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Organic solvent selection for delamination of end-of-life silicon photovoltaic modules based on recyclability

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Abstract. Photovoltaic modules (PVMs) are already accepted as renewable energy sources to fulfill future energy demand. E-waste from end-of-life (EoL) PVMs will pose significant challenges for both the recovery of valuable materials and the environmental hazards in the near future. Delamination of photovoltaic modules for recycling typically involves separating the solar cell from various layers of PVM that are; glass, encapsulant ethylene-vinyl acetate, and backsheet. The solar cell is encapsulated by ethylene-vinyl acetate (EVA) layers; the photovoltaic module can be delaminated by a swelling encapsulant layer using organic solvents.

The recyclability of organic solvents is investigated in this work, with an emphasis on possible contamination by encapsulated EVA during EoL PVM delamination. To investigate the interactions of encapsulant EVA with solvents, it was extracted from EoL PVMs. This work investigates contamination of organic solvents in the temperature range of 25–55 °C and suggests solvent recyclability for reuse and waste reduction. Organic solvents Fourier transform infrared (FTIR) spectra after a 24-hour encapsulant EVA interaction were compared to study organic solvent contamination. This study focused on two different types of solvents: aromatic hydrocarbons (like xylene) and chlorinated hydrocarbons (like CH_2Cl_2). Xylene demonstrated resistance to EVA contamination, making it a preferred organic solvent for recycling EoL PVMs.

Key words: ethylene-vinyl acetate (EVA), e-waste, organic solvent, photovoltaic modules, recycling, solar cell.

INTRODUCTION

Electronic waste (E-waste), refers to discarded electrical or electronic devices. The amount of E-waste generated globally has been increasing due to the rapid growth in technology. The inadequate manners for recycling/disposal of E-waste can have adverse environmental and health impacts, as well as loss of valuable materials(Ghulam & Abushammala, 2023). Photovoltaic modules (PVMs), commonly known as solar panels, are a key component of solar energy systems. The PVMs convert sunlight into electricity and are widely used for renewable energy generation. The widespread implementation of photovoltaic (PV) technologies in the coming years has the potential to cut global carbon emissions by 50% (Deng et al., 2019; Trivedi et al., 2023b). As PV technology becomes more prevalent, there is a growing concern about the management of end-of-life (EoL) PVMs. The forecast suggests that the global waste produced by PVMs is projected to increase by 60 to 78 million tonnes by 2050 (Ko et al., 2023; Trivedi et al., 2023b). Silicon-based PVMs, both monocrystalline and polycrystalline, have traditionally dominated the solar market. These constitute a significant majority of the global PVM market (Ballif et al., 2022).

The importance of silicon photovoltaic modules (Si PVMs) in today's world cannot be overstated. These modules are the cornerstone of solar energy generation, which is crucial for transitioning towards a sustainable and renewable energy future. As the world seeks to reduce reliance on fossil fuels and mitigate climate change, Si PVMs have emerged as a key solution due to their low cost and easy maintenance(Tan et al., 2022; Trivedi et al., 2023b). Si PVMs reduce dependence on finite and environmentally harmful energy sources like coal, oil, and natural gas. It helps mitigate climate change by reducing carbon dioxide and other pollutants released into the atmosphere. While silicon PVMs have a relatively long lifespan (25–30 years), they eventually reach the end of their operational life (Mao et al., 2024). When decommissioned, these modules contribute to E-waste. The damaged Si PVMs also contribute to E-waste caused due to environmental conditions like hail, storms, and other reasons that cause physical impact on PVMs (Denz et al., 2022; Goudelis et al., 2022). The physical impact/damages lead to a reduction in solar energy production efficiency. The PVM is considered its end-of-life when output efficiency drops to 80% of its starting efficiency (Trivedi et al., 2023b). Efforts are being made to develop recycling technologies for the various materials used in solar panels, including silicon, glass, and metals.

As the Si PVMs industry grows, the volume of end-of-life Si PVMs is increasing, posing challenges for waste management and environmental stewardship. Si PVMs contain valuable materials such as silicon, glass, aluminum, silver, copper wires, and busbars. Recycling these materials reduces the need for virgin resource extraction, conserves energy, and minimizes environmental impacts associated with mining and processing raw materials(Trivedi et al., 2023a, 2023b). Recycling Si PVMs aligns with the principles of a circular economy, where resources are reused, recycled, or repurposed to minimize waste and maximize resource efficiency. By implementing robust recycling infrastructure and processes, the Si PVMs industry can contribute to a more sustainable and resilient energy ecosystem. Policymakers, industry stakeholders, and consumers must collaborate to develop and implement effective recycling strategies that minimize waste, conserve resources, and promote the transition to a circular economy (Fthenakis, 2009; Tan et al., 2022; Trivedi et al., 2023b).

To extract valuable and hazardous materials from solar cells, EoL Si PVM delamination is required. Thermal treatment and organic solvent interaction techniques are frequently used to delaminate EoL PVMs (Trivedi et al., 2023c). Moreover, end-of-life Si PVMs are sometimes delaminated mechanically or using a combination of mechanical, thermal, and chemical methods. Thermal treatment is the process of heating Si PVM in a furnace to a higher temperature (500–600 °C), which causes the encapsulant ethylene vinyl acetate to decompose and facilitates the separation of the module's glass and silicon solar cells. This process necessitates high temperatures, which can consume a lot of energy and contribute to greenhouse gas emissions. Heating the modules requires high equipment and energy costs, especially in large-scale operations(Dobra et al., 2022; Trivedi et al., 2023b).

Mechanical delamination is the physical separation of the layers of a Si PVMs using mechanical force. Shredding, grinding, milling, crushing (mechanical and high voltage), cutting, and peeling can all be used to separate the module's constituent materials (Deng et al., 2022; Trivedi et al., 2023b). Mechanical delamination, on the other hand, results in downcycling products and is not appropriate for delamination with delicate layers of Si PVM (Trivedi et al., 2023a). Delamination with an organic solvent is a promising method for delaminating various layers of PVMs. It does not cause damage to silicon cells and can be used to recover intact silicon wafers (Dias et al., 2021; Xu et al., 2021; Ghahremani et al., 2024). It is also an energy and cost-effective method compared to other methods of delamination(Vaněk et al., 2023). Several solvents have previously been reported to interact with the encapsulant EVA to cause EoL Si PVM delamination. Dibasic ester (DBE), trichloroethylene, d-limonene, toluene, xylene, N,N-dimethylpropenylurea (DMPU), ethylene glycol diacetate (EGDA), hexane, etc. are a few examples (Chitra et al., 2022; Li et al., 2022; Prasad et al., 2022; Abdo et al., 2023; Li et al., 2023; Min et al., 2023; Trivedi et al., 2023c; Trivedi et al., 2024). However, delamination with organic solvents necessitated a closed system to prevent the vaporization of organic solvents.

In our prior study, we examined the swelling behavior of encapsulant EVA through an analysis of both weight and size ratios. The significance of the size ratio becomes pronounced when considering the delamination process in PVMs, as a higher size ratio facilitates the effective separation of EoL PVM layers(Trivedi et al., 2023c). Higher temperatures will cause organic solvents to vaporize more because they are volatile and have low boiling points. It is observed that organic solvents degrade at higher temperatures as well (Rice & Herzfeld, 1934; Kim & Lee, 2012). These are the reason the temperature range of 25 to 55 °C was chosen for the study. Organic solvents are contaminated during interaction by the encapsulant EVA. This contamination reduces their ability to be recycled, and waste is produced when organic solvents are used for delamination. The recyclability and reusability of solvents used for recycling is a major concern (Quah et al., 2023).

This study conducts a systematic investigation of Si PVM delamination using organic solvents, as well as contamination of organic solvents by encapsulant EVA after the interaction. The objective of ongoing work is to study the recyclability of organic solvents, specifically concerning their contamination when used for delamination of EoL silicon photovoltaic modules (Si PVMs). The single-time use of organic solvent for delamination may be seen as an expensive investment, therefore the recyclability of the organic is necessary. It requires that the solvent used for recycling EoL PVMs must be both recyclable and have minimal contamination in order to be reused effectively and produce the least amount of waste. Organic solvents are compared based on the least amount of contamination, analyzed by Fourier transform infrared (FTIR) data.

THEORETICAL BACKGROUND

The heart of the silicon PVM is the solar cells, which are typically made of crystalline silicon, Fig. 1. Ethylene-vinyl acetate (EVA) is used to encapsulate the solar cells and provide mechanical support to the module. This layer also helps in bonding the various components together and protecting the cells from moisture and other environmental conditions. A Si PVM protected by tempered glass from the front side. It protects the solar cells from environmental factors such as rain, snow, and hail while allowing sunlight to pass through. The glass is designed to be transparent to allow maximum light penetration. The backsheet is made of layers of polyvinyl fluoride (PVF)/ polyethylene terephthalate (PET)/ polyvinyl fluoride (PVF). The backsheet protects PVM from environmental conditions and provides mechanical support from the backside. The whole assembly is encompassed by an aluminium frame. This frame provides structural support to the solar module and helps protect it from mechanical stresses, such as wind and snow loads (Mahmood & TamizhMani, 2023; Trivedi et al., 2023b).

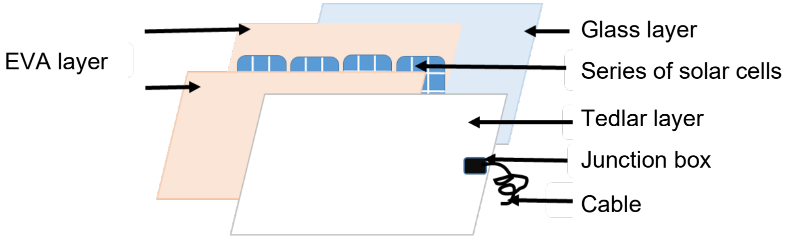

Figure 1. Schematic diagram of silicon photovoltaic mõdule.

EVA is almost transparent to sunlight, allowing the solar cells to receive maximum sunlight for energy conversion (Sharma et al., 2020). EVA adheres strongly to a variety of materials, including glass, metals, and solar cell surfaces. This property is critical to maintaining the structural integrity of the PVM (Tan et al., 2022). EVA has excellent resistance to ultraviolet (UV) radiation, ensuring long-term stability and preventing yellowing or degradation when exposed to sunlight. Extended exposure to ultraviolet (UV) radiation and elevated temperatures leads to photo and thermal oxidation and causes chemical degradation of encapsulant EVA of EoL PVMs (Sharma et al., 2020). EVA is electrically insulating, preventing electrical contact between the front and back surfaces of solar cells and reducing the risk of electrical shorts. It will also provide weather resistance, protecting the solar cells from environmental factors such as moisture, humidity, and temperature variations.

To analyze the contamination of organic solvents by encapsulating EVA, it is necessary to discuss the chemical structure of EVA and organic solvents.

Ethylene-vinyl acetate (EVA) is a copolymer, meaning it is composed of two different monomers: ethylene and vinyl acetate. The composition of EVA can vary, and the properties of the material are influenced by the ratio of these two monomers (Gu & Zhang, 2022). Typically, the vinyl acetate content in EVA used for solar encapsulation ranges from about 28% to 33%. Ethylene (C_2H_4) provides flexibility and low-temperature properties to EVA. This flexibility is important during the lamination process and helps the encapsulant conform to the irregularities on the surface of solar cells and other module components. Vinyl Acetate ($C_4H_6O_2$) contributes to the adhesion, transparency, and thermal properties of EVA.

The presence of functional groups in organic solvents influences their interaction with encapsulant EVA. Two different types of organic solvents were examined in this study. Chlorinated hydrocarbons, such as dichloromethane (CH_2Cl_2), chloroform ($CHCl_3$), and trichloroethylene (C_2HCl_3), are present in the first type of solvents, while the second type of solvents such as xylene and toluene having aromatic hydrocarbon ring. Chlorinated hydrocarbons are organic compounds that contain chlorine atoms bonded to carbon atoms. These solvents are known for their ability to interact with a variety of polymers, including EVA(Chitra et al., 2022; Prasad et al., 2022; Trivedi et al., 2023c). The interaction of EVA in chlorinated hydrocarbons can be attributed to the polar nature of the chlorinated solvents. The presence of chlorine atoms introduces polarity, allowing them to interact with the polar vinyl acetate units in the EVA copolymer. As the VA content in EVA increases, the overall polarity of the copolymer also increases. This enhanced polarity facilitates better interaction with chlorinated solvents, leading to improved interaction (Sharma et al., 2020; Trivedi et al., 2023c, 2024). In some cases, interaction may be influenced by temperature. Elevated temperatures can increase the kinetic energy of polymer molecules, making them more prone to interact with and contaminate the organic solvent.

Aromatic hydrocarbons, such as toluene ($C_6H_5CH_3$) and xylene ($C_6H_4(CH_3)_2$), are another category of solvents known for their effectiveness in interaction EVA with higher VA content (Xu et al., 2021; Li et al., 2022; Trivedi et al., 2023c). Aromatic hydrocarbons have a distinct cyclic structure with alternating single and double bonds. While they are not as polar as chlorinated solvents, they still exhibit some degree of polarity due to the presence of π-electron clouds in the aromatic rings. Also, it is investigated that after degradation of encapsulant EVA of EoL PVM, it produces some unsaturated hydrocarbon (double bond) which can attract unsaturated aromatic hydrocarbon due to like interact like principle(Giurginca et al., 2003; Sharma et al., 2020).

To understand the applicability, the experiments were conducted with different organic solvents at various temperatures for delamination of end-of-life PVM. This study represents the capacity of the organic solvents to retain the pristine chemical composition after the interaction with encapsulant EVA.

MATERIALS AND METHODS

Encapsulant ethylene vinyl acetate (EVA) was recovered from an end-of-life (EoL) photovoltaic module (PVM) made by the company, Emmvee Crystal, procured from the Indian Institute of Technology Kanpur, Uttar Pradesh, India. The PerkinElmer spectrum (V.10.03.06) instrument was employed for FTIR spectra of organic solvents and EVA. Rankem and Fisher scientific brands of organic solvents were used in the study. A total of five organic solvents were selected for study in order to understand contamination by encapsulant EVA, containing chlorine group (dichloromethane, trichloroethylene, and chloroform) and aromatic ring (toluene and xylene). These solvents exhibit good interaction with encapsulant EVA and can delaminate EoL PVMs (Trivedi et al., 2023c).

Dismantling of end of life photovoltaic module

The process of physically dismantling the EoL PVM assisted in the recovery of EVA. A saw was used to cut off the aluminium frame of EoL PVM. After removing the aluminium frame, a piece of EoL PVM was cut using a saw, Fig. 2. It contained a single 15.6×15.6 cm² solar cell. Using tweezers, broken glass pieces from the EoL PVM were removed. The encapsulant EVA layer was separated manually from the surface of the solar cell. For sample preparation, the undamaged EVA layer was used. Each EVA sample weighed 0.025 ± .002 g and measured approximately 0.49 cm² used for interaction with organic solvents.

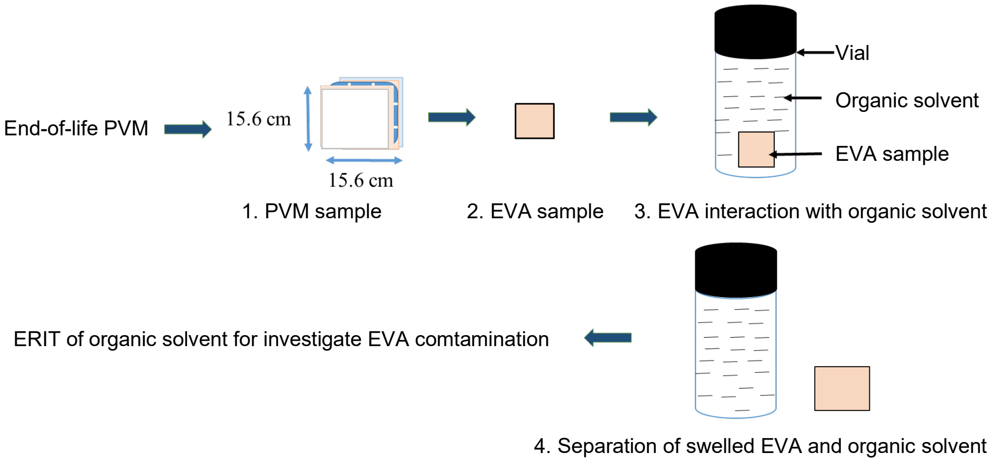

Figure 2. Schematic of interaction of organic solvent with encapsulant ethylene vinyl acetate (EVA).

Encapsulant EVA was recovered from the EoL PVM to analyze the actual state at end-of-life and its interaction with organic solvents. Subsequently, the EVA samples were placed in glass vials with various types of organic solvents for 24 hours duration. Each vial contained 5mL of organic solvent. After 24-hour interaction, the EVA samples were removed from the organic solvents, Fig. 2. To investigate the resistance of contamination of the organic solvents after interaction with encapsulant EVA, Fourier Transform Infrared (FTIR) spectra of the solvents were examined.

The reusability of organic solvents is discussed on the basis of single-time interaction with encapsulant EVA.

RESULTS

The swelling of the encapsulant EVA as a result of interaction with an organic solvent leads to delamination of the EoL photovoltaic module (McLoughlin et al., 2023; Vaněk et al., 2023). However, organic solvents become contaminated as a result of the encapsulant EVA's interaction. FTIR spectra are used to analyze organic solvents contamination after interaction with the encapsulating EVA at various temperatures. The FTIR spectra of all five organic solvents at temperatures 25, 35, 45, and 55 °C and the chemical structure of EVA and organic solvents used for the study is mentioned in Fig. 3.

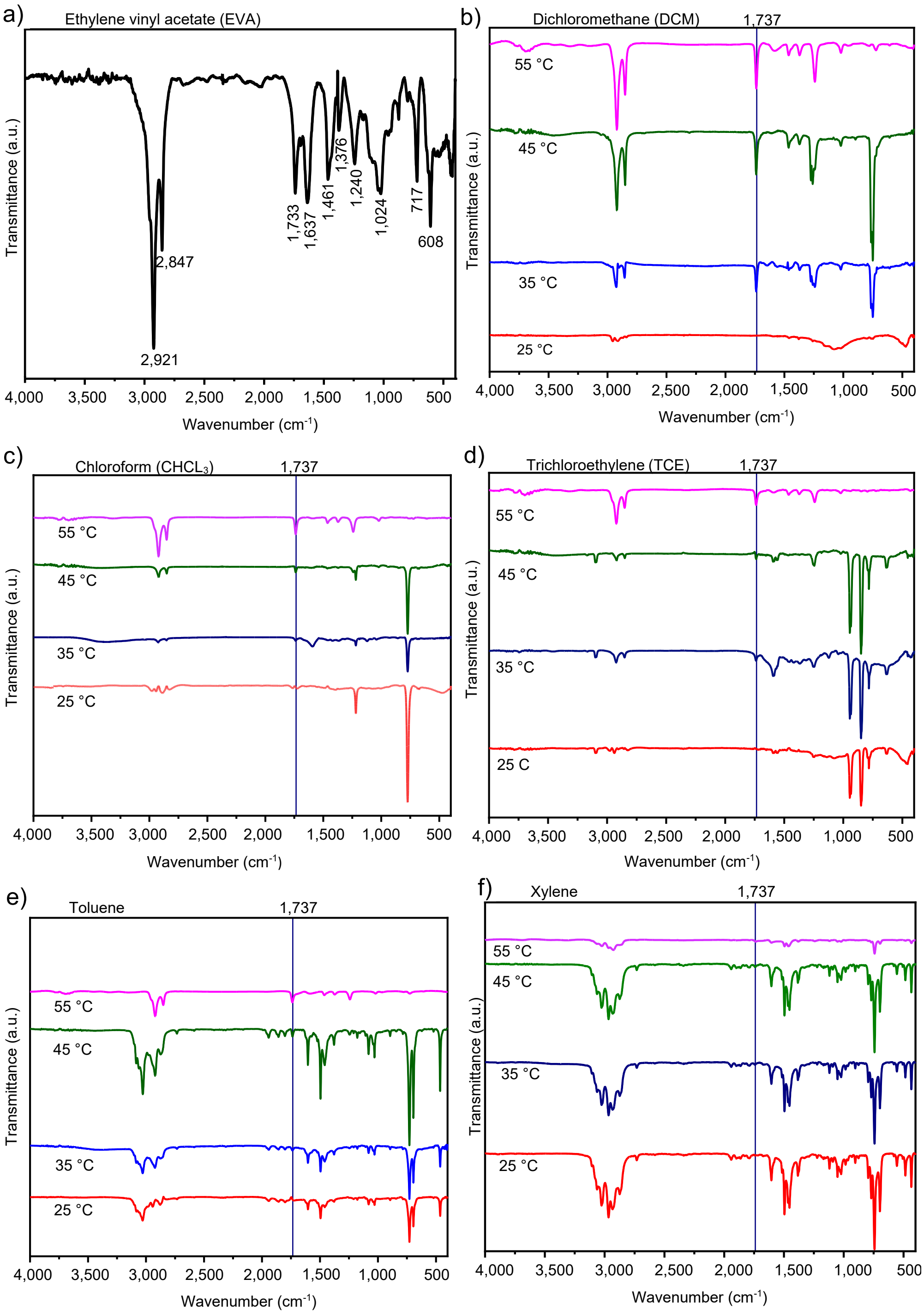

Figure 3. FTIR spectra a) Encapsulant ethylene vinyl acetate(EVA), b) Dichloromethane (DCM), c) Chloroform ($CHCl_3$), d) Trichloroethylene (TCE), e) Toluene, and f) Xylene.

Table 1 shows the FTIR peaks obtained from the encapsulant EVA of EoL PVMs. The peaks of various groups of EVA have been matched with the peaks identified in the literature.

Table 1. FTIR spectra peaks of encapsulant ethylene vinyl acetate (EVA) of end-of-life photovoltaic modules (EoL PVM)

	FTIR peaks of encapsulant EVA (cm^{-1})			
	Current study	Reference 1 (Khodkar & Ebrahimi, 2011)	Reference 2 (Jiang et al., 2017)	Reference 3 (Adelnia et al., 2015)
Acetate group peaks	1,737	1,734	1,739	1,742
	1,637	-	-	-
	1,243	1,234	1,242	1,234
	1,028	1,018	1,020	1,025
	608	607	-	-
Ethylene group peaks	2,926	2,915	2,919	2,916
	2,846	2,846	2,852	2,850
	1,466	1,460	1,467	1,466
	1,376	1,367	1,371	1,371
	721	720	721	-

As per Table 1, the FTIR peaks at 1,737, 1,243, 1,028, and 608 cm^{-1} are indicative of the acetate group, while the peaks at 2,926, 2,846, 1,466, 1,376, and 721 cm^{-1} indicative of the ethylene group in the EVA encapsulant. However, the peak at 1,637 cm^{-1} is absent in references. This particular peak (1,637 cm^{-1}) represents the C=C vibration of the vinyl group. The degradation of the encapsulant EVA is the main cause of this peak. When EVA is exposed to high temperatures and γ-irradiation, it degrades and produces unsaturated hydrocarbon, Eq. 1 (Giurginca et al., 2003).

$$\text{-CH}_2\text{-CH}_2\text{-CH(OOCCH}_3\text{)-CH}_2\text{-} \longrightarrow \text{-CH=CH-CO-CH}_2\text{-} + \text{CH}_3\text{CHO} \qquad (1)$$

A comparison between the FTIR spectra of organic solvents at a temperature range of 25–55 °C after interaction with encapsulant EVA is discussed in the upcoming section. The characteristic peak of EVA after a 24-hour interaction was analyzed to determine the levels of contamination in organic solvents.

DISCUSSION

To assess encapsulant EVA contamination, its characteristic FTIR peak should be evident in organic solvents after 24 hours of interaction. The ethylene group peaks at 2,926, 2,846, 1,466, 1,376, and 721 cm^{-1}, attributed to C-H vibration, may cause confusion due to the presence of C-H groups in organic solvents. The fingerprint region in Fourier-Transform Infrared (FTIR) spectroscopy, typically spanning from 1,500 cm^{-1} to 500 cm^{-1}, exhibits intricate and distinct peak patterns arising from the bending vibrations of various functional groups. However, the dense and complex peaks in this region contribute to confusion, especially regarding the FTIR peaks of the acetate group at 1,243, 1,028, and 608 cm^{-1}. Notably, the C=O peak at 1,737 cm^{-1} stands out as a signature of EVA contamination, as this functional group is absent in all solvents considered in the ongoing study.

The contamination of various organic solvents can be investigated by observing a 1,737 cm^{-1} peak in FTIR spectra of organic solvent after EVA interaction of 24 hours. It can be concluded that a peak appears at 1,737 cm^{-1} in dichloromethane (CH_2Cl_2), chloroform ($CHCl_3$) and trichloroethylene (C_2HCl_3) and toluene ($C_6H_5CH_3$). The 1,737 cm^{-1} peak's intensity increased as temperature increased (from 25 to 55 °C), indicating that contamination increased as temperature increased. However, xylene ($C_6H_4(CH_3)_2$)behaved differently compared to other solvents. FTIR peak 1,737 cm^{-1} is not observed in xylene despite temperature increases up to 55 °C. Although both xylene and toluene have aromatic rings, toluene is more reactive than xylene and can cause contamination, whereas xylene can only cause swelling in the encapsulant EVA and resist contamination (Azeumo et al., 2019; Trivedi et al., 2023c). By being resistant to contamination following contact with the encapsulant EVA, xylene can be made more recyclable. Because of its higher recyclability, xylene is a suitable option for the delamination of EoL PVMs for recycling.

To efficiently recycle the electronic waste generated by EoL PVMs in the coming year, the recycling process must be scaled up. However, the use of solvents on a large scale must comply with health, safety, and environmental regulations. Xylene was found to be the best-performing out of the five solvents, on the basis of resistance to encapsulant EVA contamination. Conversely, Chloroform ($CHCl_3$) and trichloroethylene (C_2HCl_3) are not suitable for extensive use due to concerns related to health, safety, and the environment (Bruckner et al., 2023). Dichloromethane (CH_2Cl_2), with its low boiling point of 39.6 °C, is highly volatile (Mu et al., 2022). These three solvents ($CHCl_3$, $C2HCl_3$, and CH_2Cl_2) are also contaminated by the encapsulant EVA and are unsuitable for recycling for the delamination of end-of-life PVMs. Sanofi's Solvent Selection Guide suggests that xylene and toluene both solvents have similar health, safety, and environmental hazard criteria. However, the International Council for Harmonisation of Technical Requirements for Pharmaceuticals for Human Use (ICH) limit for xylene is 2,170 ppm, compared to toluene's 890 ppm as per Sanofi's Solvent Selection Guide (Prat et al., 2013).

According to the literature, toluene is an organic solvent used for the delamination of end-of-life photovoltaic modules(Azeumo et al., 2019; Xu et al., 2021; Sah et al., 2023; Vaněk et al., 2023). However, the present study suggests that xylene is a better alternative in terms of recyclability and ICH limit. The interaction studied at various temperatures allows for the use of higher temperatures (55 °C), reducing delamination time and increasing the effectiveness of the recycling process. Xylene's resistance to encapsulant EVA contamination and its superior alignment with Sanofi's solvent selection parameters render it the preferred choice for the recycling of end-of-life photovoltaic modules (EoL PVMs) (Prat et al., 2013). The presence of encapsulant EVA in organic solvents suggests different (unclear) mechanisms of interaction. Xylene was found free from EVA contamination as per the FTIR spectra, at various temperatures of the experiment.

CONCLUSIONS

This study compares the recyclability of organic solvents, specifically focusing on their potential contamination when used for the delamination of EoL PVMs. Delamination unlocks a plethora of valuable materials for recovery. The encapsulant EVA interacts with the chloride group and the aromatic ring containing organic solvents and causes contamination. Fourier Transform Infrared (FTIR) spectra of the organic solvents explore potential contamination after a 24-hour interaction with EVA. The EVA contamination in organic solvents increases with temperature. It is explained by the interaction of EVA with organic solvents at a temperature range of 25 to 55 °C. The presence of the C=O peak at 1,737 cm^{-1}, is considered a signature of EVA contamination. This peak is noted in all solvents except xylene. Xylene exhibits resistance to encapsulant EVA contamination, making it a preferred choice for use in the delamination of end-of-life photovoltaic modules (EoL PVMs). Ensuring the recyclability of organic solvents is crucial for their prolonged reuse and minimizing waste production in the delamination process of end-of-life photovoltaic modules (EoL PVMs) recycling. The environmental burden can be reduced with the reuse of organic solvents for delamination.

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