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Delamination of end-of-life photovoltaic modules as key for effective recycling

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Abstract: The delamination of ethylene–vinyl acetate (EVA) encapsulant is a critical step for the recycling of end-of-life (EoL) photovoltaic (PV) modules. In this study, a solvent-based approach was systematically investigated to understand EVA–solvent interactions and to evaluate differences between pristine EVA and EVA recovered from EoL PV modules. An initial screening of fifteen organic solvents was conducted using standard pristine EVA at ambient temperature to assess the solubility behavior and establish baseline polymer–solvent compatibility. Hansen solubility parameter (HSP) analysis was employed to support the experimental observations and to guide the selection of promising solvents. These candidate solvents were subsequently applied to EVA samples extracted from EoL PV modules, including crosslinked EVA, under both ambient and elevated temperature conditions to evaluate delamination efficiency. Furthermore, thermogravimetric analysis (TGA)/differential scanning calorimetry (DSC), and Fourier-transform infrared spectroscopy (FTIR) were used to characterize the thermal and chemical differences between pristine and EoL EVA. The results provide insights into how degradation and crosslinking influence EVA solubility and identify solvent systems with potential for effective delamination of EoL PV modules.

1. Introduction

As the photovoltaic (PV) industry continues to expand globally, the volume of end-of-life (EoL) PV modules is increasing at an unprecedented rate, creating a growing environmental burden and significant challenges for waste management systems. Current projections estimate that cumulative global PV waste could reach approximately 78 million tons by 2050 [1]. Crystalline silicon (c-Si) PV modules, which dominate the market, contain valuable materials such as tempered glass, high-purity silicon, silver, aluminium, and copper, making recycling both an environmental necessity and an economic opportunity. However, efficient recovery of these materials remains challenging due to the presence of the polymeric encapsulant, ethylene vinyl acetate (EVA), which strongly bonds the module layers together. EVA exhibits high adhesion strength, chemical stability, and a partially crosslinked structure formed during the lamination process, properties that are further reinforced during long-term outdoor exposure through thermal aging, UV irradiation, and oxidation [2,3]. These characteristics significantly hinder component separation, limiting the effectiveness of conventional mechanical and thermal recycling routes and often leading to material downcycling or energy-intensive treatment steps [4]. As a result, advanced recycling strategies combining mechanical, thermal, and chemical approaches are increasingly being explored to improve PV module delamination efficiency and enhance material recovery yields [2,5].

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EVA is a random copolymer of ethylene and vinyl acetate widely used in PV modules to encapsulate silicon cells and bond them to glass and back sheet layers, providing mechanical integrity and long-term environmental protection. From a recycling perspective, the selective removal of EVA remains a critical bottleneck, particularly for EoL modules in which the encapsulant has undergone significant degradation, crosslinking, and loss of solubility during service life [3,6]. These aging-induced changes alter the polymer's chemical structure and swelling behaviour, complicating solvent penetration and dissolution. Unlike conventional low-molecular-weight solute–solvent systems, polymer–solvent interactions are governed by complex thermodynamic and kinetic factors related to polymer chain length, crystallinity, crosslink density, and solvent diffusivity. The dissolution of EVA therefore often proceeds through intermediate swollen or gel-like states rather than complete solubilization, making the selection of appropriate solvents and operating conditions particularly critical for solvent-based delamination processes [5,6]. A detailed understanding of EVA–solvent interactions is thus essential for designing effective and scalable chemical recycling routes for PV modules.

In this work, a solvent-based chemical delamination approach is systematically investigated to evaluate the dissolution behaviour of EVA encapsulants. A range of organic solvents is assessed to examine polymer–solvent interactions and to compare the behaviour of pristine EVA with EVA extracted from EoL PV modules. In addition, thermogravimetric analysis (TGA) and Fourier-transform infrared spectroscopy (FTIR) are employed to characterize the thermal stability, degradation pathways, and chemical structure of the different EVA samples. This combined experimental framework provides insight into how aging and crosslinking influence EVA solubility and supports the identification of promising solvents for efficient delamination and material recovery from EoL PV modules.

2. Methods

A systematic solvent-based methodology was employed to study polymer–solvent interactions and facilitate delamination of end-of-life (EoL) PV modules. Standard pristine EVA was first used to screen a range of organic solvents at ambient temperature, followed by comparative dissolution studies on EVA extracted from EoL PV modules. Representative images of pristine EVA, extracted EoL EVA and a typical EoL PV module are shown in figure 1 (a–c). Selected solvents were subsequently evaluated at elevated temperatures to assess their delamination efficiency. Thermal and chemical characterization techniques were applied to both pristine and EoL EVA to correlate material properties with solvent performance.

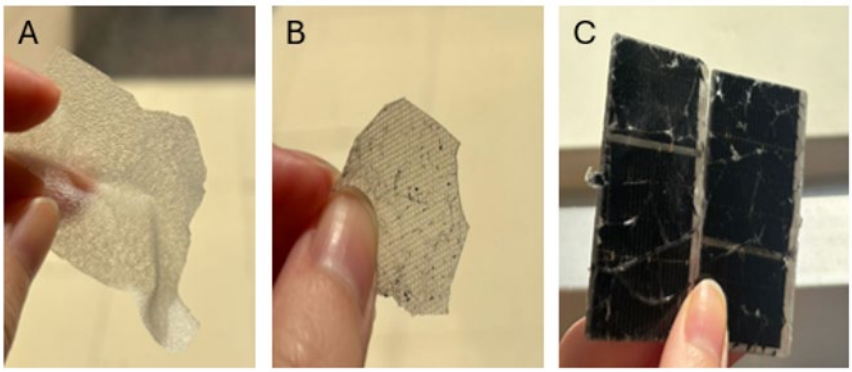

Figure 1: a) Standard pristine EVA (section 2.1), b) Extracted EVA from EoL PV (section 2.2), c) Typical EoL PV module (section 2.3)

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2.1 Assessment of pristine EVA in different solvents using HSPiP software at ambinat temperature

The chemical delamination of EoL PV modules was initially explored through screening experiments with a range of fourteen organic solvents. These experiments aimed to evaluate the dissolution behavior of the EVA encapsulant. The Cold/ambient dissolution test was performed on pristine EVA by immersing samples in 30 mL of solvent at ambient temperature under continuous stirring. The EVA-to-solvent molar ratio was fixed at 1.5%, and experiments were conducted for four hours. The extent of dissolution was primarily assessed through visual and viscosity change inspection to determine whether the EVA had dissolved in each solvent.

To support experimental work and identify the suitable solvents for EVA, the Hansen Solubility Parameters in Practice (HSPiP) software [7] was used to predict EVA–solvent compatibility with various organic solvents. The solubility parameter (δ) of a compound, originally defined by Hildebrand (1936) as the square root of its cohesive energy density c_i (Eq. 1), describes the energy required to overcome intermolecular forces during dissolution.

$$\delta_i = \sqrt{c_i} = \left(\frac{\Delta_{vap}H_{m,i}RT}{V_{m,i}}\right)^{0.5} \quad (1)$$

where $\Delta_{vap}H_{m,i}$ is the enthalpy of vaporization of the compound i, R is the universal gas constant, T is the absolute temperature and $V_{m,i}$is the molar volume of the i compound. While useful, Hildebrand's single-parameter approach only accounts for dispersive interactions. Therefore, Hansen [8] extended this concept by dividing δ into three contributions: dispersion (δ_d), polar (δ_p), and hydrogen bonding (δ_h), such that the overall parameter δ_i is given by Eq. (2).

$$\delta_i^2 = \delta_{d,i}^2 + \delta_{p,i}^2 + \delta_{h,i}^2 \quad (2)$$

where δ_i indicates the solubility parameter of a solvent i. The polymer dissolution occurs when the solubility parameters of the polymer and the solvent are sufficiently close. The affinity between EVA and different solvents was quantified using the interaction distance R_a and the relative energy difference (RED), solubility.

$$R_a = \sqrt{4\,(\delta_i - \delta_p)^2 + (\delta_i - \delta_p)^2 + (\delta_i - \delta_p)^2} \quad (3)$$

$$RED = \frac{R_a}{R_O} \quad (4)$$

where subscripts i and p refer to solvent and polymer, respectively, and R_0 is the interaction radius of the polymer. A solvent is classified as "good" when RED < 1 and "poor" when RED ≥ 1. This model-based approach, provides a theoretical basis for identifying solvents with high affinity toward EVA which allows pre-screening of solvents, reducing the need for extensive trial-and-error experiments and guiding solvent selection for EVA dissolution studies. Each solvent is described by Hansen solubility parameters as shown in table 1 and considering dispersion, polar and hydrogen bonding forces [8]. Figure 2 shows a three-dimensional Hansen solubility parameter (HSP) diagram showing the distribution of solvents relative to the polymer solubility sphere. Solvents located within the interaction radius (R_0) are considered compatible with the polymer, whereas solvents outside the sphere exhibit poor solubility. The relative energy difference (RED = R_a/R_0) quantifies solvent–polymer affinity.

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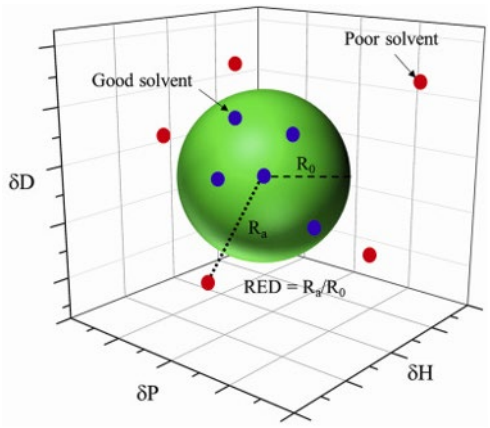

Figure 2: Three-dimensional Hansen solubility parameter (HSP) diagram showing the distribution of solvents relative to the polymer solubility sphere

2.2 Assessment of candidate solvents for EoL EVA (Ambient temperature)

The organic solvents identified as candidate solvents during the initial screening with pristine EVA (section 2.1) were selected for further evaluation using EVA extracted from end-of-life (EoL) PV modules. EVA encapsulant was extracted from three representative EoL PV modules and exposed to each solvent under identical experimental conditions. All dissolution experiments were conducted at ambient temperature with continuous shaking for 4 h to ensure consistent solvent–polymer contact. This procedure was used to assess the interaction between the selected solvents and EoL EVA under room-temperature conditions prior to subsequent high-temperature delamination studies.

2.3 Assessment of candidate solvents for EoL EVA (High temperature)

Based on the ambient-temperature screening, selected candidate solvents were further evaluated for the delamination of end-of-life (EoL) PV modules under elevated temperature conditions. Intact EoL PV laminates were immersed in the selected solvents and treated in a sealed autoclave reactor to enable controlled heating above ambient temperature and promote solvent penetration and EVA softening. The delamination process was conducted in a stepwise manner comprising solvent filling, heating to the target temperature, cooling, and subsequent mechanical separation of the module layers. Experiments were performed under controlled thermal conditions to assess the influence of temperature on EVA–solvent interactions and delamination efficiency. In parallel, the dynamic viscosity of the solvent–EVA mixtures was measured at different operating temperatures to provide an indirect assessment of polymer dissolution behavior during the process.

2.4 Analytical methods

Three representative EoL PV modules were initially subjected to preliminary treatments, manual deframing, water-jet cutting and manual exfoliation to expose and extract the encapsulant EVA layers. Subsequently, these extracted EoL polymeric EVAs (EVA_PV1, EVA_PV2, EVA_PV3) along with a pristine factory sample. These sample were analyzed using different analytical tools using TGA/DSC and FTIR.

2.4.1 Simultaneous TGA/DSC

Thermogravimetric analysis (TGA) coupled with differential scanning calorimetry (DSC) was carried out using a SDT Q600 instrument (TA Instruments) to characterize the thermal stability and decomposition behavior of the samples. Nearly 30mg of EVA material was placed in a platinum pan connected to a high-sensitivity microbalance and heated in a sealed furnace equipped with a thermocouple for precise temperature control. The experiments were performed in an oxygen-rich atmosphere at a constant heating rate of 10 °C/min, with the temperature ramped to

~650–700 °C to ensure complete degradation of the organic components. Throughout the heating process, the instrument simultaneously recorded weight loss and heat flow, providing information on both the mass changes associated with thermal decomposition and the exothermic effects linked to combustion. The high-resolution temporal/thermal data allows accurate tracking of decomposition stages and combustion kinetics under controlled atmospheric conditions.

2.4.2 Fourier Transform Infrared Spectroscopy (FTIR)

The Fourier-transform infrared (FTIR) spectroscopy was carried out using a Bruker Vertex 80v system in an attenuated total reflection (ATR) configuration. A single-reflection diamond/ZnSe crystal was employed as the ATR element. The IR spectra were recorded in a spectral range from 400 to 4000 cm^{-1} wave number with a spectral resolution of 4 cm^{-1}, and 128 scans were averaged to improve the signal-to-noise ratio. The FTIR spectroscopy is a powerful analytical technique based on the interaction of infrared radiation with molecular vibrational modes, providing structural and chemical information about organic and polymeric materials.

3. Results and Discussion

3.1 Solvent–pristine EVA compatibility based on HSPIP predictions

An appropriate solvent must provide sufficiently strong interactions with the polymer to enable dissolution. The HSP approach facilitated the identification of such solvents for EVA. The results of pristine EVA screening in various solvents using HSPiP software are summarized in Table 1 and figure 3. The three-dimensional δD–δP–δH plots show the position of selected organic solvents relative to the EVA solubility sphere, with solvents inside or near the sphere predicted to exhibit stronger polymer–solvent compatibility. Solvents were scored as "1" when complete dissolution was observed (good solvents) and "0" when no or only partial dissolution occurred, the latter also being classified as unsuitable. Based on these results, five solvents including benzene, chloroform, cyclohexane, toluene, and xylene were identified as effective solvents for EVA and are represented within the solubility sphere (see figure 4). In the Hansen solubility sphere, the green dot represents the polymer (EVA). Blue dots correspond to *good* solvents located inside the sphere, indicating closer HSP values to the polymer including benzene, chloroform, cyclohexane, toluene, and xylene. Red dots represent poor solvents positioned outside the sphere, reflecting larger differences in HSP values from the polymer such as acetone, di-chloromethane, methanol, ethyl acetate and acetone ect.

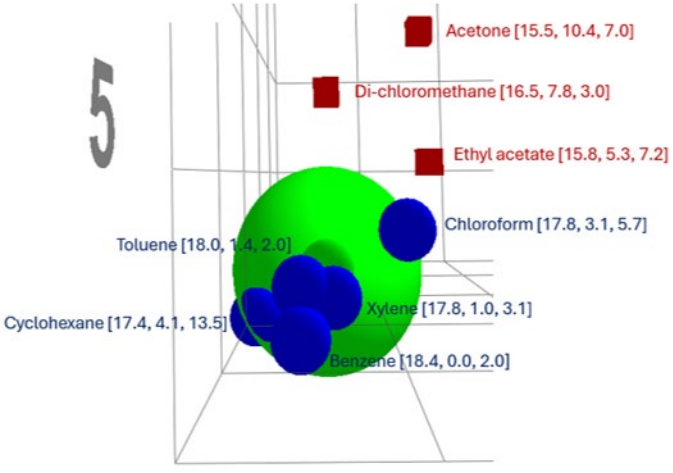

Figure 3: Hansen Plot with estimated Hansen sphere as calculated by HSPiP software

Table 1: Experimental evaluation of pristine EVA solubility in different solvents along with their Hansen solubility parameters

Sample N°	Solvent	Dispersion $\delta_{D,i}$ (Mpa$^{1/2}$)	Polar δ_P (Mpa$^{1/2}$)	Hydrogen bonding $\delta_{H,i}$ (Mpa$^{1/2}$)	RED	Molar Volume	Observation	Score
1	Benzene	18.4	0.0	2.0	0.641	89.5	Dissolved	1
2	Heptane	15.3	0.0	0.0	1.250	-	Not dissolved	0
3	Hexane	14.9	0.0	0.0	1.321	131.4	Not dissolved	0
4	Methanol	14.7	12.3	22.3	2.593	40.6	Not dissolved	0
5	Ethyl acetate	15.8	5.3	7.2	1.001	98.6	Not dissolved	0
6	Dichloromethane	16.5	7.8	3.0	1.217	-	Partially dissolved	0
7	Chloroform	17.8	3.1	5.7	0.496	80.5	Dissolved	1
8	Acetonitrile	15.3	18.0	6.1	2.363	52.9	Not dissolved	0
9	Cyclohexane	16.8	0.0	0.2	0.998	108.9	Dissolved	1
10	Cyclohexanol	17.4	4.1	13.5	3.152	-	Not dissolved	0
11	Toluene	18.0	1.4	2.0	0.688	106.6	Dissolved	1
12	Xylene	17.8	1.0	3.1	0.579	121.1	Dissolved	1
13	Acetone	15.5	10.4	7	1.515	73.8	Not dissolved	0
14	Ethanol	15.8	8.8	19.4	1.991	58.6	Not dissolved	0
15	Water	15.5	16.0	42.3	4.783	18.0	Not dissolved	0

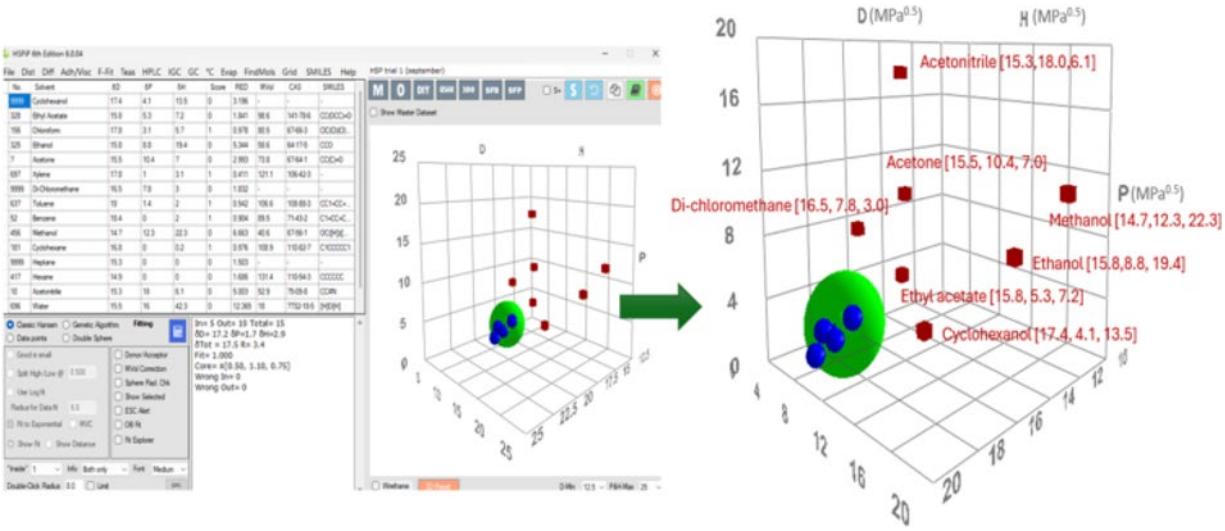

Figure 4: Hansen solubility parameter plot of selected solvents relative to the pristine EVA solubility sphere

3.2 Ambient-temperature solvent interaction with EoL EVA

Five organic solvents: benzene, chloroform, cyclohexane, toluene, and xylene, previously identified as candidate solvents during the initial pristine EVA screening were subsequently evaluated for their ability to dissolve EVA extracted from end-of-life (EoL) PV modules. All experiments were conducted under identical conditions, with EoL EVA samples subjected to continuous shaking for 4 h at ambient temperature. In contrast to the behavior observed for pristine EVA, no dissolution of EoL EVA was observed in any of the selected solvents under these conditions. This lack of solubility is attributed to structural and compositional changes occurring during module service life, including increased crosslinking and the incorporation of fillers and contaminants such as glass dust and additives, which reduce polymer chain mobility and hinder solvent diffusion. These results demonstrate that ambient-temperature, single-solvent treatments

are insufficient for effective EoL EVA delamination and indicate that elevated temperatures and/or extended exposure times are required to facilitate solvent-based delamination.

Table 2: Experimental evaluation of end-of-life EVAs solubility in five candidate solvents at ambient temperature

Sample Nº	Solvent	Observation
1	Benzene	Not dissolved
2	Chloroform	Not dissolved
3	Cyclohexane	Not dissolved
4	Toluene	Not dissolved
5	Xylene	Not dissolved

3.3 High-temperature solvent-assisted delamination of EoL PV Modules

Raising the treatment temperature markedly improved the performance of solvent-based delamination for end-of-life (EoL) PV modules as shown in table 3. At elevated temperatures, aromatic solvents such as xylene and toluene interacted more effectively with the EVA encapsulant, promoting polymer softening and facilitating separation of the module layers. As temperature increased, the dynamic viscosity of the solvent–EVA mixtures also increased, indicating enhanced EVA swelling and partial dissolution. This response highlights the key role of heat in increasing polymer chain mobility and improving solvent diffusion, which are otherwise restricted in aged and crosslinked EVA. These observations confirm that temperature is a critical parameter for effective solvent-assisted delamination and demonstrate the suitability of selected aromatic solvents for high-temperature recycling of EoL PV modules

Table 3: Experimental evaluation of end-of-life EVAs solubility in five candidate solvents at high temperature

Sample Nº	Solvent	Observation
1	Benzene	Not dissolved
2	Chloroform	Not dissolved
3	Cyclohexane	Not dissolved
4	Toluene	Dissolved
5	Xylene	Dissolved

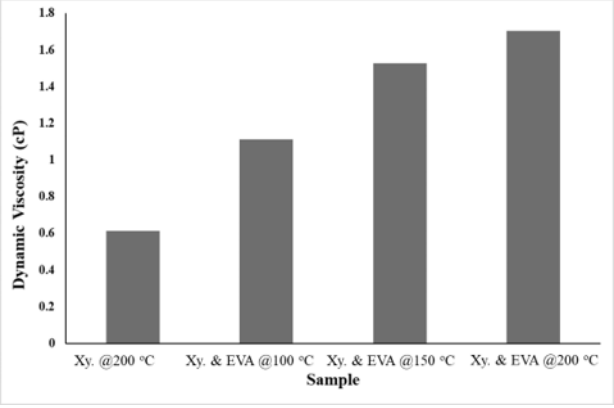

Figure 5: Variation in dynamic viscosity of pure xylene and xylene–EVA mixtures at different operating temperatures

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3.4 Analytical tools results

3.4.1 TGA results

Preliminary thermogravimetric and differential scanning calorimetry (TGA/DSC) results for three EVA samples extracted from end-of-life (EoL) PV modules (EVA_PV1, EVA_PV2, and EVA_PV3), together with a pristine factory EVA reference, are shown in Figure 1. Figure 1a presents the TGA curves, where weight percentage is plotted as a function of temperature over a 70 min heating period. All EVA samples (pristine and PV-derived) exhibit a two-step thermal degradation behavior typical of ethylene–vinyl acetate copolymers. The materials remain thermally stable up to approximately 300 °C. The first major mass-loss event occurs between 330 and 350 °C and corresponds to the deacetylation of vinyl acetate side groups, resulting in the release of acetic acid and accounting for approximately 24% weight loss. The second, more pronounced degradation step takes place between 400 and 500 °C, with a mass loss of roughly 75%, and is attributed to the decomposition of the polyethylene backbone. By 550–600 °C, nearly complete mass loss is observed, leaving negligible residual material.

The extent of deacetylation was quantified based on the weight loss associated with the first decomposition stage. As shown in the inset of Figure 1a, the deacetylation temperature shifts toward higher values with increasing curing temperature of the EVA. This trend suggests that a higher degree of crosslinking strengthens the bonding between vinyl acetate groups and the ethylene backbone, thereby reducing the extent and delaying the onset of deacetylation. In general, EVA with higher vinyl acetate (VA) content undergoes greater deacetylation due to the increased availability of relatively weaker VA linkages, as reported in the literature [9,10].

Figure 1b shows the DSC thermograms of the pristine and EoL EVA samples plotted as heat flow versus temperature. Two main thermal transitions are observed, corresponding well with the degradation steps identified in the TGA analysis. A minor endothermic peak appears in the range of 240–250 °C, with a maximum at approximately 244.56 °C, which is attributed to the initial deacetylation of vinyl acetate side groups. This event is associated with a relatively low heat flow of about 1.8 W g^{-1}, indicating a modest enthalpic contribution. A second, more prominent exothermic peak is observed between 440 and 460 °C, with a maximum near 450.31 °C and a substantially higher heat release of approximately 5.5 W g^{-1}, corresponding to the decomposition of the polyethylene backbone. Although all samples display similar thermal features, subtle differences are evident. The pristine EVA exhibits slightly lower onset and peak temperatures and reduced heat flow intensity, suggesting earlier degradation and lower thermal resistance. In contrast, EVA_PV2 and EVA_PV3 show sharper peaks at higher temperatures, indicating enhanced thermal stability, while EVA_PV1 exhibits intermediate behavior. These DSC results corroborate the TGA findings and demonstrate that, while the fundamental degradation mechanisms remain similar, aging and service-related effects in PV modules influence the thermal stability of EVA.

Table 4: Estimated values of VA and ethelene contents from TGA analysis for EVA samples

Sample	Stage 1 (VA content)	Stage 2 (Ethylene)
Prestin EVA	24.04 %	75.34 %
EVA_PV1	28.29 %	70.93 %
EVA_PV2	30.46 %	68.88 %
EVA_PV3	25.96 %	72.79 %

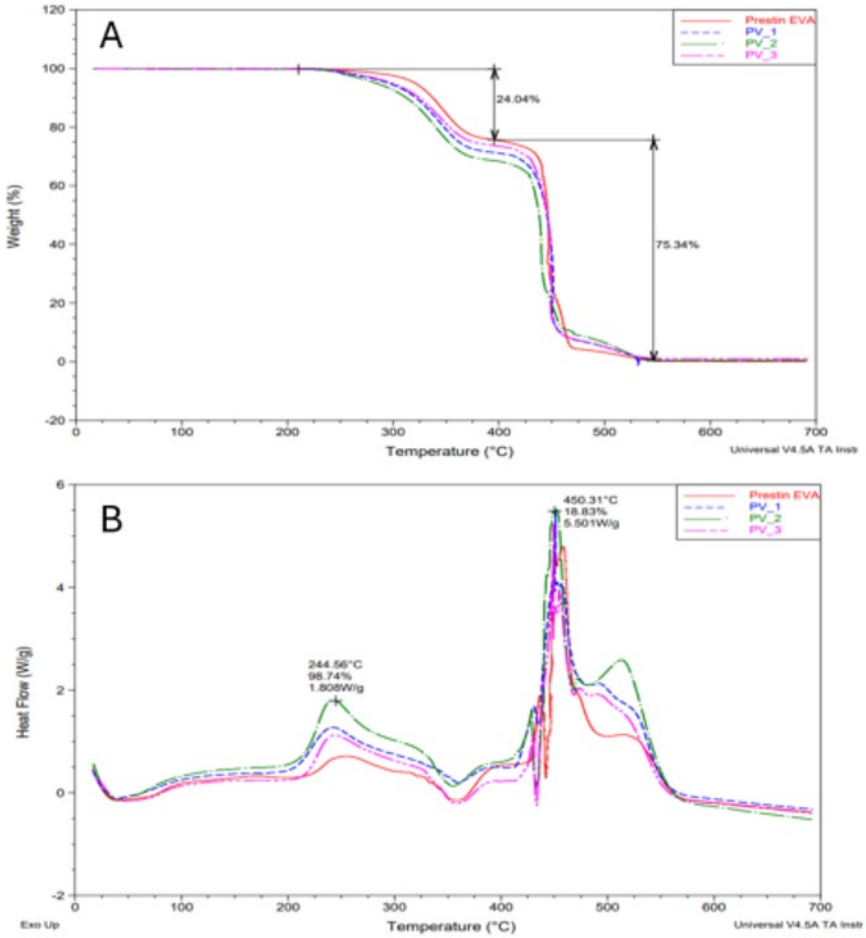

Figure 6: a) TGA curves and b) DSC thermograms of pristine EVA and three extracted end-of-life EVA samples (PV1, PV2, PV3)

3.4.2 FTIR results

The FTIR spectra of EVA 0–3 illustrated in figure 3 display the characteristic bands of ethylene–vinyl acetate, including C–H stretching at 2920 and 2850 cm^{-1}, the ester carbonyl stretch at ~1740 cm^{-1}, C–O stretching in the 1240–1020 cm^{-1} region, and CH_2 rocking vibrations at ~730/720 cm^{-1}. EVA 0 (pristine) shows strong CH_2 rocking bands, indicative of higher polyethylene crystallinity. In the crosslinked samples (EVA 1–3), all major EVA bands remain, confirming that the base polymer chemistry is unchanged; however, the CH_2 rocking bands become progressively less intense, consistent with reduced crystallinity as crosslinking increases. Meanwhile, the carbonyl and C–O bands maintain their positions but show subtle intensity and shape variations, reflecting changes in the local environment of the acetate groups. The spectra demonstrate that crosslinking primarily reduces crystalline order in EVA while preserving the fundamental chemical structure.

Table 5: FTIR peak assignments for EVA samples

Wavenumber (cm^{-1})	Assignment
2920-2850	C–H stretching (CH_2, CH_3)
1740	Ester carbonyl stretch
1240–1020	C–O stretching (ester)
730/720	CH_2 rocking

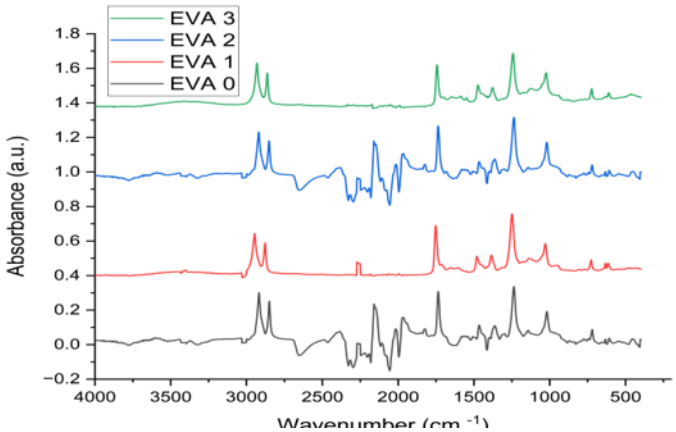

Figure 7: FTIR results of Prestin EVA (EVA 0) and and three extracted end-of-life EVA samples (PV1, PV2, PV3)

Conclusion

A combined experimental screening and Hansen solubility parameter (HSP) analysis clarified the dissolution behavior of EVA. Solvents with small HSP distance, similar to benzene, chloroform, cyclohexane, toluene, and xylene, fall within the EVA solubility sphere and show strong compatibility, matching the observed swelling/dissolution at room (and elevated for EoL EVA) temperature. Complementary TGA/DSC and FTIR indicate that crosslinking lowers crystallinity and raises thermal stability while preserving the EVA backbone, thereby increasing the gel fraction and reducing the soluble fraction. HSP-guided solvent selection combined with multi-technique characterization provides a robust basis for designing efficient, cleaner recycling routes for EVA in PV modules. Crucially, EoL EVA from different sources, though visually appear similar, varies in VA content, crosslink density, and thermal behavior, leading to measurable differences in dissolution across the tested solvents, especially post-crosslinking. This variability underscores the need to report VA wt% and gel fraction and to tailor solvent and temperature windows (e.g., 150–200 °C) to the specific EoL EVA grade and service history.

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