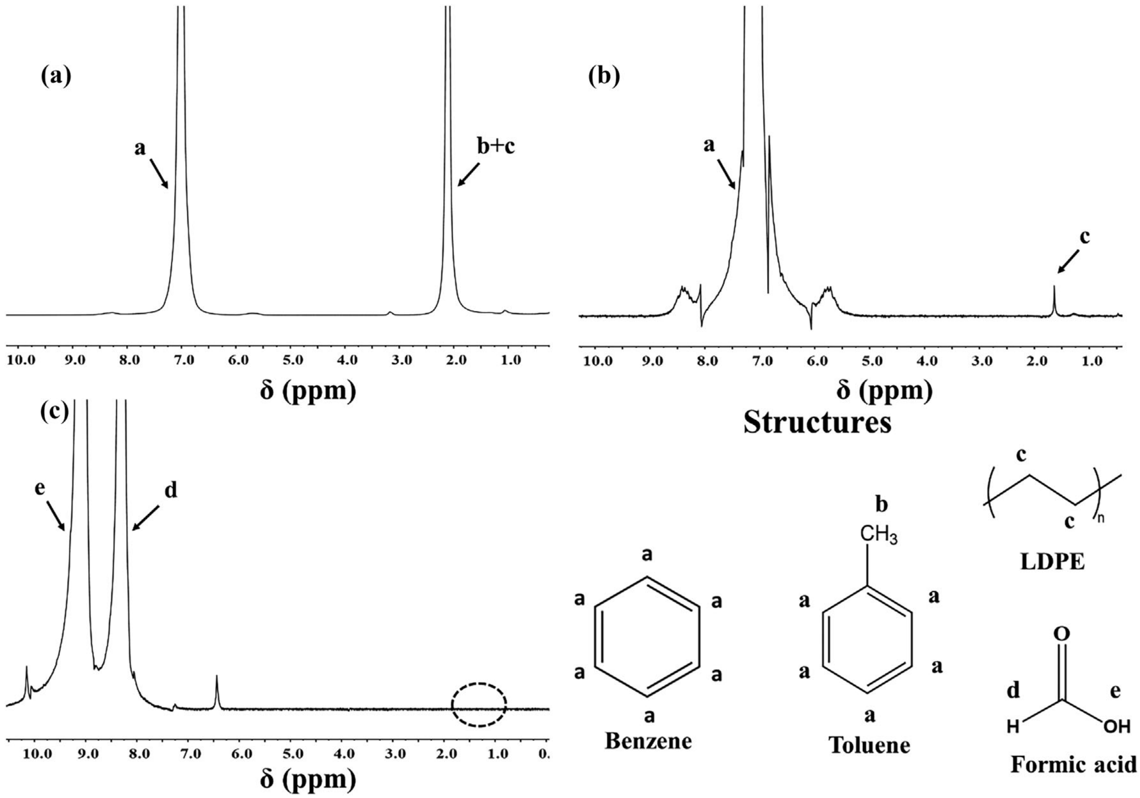

FIGURE 2 Benchtop ^{1}H NMR spectra of LDPE in (a) toluene, (b) benzene, and (c) formic acid.

and in particular the solubility parameter difference ($\delta_{solvent} - \delta_{polymer}$) (MPa$^{(1/2)}$), was first performed to identify the solvents of interest (Table 1) for our target polymers of LDPE, EVOH, and Nylon. In lieu of an exhaustive presentation of solvent solubility parameters, an abbreviated list of common solvents is shown in Table 1 for consideration here. From this list, we identified three solvents of particular interest for investigation: toluene, benzene, and formic acid. Benzene and toluene were chosen due to their small solubility parameter differences with LDPE (0.17 and 0.51, respectively) as well as their large solubility parameter differences with EVOH and Nylon. From this solubility parameter analysis, one would expect LDPE could be separated from EVOH and Nylon by a solvent extraction process with either toluene or benzene. Conversely, formic acid was chosen due to its small solubility parameter differences with EVOH and Nylon and its large solubility parameter difference with LDPE.

LDPE polymer pellets were dissolved in either toluene or benzene at 50°C and the solutions analyzed using the benchtop NMR spectrometer (Figure 2). Unfortunately, there is significant overlap in the aliphatic protons (from 1 to 2 ppm) between toluene and LDPE such that the LDPE peaks are obscured by the very large toluene methyl proton peak. However, this is not an issue with benzene (Figure 2b), which has only the aromatic protons.

We selected formic acid as a target compatible solvent for EVOH and Nylon, based on the solubility parameter differences in Table 1, and so we first confirm the inability of LDPE to dissolve in formic acid (Figure 2c). LDPE was stirred in formic acid at 50°C for 4 h, after which there was essentially no visible change to the solid phase and the solvent phase was sampled for characterization by benchtop 1H NMR spectroscopy. The resulting spectra (Figure 2c) contain only the characteristic peaks for formic acid at 8.3 and 9.24 ppm (also note the presence of a satellite peaks at ~6.5 and 10.1 ppm) and TMS with no discernable signal in the aliphatic region for the LDPE protons. This confirms the insolubility of LDPE in formic acid, a desirable result for the potential of formic acid as a selective solvent for nylon and/or EVOH.

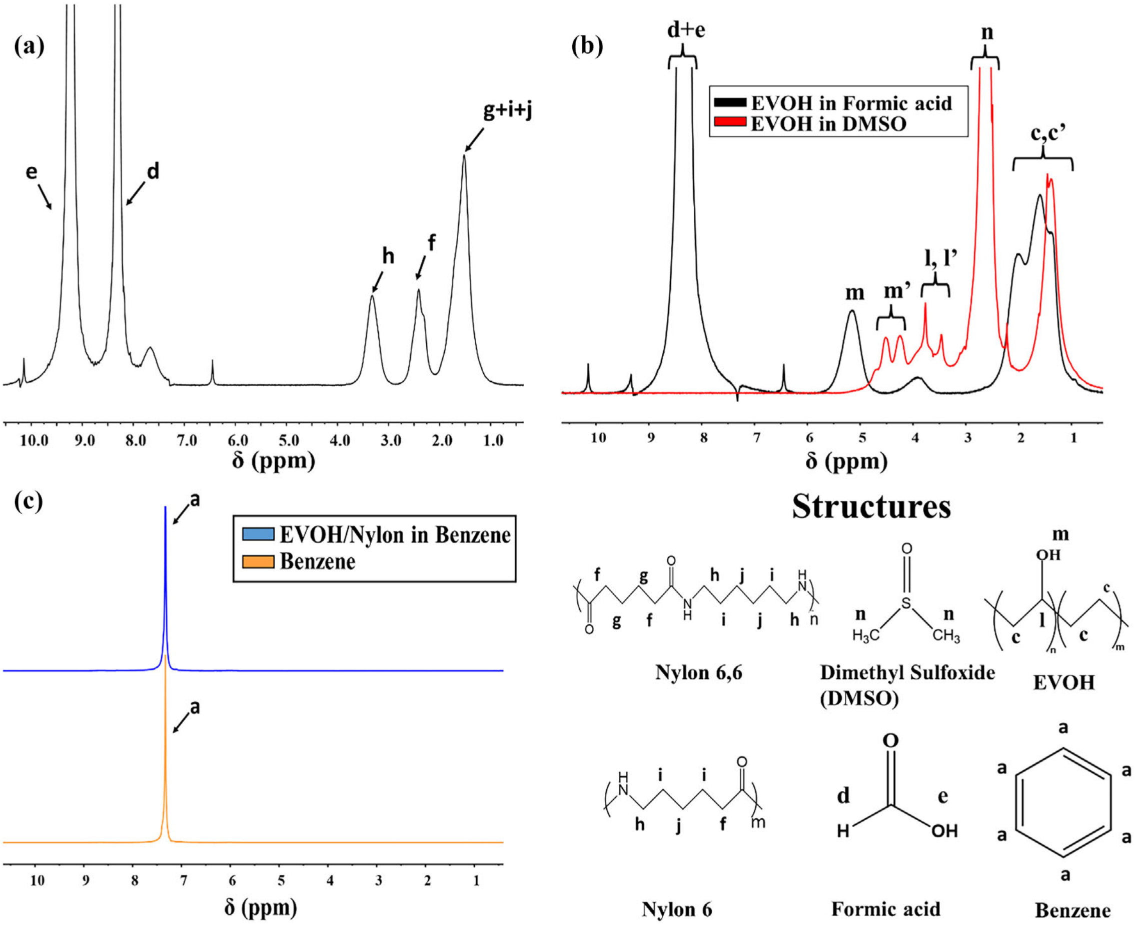

FIGURE 3 Benchtop ^{1}H NMR spectra of (a) Nylon copolymer (6/6,6) in formic acid.[30,31] (b) EVOH in formic acid and DMSO (where c', l', and m' represent EVOH's proton signals in formic acid and c, l and m in DMSO).[32] (c) Blend of EVOH and Nylon in benzene.

A similar analysis was performed for EVOH and Nylon in both benzene and formic acid to confirm both their incompatibility with benzene and their compatibility with formic acid (Figure 3). Toluene is disregarded for EVOH and Nylon, due to the above discussed overlap between the toluene methyl protons and the LDPE protons. The spectra for both Nylon (Figure 3a) and EVOH (Figure 3b) are more complex than that of LDPE due to the presence of more dissimilar protons in their structures. However, there are several discernable and well-separated peaks in the spectra obtained using the benchtop spectrometer. Note that DMSO is conventionally used as a solvent for NMR spectroscopy characterization of EVOH; however, herein, we are interested in using formic acid as the solvent, and to our knowledge, peak assignments for EVOH in formic acid have not previously been reported. Figure 3b shows the EVOH spectra for both formic acid and DMSO as the solvent. The peaks are then assigned based on the relative shielding and deshielding effects of the dissimilar protons associated with the ethylene and the vinyl alcohol groups within the EVOH copolymer. Notably, for our characterization purposes, the formic acid peaks are well separated from both the EVOH and Nylon peaks, and there are distinct EVOH and nylon peaks that also do not overlap. The hydroxyl peak in EVOH (labeled “m” in Figure 3) is shifted due to the partial hydrogen bonding with the formic acid hydroxyl and thereby appears as a single merged peak—a point we will return to below. We note also that the Nylon characterized here is a commercial product utilized in multilayer food packaging and it itself a mixture of Nylon 6 and Nylon 6,6, complicating the analysis somewhat due to the presence of similar protons on their structures. Also, the Nylon amide proton peak is at 8.1 ppm,[29] in close proximity to the formic acid hydroxyl proton. This prevents characterization of the Nylon-containing blend composition using the amide proton when formic acid is the solvent (or otherwise present in the mixture). Unfortunately, due to the similarity of the protons in these two Nylon structures and the lower resolution of the benchtop ^{1}H NMR spectrometer, we were unable to distinguish enough of the protons for compositional characterization. Lastly, a blend of EVOH and Nylon (2 g each) in 50 ml benzene (at 70°C) was prepared, and the resulting spectra (Figure 3c) contains no discernable peaks that would indicate the presence of either EVOH or Nylon. This confirms their insolubility (at least for these conditions) in benzene and thereby the potential of benzene as a selective solvent for LDPE from mixtures of LDPE–EVOH–Nylon.

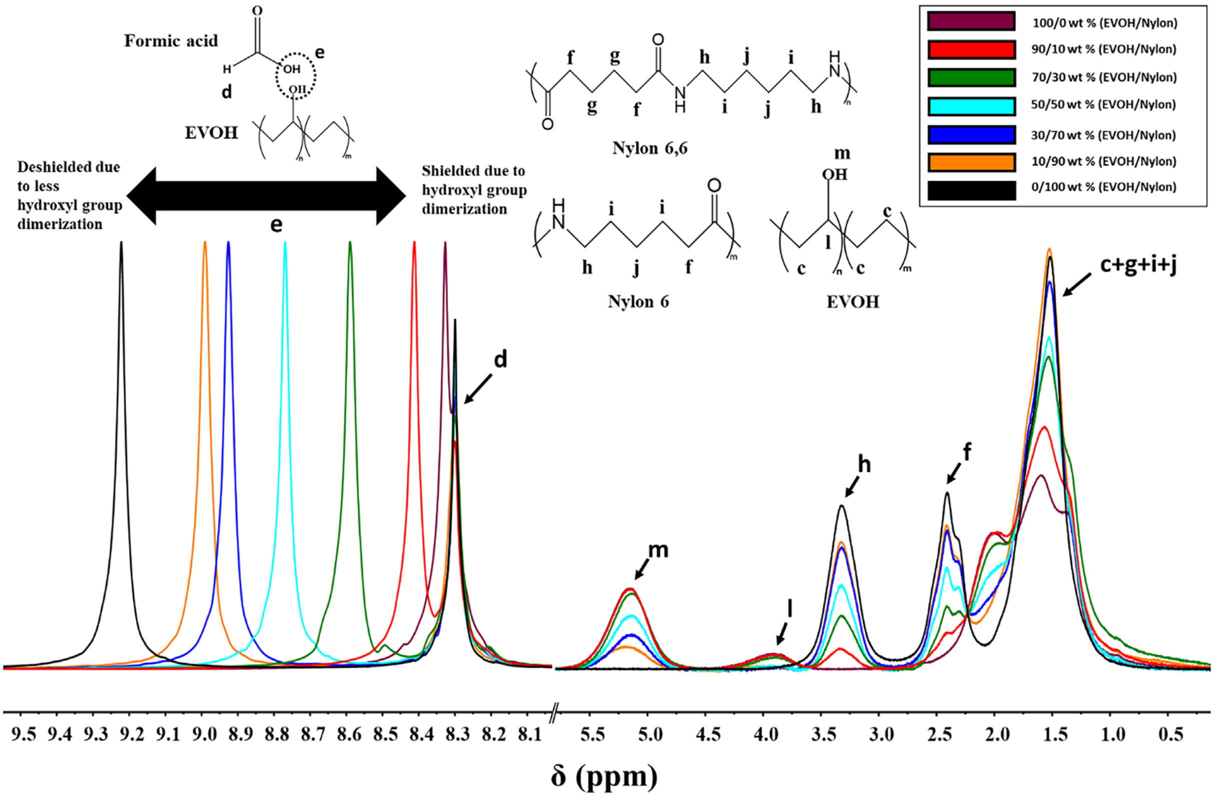

FIGURE 4 Variation of signal strength due to change in composition of EVOH and Nylon in the solution mixture with shielding and de-shielding effects on the hydroxyl proton (labeled “e”) in formic acid due to dimerization with EVOH.

3.2 | EVOH/Nylon blend compositional analysis

To access the effectiveness of low-field benchtop ^{1}H NMR spectroscopy for compositional analysis of EVOH/Nylon blends in formic acid, we prepared a series of formic acid solutions with varied EVOH/Nylon content. The polymers were weighed proportionally into vials and fully dissolved in formic acid prior to characterization using the benchtop ^{1}H NMR spectrometer. A subset of the resulting spectra are shown in Figure 4 (some spectra not shown for clarity; see Figure S1 for these additional spectra). The stacked spectra plot in Figure 4 clearly show the variation in the representative peak intensity/size as the composition is varied, with EVOH peaks increasing with EVOH content and Nylon peaks increasing with Nylon content. For example, the peak at 4.7–5.6 ppm corresponding to the hydroxyl proton on EVOH (labeled “m” in Figure 4) clearly increases with increasing EVOH content in the blend solution.

For determining the composition of the EVOH/Nylon, a calibration curve based on this hydroxyl proton is perhaps the simplest analysis and was performed here. The calibration curve was obtained using the obtained absolute integrated intensity for this peak from the spectra and normalized by the absolute intensity of this peak extracted from the 100 wt% EVOH spectra. From a quality control standpoint, a product solid sample of unknown composition could be dissolved in formic acid at this set concentration (2 g polymer per 10 ml formic acid) and the resulting EVOH hydroxyl peak intensity used to determine the blend composition (wt% EVOH). The obtained calibration curve is shown in Figure 5a, with 5 of the 11 data points within 1.5 wt% EVOH of the calibration curve, and an average deviation between the calibration data and the calibration line wt% EVOH is only ±2.9%.

Furthermore, to test the utility of the calibration curve, “unknown” blends—20/80, 50/50, and 90/10 EVOH/PA wt%—were prepared, and the integrated intensity of the EVOH hydroxyl proton peak was used with the calibration curve to calculate the blend composition. This calculated composition is plotted against the prepared composition in Figure 5c, where the average deviation from the known value was 2.2%. A similar procedure was followed to obtain a calibration plot (Figures S2 and S3) with the high-field (500 Mhz) ^{1}H NMR spectrometer. The data for the calibration curve for the high-field data contain fewer data points and produces similar results where the average deviation between the data and the calibration line is 3%. Considering the potential for slight error in the prepared composition from gravimetric preparation of these blends, for both the calibration and the unknowns, this is a very good result that we consider to be confirmation of the ability to perform this analysis using a benchtop spectrometer.

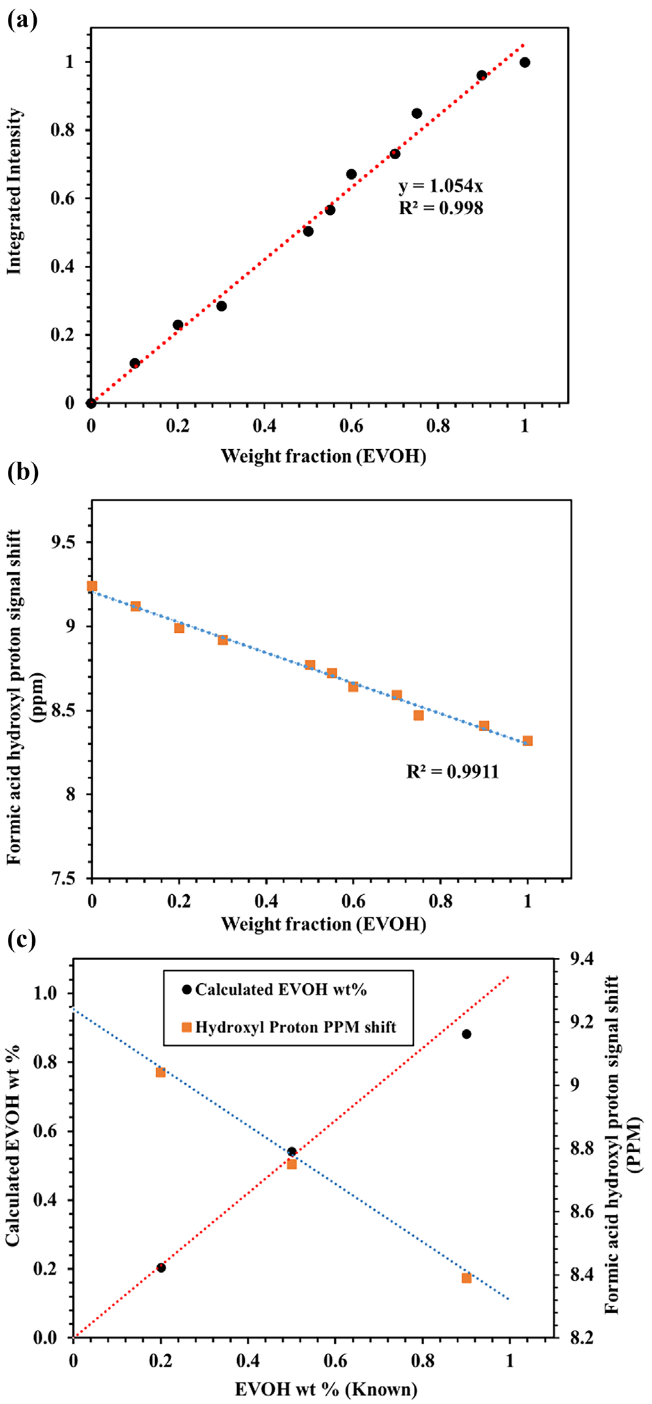

FIGURE 5 Calibration curve. (a) Integrated intensity versus weight fraction of EVOH in solution. (b) Formic acid hydroxy proton signal shift with increasing EVOH content. (c) Calculated EVOH weight percent of unknown samples from the calibration curves (a) and (b).

Interestingly, during this analysis, we observed a shift in the formic acid peak (Figure 4). It was seen that, the peaks corresponding to the carboxylic acid proton and the hydroxyl group in the formic acid both shift with increasing EVOH content in the solution. The signal corresponding to the hydroxyl proton in formic acid experiences a noticeable shielding effect. We attribute this shielding effect to dimerization of the hydroxyl proton in the formic acid due to the partial electronegativity of the hydroxyl oxygen in the polyvinyl alcohol repeat units of the EVOH copolymer. Thus, increasing in the concentration of EVOH in the solution, increases the dimerization of the molecules forming clusters and results in this observed spectral shift. Upon close analysis of this shift, we found that it corresponded linearly with the composition (Figure 5b) such that calibration plot could be constructed using the hydroxyl peak shift. For these calibration data, the average deviation between the data and the calibration line was only 2.6%. This provides another route toward determining the relative composition of EVOH from a mixture toward determining feed or product purity in the recycling of multilayer plastics. This could be also verified as this shielding effect decreases with increase in the Nylon concentration in the solution and showing de-shielding effects.

To directly compare the above analysis for samples characterized using both the benchtop and the 500-MHz spectrometer, two samples (50/50 and 90/10 EVOH/Nylon) were prepared and characterized using both spectrometers. For this tube-in-tube NMR, tubes were used (3 mm in 5 mm) to characterize the exact samples in both spectrometer (no solvent and deuterated DMSO in the outer tube for the 500-MHz spectrometer, respectively). The two obtained spectra are overlaid in Figure 6. The obtained spectra were then analyzed to determine the composition using the peak calibrations described above (Table 2). As expected, the peaks in the spectra from the 500-MHz spectrometer show greater definition and separation compared to the broad peaks in the spectra obtained from the low-field spectrometer. For the quantitative analysis, the calculated composition using the 500-MHz NMR spectra deviates on average 3.8 wt% from the prepared composition. For the low-field benchtop NMR spectra, the composition was calculated using both the peak area and peak shift methods discussed above. The peak area calibration yields compositions that deviate on average 2.9 wt% from the expectation, while using the formic acid hydroxyl peak shift yields compositions that deviate 2.1 wt% from the expectation. Notably, the analysis for the 90/10 sample yields compositions much closer (within 1%–2%) of the prepared sample composition, whereas the 50/50 yields compositions that are consistent but higher than the prepared sample composition (53, 55, and 57 wt% EVOH). Overall, this analysis finds that the results using these methods for the low-field NMR spectrometer are analogous to those from

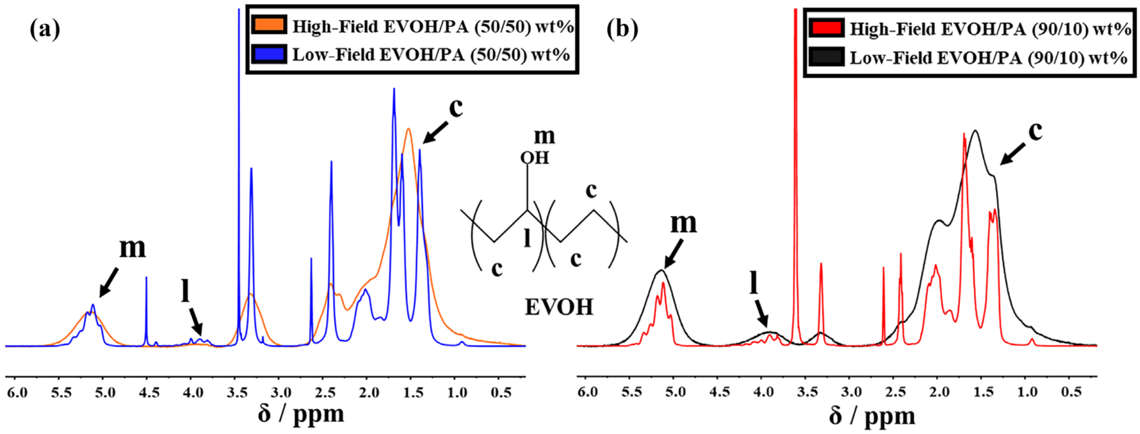

FIGURE 6 Overlay of spectra from benchtop and 500-MHz spectrometer for (a) 50/50 EVOH/Nylon in formic acid and (b) 90/10 EVOH/nylon in formic acid.

TABLE 2 Benchtop NMR versus 500 MHz NMR EVOH wt% calculation analysis.

Composition (EVOH/PA)	EVOH wt% calculated using benchtop spectra peak area calibration curve	EVOH wt% calculated using benchtop spectra peak shift calibration curve	EVOH wt% calculated using 500 MHz spectra peak area calibration curve
50/50	55	53	57
90/10	91	88	91

higher field spectrometers with respect to extracting compositions for mixtures of EVOH/Nylon since when we compare the compositions determined by the low-field and high-field spectra with each other, the results are in good agreement. Namely, we find the same result for the 90/10 ENOG/Nylon (91 wt% EVOH) using the peak area calibrations, whereas the low-field peak shift analysis is slightly lower (88 wt% EVOH), and for the 50/50 EVOH/Nylon, all results are within 4%.

4 | CONCLUSIONS

We investigated the use of benchtop low-field ^{1}H NMR spectroscopy of target constituents from multilayered plastics in the context of solvents of interest for solvent-based recycling systems. Polymer (LDPE, EVOH, Nylon) and polymer blend (EVOH/Nylon) solutions were characterized using a 60-MHz benchtop ^{1}H NMR spectrometer. For the polymer solutions, the spectra confirmed the expected dissolution of each polymer in the solvents (benzene, toluene, and formic acid) chosen based on a Hildebrand solubility analysis. For the polymer blend solutions of EVOH and Nylon in formic acid, calibration plots were constructed based on the EVOH hydroxyl proton and an observed shift in a formic acid peak where calibration data were found to be linear and characterized "unknown" samples yielded compositions in good agreement (within 2–3 wt% on average depending on calibration used) with the prepared "unknown" compositions. Analogous samples were also characterized using both the low-field and a 500-MHz spectrometer where good agreement was obtained (within 2 wt% on average) between the compositions determined from the high-field and low-field spectra. Overall, this work demonstrates how benchtop ^{1}H NMR spectroscopy can be a useful and reliable tool for analyzing some polymers and polymer blends both in our research laboratories and in the context of commercial quality control.

AUTHOR CONTRIBUTIONS

Conceptualization: Harrish Kumar Senthil Kumar, Yoorae Noh, Adam L. Bachmann, and Bryan S. Beckingham. *Methodology*: Harrish Kumar Senthil Kumar, Yoorae Noh, Adam L. Bachmann, and Bryan S. Beckingham. *Software*: Harrish Kumar Senthil Kumar. *Validation*: Bryan S. Beckingham. *Formal analysis*: Harrish Kumar Senthil Kumar. *Investigation*: Harrish Kumar Senthil Kumar. *Resources*: Bryan S. Beckingham. *Data curation*: Harrish Kumar Senthil Kumar. *Writing—original draft preparation*: Harrish Kumar Senthil Kumar and Bryan S. Beckingham. *Writing—review and editing*: Harrish Kumar Senthil Kumar, Yoorae Noh, Adam L. Bachmann, and Bryan S. Beckingham.; *Visualization*: Harrish Kumar Senthil Kumar. *Supervision*, Bryan S. Beckingham. *Project administration*: Bryan S. Beckingham. *Funding acquisition*: Bryan S. Beckingham. All authors have read and agreed to the published version of the manuscript.

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CONFLICT OF INTEREST STATEMENT

There are no conflict of interests to declare.

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