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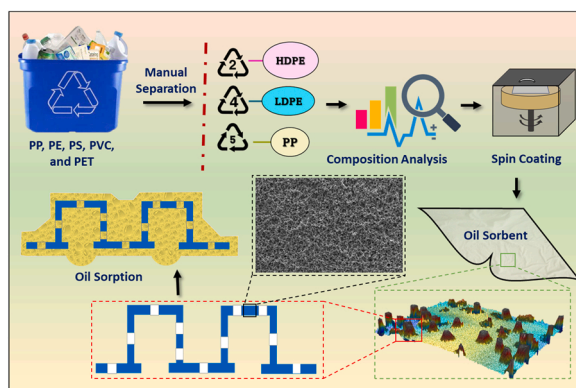
Up-cycling plastic waste into swellable super-sorbents

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HIGHLIGHTS

- Polyolefins, which make up 60% of plastic waste, were used to produce a super-sorbent for oil and organic removal from water.
- The super-sorbent has pores and cavities, with an oil uptake capacity of 70–140 g/g.
- Each cavity can swell up to twenty times the thickness of the sorbent, exhibiting sponge-like behavior.
- The sorbent can be squeezed mechanically or manually to recover the sorbed oil, with at least 97% efficiency.
- This technique can lead to greater utilization of plastic waste as a source of value-added materials.

GRAPHICAL ABSTRACT



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ABSTRACT

Environmental pollution caused by plastic waste and oil spills has emerged as a major concern in recent years. Consequently, there has been a growing interest in exploring innovative solutions to address these challenges. Herein, we report a method to upcycle polyolefins-based plastic waste by converting it into a bimodal super-oleophilic sorbent using dissolution, spin-coating, and annealing techniques. The resulting sorbent possesses an extensive network of pores and cavities with a size range from 0.5 to 5 μm and 150–200 μm , respectively, with an average of 600 cavities per cm^2 . Each cavity can swell up to twenty times the thickness of the sorbent, exhibiting sponge-like behavior. The sorbent had an oil uptake capacity of 70–140 g/g, depending on the type of sorbate and dripping time. Moreover, the sorbent can be mechanically or manually squeezed to recover the sorbed oil. Our integrated methodology provides a promising approach to upcycling plastic waste as an abundant source of value-added materials.

1. Introduction

Plastic pollution is a growing issue with over 380 million tons of

plastic produced annually, and less than 20% of it being recycled. Unfortunately, the recycling efforts focus on producing low-value items or energy recovery, making it challenging to address plastic pollution. To

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combat this issue, upcycling strategies have been developed, which involve the use of plastic waste as a feedstock for synthesizing value-added products, being molecules, materials, or polymers [1–3]. While these technologies have been effective in reducing plastic pollution to some extent [4–6], they tend to use isolated plastic waste or single plastic streams. Since post-consumer plastic waste is mixed, it becomes difficult to separate and identify individual polymers [7–9]. This complexity makes separation methods expensive and time-consuming, limiting the recycling or upcycling of these materials [10–12]. Hence, upcycling processes that deconstruct mixed plastics into valuable products without sorting could bypass this limitation.

The problem is further exacerbated in the case of polyolefins, which are 60% of total plastic waste [13], [14]. They comprise polyethylene PE {high-density polyethylene (HDPE), low-density polyethylene (LDPE)}, and polypropylene (PP). They are commonly used in the production of low-cost and disposable items, such as bags, wrappers, diapers, sanitary pads, and sorbent mats. However, due to their similar densities, it can be challenging and energy-intensive to separate these two materials. To address this issue, there has been a growing trend of blending PE and PP with or without a compatibilizer to create a blended material or pellets that have improved processability, flowability, and compatibility. This blending approach offers a more efficient and cost-effective way to reuse and recycle these materials while also reducing waste. [15–21]. These blended pellets can be used as a feedstock for various applications, and one such application is the preparation of reusable oil sorbent.

Oil spills are a significant environmental problem, and researchers have developed various sorbents to address this issue, including both natural and synthetic materials. Natural sorbents comprise inorganics such as clay, talc, zeolites, calcium fly ash [22,23], and organics such as corncob, hull, cotton fibres, bagasse, etc. [24–26]. While they are cheaper and readily available, they have limited oil sorption capacities and high-water uptake [27]. Additionally, synthetic sorbents made up of graphene, carbon nanotubes, and polymers in the form of aerogels [28], thin films [29], pads [30,31], and nonwoven fibres [32] have been widely explored. Although aerogels offer high uptake values, their light weight makes handling difficult for the user. In contrast, thin films, pads, and nonwoven fibres can quickly absorb oil but have low uptake capacities and retention values. Oil sorbent membranes were prepared from porous and nonporous fluorinated polyamides to rapidly clean the oil spills with an oil sorption capacity of 109 g/g [33].

There have been efforts to synthesize oil sorbents using plastic waste. Waste surgical masks based on polypropylene decorated by hydrophobic metal-organic frameworks were prepared for oil spill cleanup with an oil adsorption capacity of 2.5 g/g [34]. Further, waste polyethylene terephthalate was utilized to synthesize nanofibrous membranes for oil removal that sorb 30 g/g of crude oil [35]. Additionally, isolated waste HDPE materials was used with an oil sorption capacity between 90 and 100 g/g [14,36].

In addition, commercial sorbent pads, such as 3 M-HP-255, 3 M-156, Chemtex-BP-9 W, and Alsorb, are often used as part of an oil spill kit. These pads are made from polypropylene and typically consist of a nonwoven fabric. However, their thickness, which is at least 5 mm, limits their efficiency in thin water-borne oil films as they tend to pick up undesirable water due to a portion of the material floating below the surface. This has resulted in a trade-off between thickness and oil uptake capacity. Sorbent films with higher thickness have slower uptake kinetics and cannot efficiently remove thin water-borne oil layers. In contrast, lower-thickness films have limited volume and lower uptake capacity. Although creating pores of varying sizes can enhance the internal surface area, the oil uptake capacity cannot be significantly increased without increasing the thickness of the sorbent films.

Our research group has developed a new approach to address the limitations of current commercial sorbent pads for oil spill cleanups. We created a microporous thin film with hundreds of swellable cavities on its surface, with each cavity capable of swelling to twenty times the thickness of the film. When the thin film sorbent comes in contact with

oil, the cavities swell to their maximum capacity of around 150 μm , significantly increasing the oil uptake capacity. Additionally, after oil sorption, the film can release up to 98% of the oil when squeezed, making it reusable and recyclable. Moreover, our films can be compressed for more efficient packaging and occupy more sheets within a given space. Our approach to utilizing plastic waste as a raw material for the creation of a bimodal sorbent for oil and organic removal represents a valuable addition to the field of plastic waste upcycling.

2. Experimental section

2.1. Materials and methods

A stream of plastic waste was collected, including milk bottles, food disposables, water pipes, polybags, and water bottles. An isomeric mixture of xylene was purchased from VWR chemicals and was used as a solvent without further purification. The glass substrate plates were cut to a size of 5 cm \times 5 cm and can be fixed on the spin coater's chuck. Spin coating was carried out on a Spin coater from Ossila. The chuck was customized and received from Ossila instruments. A hot air oven made of MINO/30/TDIG by Genlab Ltd was used for heating and annealing. Hot plate annealing was carried on Heidolph magnetic hotplate stirrer.

Friction/peel tester Lloyd instruments Ltd Bognor Regis, UK, was used for tensile strength and modulus calculations. SEM images were captured with FEI Quanta650FEG. X-ray diffraction (XRD) measurements were measured using PANalytical Empyrean multipurpose XRD by Malvern Panalytical, Malvern, UK. XPS was measured using the Thermo Fischer Escalab 250XI platform. A monochromated X-ray source (Al K α : 1486.6 eV) is used. FTIR was carried out using the PerkinElmer Frontier instrument. Profilometry imaging was carried out using a Leica DCM8 microscope. The optical contact angle was calculated using OCA 35, Dataphysics instruments GmbH – Filderstadt, Germany. Porosity was calculated using the wet porosity method, where the difference between the weight of the thin film with ethanol and the weight of the thin film was converted into the volume by dividing it by density. Thickness was measured with a micrometer and cross-referenced with Deflesko FS3 PosiTector 6000 using a ferrous metal base.

2.2. Separation of polyolefins from plastic waste and composition analysis

A stream of plastic waste was collected from the local landfill site. Identified plastic waste was sorted as per SPI classification. The unidentified plastic was shredded into small pieces and subjected to float-sink separation based on densities with respect to water. The plastic floating on the water's surface was collected and investigated further. It was needed to know the amount of polyethylene and polypropylene contents. Thus, DSC was studied to provide qualitative and quantitative results.

A detailed DSC study was performed using known compositions of polyolefins. Polyolefins comprised polyethylene and polypropylene. Six compositions were made using polyolefins. Each composition was made up of 1 g polyolefin.

2.3. General procedure to prepare DSC samples

1 gm of polyolefins was taken in a conical flask and dissolved in 10 ml of xylene at 130° C for 20 min. Polyethylene comprises equal amounts of LDPE and HDPE. The reaction was stirred using a magnetic stirrer till a clear homogenous solution was achieved. The solution was then cooled to room temperature, resulting in the precipitation of the polymer. Polymer powder was extracted by separating and removing the solvent through filtration and vacuum condensation. Up to 98% of the solvent was recollected and reused. The resultant powder was sent for a DSC study.

The above general procedure was followed for the following compositions with respective weights.

- a) *Preparation of PE75PP25*: 0.75 gm of PE and 0.25 gm of PP were used.
- b) *Preparation of PE50PP50*: 0.5 gm of PE and 0.5 gm of PP were used.
- c) *Preparation of PE25PP75*: 0.25 gm of PE and 0.75 gm of PP were used.
- d) *Preparation of PE66PP33*: 0.66 gm of PE and 0.33 gm of PP were used.
- e) *Preparation of PE(LD)100*: 1 gm of PE(LD) was used
- f) *Preparation of PE(HD)100*: 1 gm of PE(HD) was used.
- g) *Preparation of PP100*: 1 gm of PP was used.
- h) *Preparation of unknown sample (PEXPY)*: 1 gm of the unidentified polyolefin composition was used.

2.4. DSC characterization

5 mg of powder was submitted for DSC analysis. The heating rate was kept at 10 °C/min till it reached 200 °C. The cooling rate was also held at 10 °C/min under a nitrogen atmosphere. The second cycle of heating was carried out under the same conditions. The enthalpy calculations and DSC graphs were collected from the second heating cycle for comparison. From the graphs, the melting peaks were assigned for respective polymers. Since the powder was more amorphous, the first cycle was not considered because broad peaks made it challenging to identify the melting points of polymers.

2.5. XRD characterization

Initially, XRD was carried out for pure samples to determine the characteristic peaks of individual polymers, followed by the investigation of known blends. These data revealed the presence or absence of characteristic peaks of respective polymers. It was qualitatively evident that polyethylene or polypropylene was present in the unknown sample.

2.6. Preparation of oil sorbent

1 gm of polymer (PE and/or PP) was taken in a conical flask and dissolved in 10 ml of xylene at 130 °C for 20 min, or till a clear solution was achieved. Then, 2 gm of sodium chloride as a filler with a particle size of 150–200 mm was added into the polymer solution and stirred for 10–15 min or till a homogenous dispersed solution was achieved. The hot solution was poured on a glass plate mounted on a spin coater chuck and allowed to spin. The spin coating was programmed in three steps to achieve a uniform thin film. (a) It includes the rotational speed of 400 rpm for 10 s, (b) The rotational speed of 1000 rpm was kept for 60 s, and (c) The rotational speed of 3000 rpm was maintained for 120 s

The excess polymer and solution were collected through a drain. The glass substrate with the thin film was detached from the chuck and was subjected to annealing at 160 °C in a hot air oven for 20 min. The thin film was then isolated from the glass substrate and washed with water to remove the filler. EDX analysis presented in Table 1 confirmed the removal of sodium chloride.

It is to note that the stirrer and spin coater were placed side-by-side to avoid solvent exposure to the environment, and the whole setup was kept inside the fume hood. The spin coater drain was connected to a receiver to collect solvent and excess polymer. After spin coating, a hot plate can also be used instead of heating in a hot air oven; however, the time for heating will be reduced to 5–15 s. Heating/ Annealing of the thin film is a critical step and requires a detailed study before finalizing the optimum time and temperature, as it leads to various porosity and strength.

Table 1
EDX Analysis of prepared samples.

Elements	Mass [%]	Atom [%]
Carbon (C)	98.20	98.76
Oxygen (O)	1.42	1.07
Sodium (Na)	0.19	0.10
Chloride (Cl)	0.20	0.07

In addition, the spin coater chuck was customized to accommodate glass substrate at high speeds. The glass substrate was heated to a temperature below the solvent's boiling point; thus, we used 120 °C, which is less than the boiling point of xylene. The hot polymer solution was poured onto the hot glass substrate attached to the chuck. The spin coating was programmed in three stages. In the first stage, the speed was kept low because it was easy to facilitate the binding of the polymer to the glass substrate, followed by medium speed; this step facilitates the spreading of the polymer to achieve uniform thickness. The speed was further increased in the last step to remove the solvent molecules from the thin film and refine the thickness.

After the spin coating process, the thin film was detached from the glass substrate and used in two ways; a) without further heating, which will be referred to as "Before-anneal", b) with heating at 160 °C for 20 min, which will be referred as "After-anneal". In the before-anneal substrate films, the films could not be separated from the substrate as a freestanding thin film because they did not possess sufficient strength. A freestanding thin film must maintain its integrity without a supporting layer or a substrate. For the after-anneal substrate films, the films can be easily separated from the substrate as a freestanding thin film, which has sufficient strength to hold its structure.

2.7. Oil sorption

Each measuring 25 cm², a series of sorbents were investigated for oil sorption studies. In the saturation studies, the sorbent was placed on the sorbate for 5, 10, 20, 30, and 60 s and was then taken out using a tweezer and allowed to hang for a specific duration so that the loosely connected sorbate was drained until the equilibrium was reached, the saturation capacity was then measured. In the dripping kinetic, the sorbent was placed in the sorbate until saturation was reached and then allowed to hang for different times, i.e., 0.5, 1, 2, 5, and 15 min. An oil uptake capacity is calculated as the mass of sorbate taken up by a unit mass of dry sorbent. Recyclability was performed to examine the sorbents' reusability. We performed recyclability in two ways, mechanical squeezing and solvent washing.

3. Results and discussion

3.1. Determination of PE and PP content in a composition of polyolefins through DSC

DSC is amongst the most straightforward techniques to identify the composition of various polymers in mixed plastic waste. Initially, pure polyolefins and known compositions of polyolefins were submitted for DSC to achieve reference data. Reference data was generated by calculating the area under the curves and the enthalpies for the pure and known compositions of polyolefins comprising different ratios of polyethylene and polypropylene.

The characteristic DSC peaks for PP, PE(HD), and PE(LD) were shown in Fig. 1(a). The melting peaks of PP, PE(HD), and PE(LD) were observed at 170 °C, 130 °C, and 110 °C, respectively. The DSC plots for the mixtures of PE and PP, including PE and PP in 25:75, 50:50, 66:33, and 75:25 ratios, were shown in Fig. 1(a).

We observed a decrease in intensity and shifting of the melting peak of PP with a reduction in PP content. Further, the melting peak of PP in PP-PE blends shifted closer to PE, which contributed to the homogeneous mixing. Similarly, the area under the curve and peak intensity of PE increased with an increase in PE content. However, PE(HD) showed a significant peak intensity and the area under the curve compared with PE(LD). We did not further deconvolute the PE peak region because the LDPE and the HDPE were miscible with each other, as we aimed to upcycle the plastic waste.

From these DSC graphs, the area under the curves of each polymer in composition was noted, and calculations were carried out for PP and the average of PE(HD) and PE(LD). This data helps in the quantification of

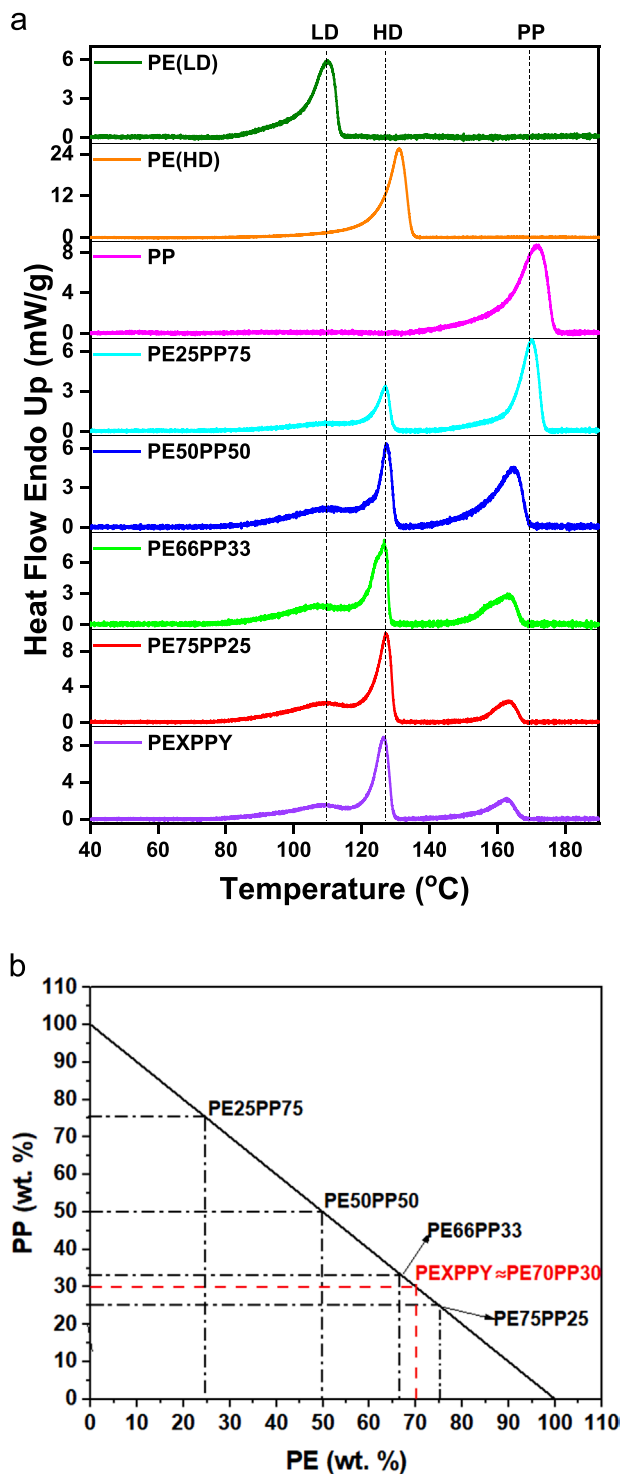


Fig. 1. (a).DSC plot of pure polyolefins including PE(LD), PE(HD), PP; and combinations of polyolefins. (b). Quantification of unknown composition of polyolefin (PEXPPY) obtained from DSC plot of pure polyolefins including PE (LD), PE(HD), PP; and combinations of polyolefins.

unknown compositions of polyolefins, as described in Fig. 1(b).

To determine the percentage of PP present in a sample, the area under the curve of PP was calculated and divided by the area under the curve of pure PP. The percentage of PE was then calculated by subtracting the PP percentage from 100. However, the individual components of PE(LD) and PE(HD) can be determined by deconvoluting the PE peak region and further calculations.

An unknown sample was taken and submitted for the DSC, where we observed different peaks showing the presence of polyethylene and polypropylene. We calculated the area under the curve of individual compositions of polyolefins to determine the amount of PP and PE as reported in the supplementary section (Table S1). Then it was overlaid on the reference graph to identify the percentage composition of the individual polymer. Qualitatively, an overview of the graph showed the unknown sample falls in the region between PE66PP33, and PE75PP25, which means the unknown sample contains PP in the ratio of 25 and 33, while PE made up the rest. Further, the PP and PE contents were found to be $30 \pm 2\%$ and $70 \pm 2\%$ from the area under the curve calculations. Thus, from the DSC analysis, the amount of PE and PP can be determined, which can be upcycled into value-added products without separating polyolefins.

3.2. Determination of PE and PP content in a composition of polyolefins through XRD, FTIR, and XPS

Further, XRD was carried out to support DSC results. We have investigated the XRD of pure polymers to explore their characteristic peaks. We have observed that the characteristic peaks of PE were at 22° and 24° , while for PP the characteristic peaks were at 13° and 17° . Similar to DSC, we observed a decrease in peak intensity with the reduction in the content of the polymer. However, we did not observe a change or shift in peak positions. For a given unknown sample, the presence of PP and PE characteristic peaks was observed, representing qualitative but not quantitative composition (see Fig. 2). Thus, XRD suggests the presence or absence of different polyolefins.

The molecular interactions and bonding between the polymer chains were investigated using FT-IR spectra, which assisted us in determining the qualitative composition of PE and PP. We observed characteristics of

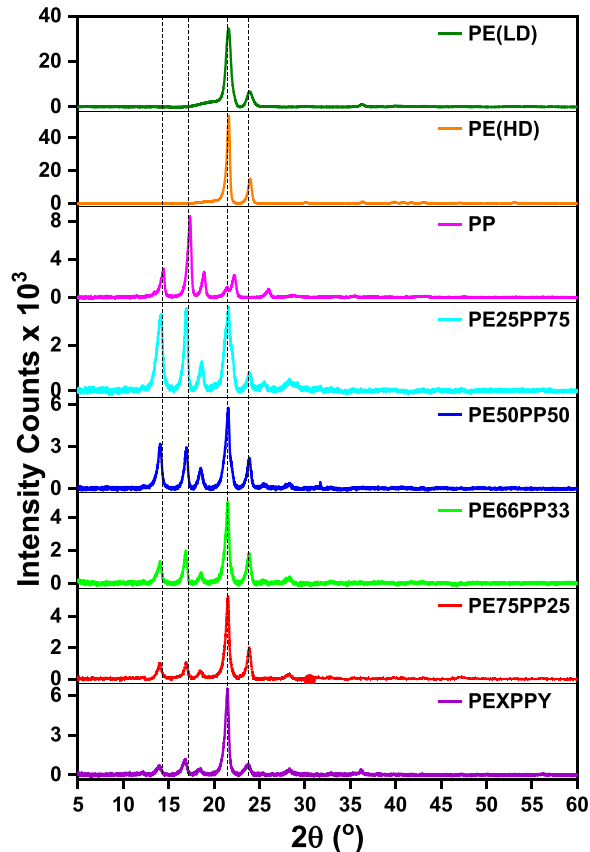


Fig. 2. XRD spectra of pure polyolefins and the different compositions of polyolefins.

C-H asymmetric and symmetric stretching peaks at 2951, 2919, 2869, and 2836 cm^{-1} and C-H symmetrical bending peaks at 1459 and 1376 cm^{-1} for PP. Similarly, pure LDPE and HDPE showed characteristic C-H asymmetric and symmetric stretching peaks at 2914 cm^{-1} and 2848 cm^{-1} , the C-H bending peaks at 1464 cm^{-1} , and the C-H rocking peaks at 725 cm^{-1} .

It was observed that in the varied compositions of PE and PP, the peaks at 2914 and 2848 cm^{-1} , as well as the C-H rocking peak at 725 cm^{-1} , were enhanced with an increase in PE content. On the other hand, with an increase in PP content, the C-H symmetric peak at 1376 was enhanced, which is the characteristic peak of the free methyl group. When any composition of PE and PP was present in a polymer sample, all the characteristic peaks of PE and PP were observed in their respective percentages, as shown in Fig. 3.

XPS was investigated to illustrate the polymer surface chemical composition and mainly to validate the absence of xylene as a residual solvent in the polymeric mixture (Fig. 4). The presence of any residual solvent in the polymeric mixture will be detected by observing the peak at 292 eV because of the $\pi-\pi^*$ carbon bonding in the aromatic ring. However, we did not notice any peak around the binding energy of 292 eV, which confirmed the complete evaporation of the solvent.

3.3. Morphology analysis of polyolefins and their combinations through SEM and profilometry

SEM was carried out to examine the surface morphology of the pure polyolefins and their combinations (see Fig. 5). Pure polyethylene (PE100) was seen as a flake-like structure, while pure polypropylene (PP100) was seen as a fibrous web-like network. When the polymers were mixed, we observed a uniform homogenous compatible morphology. When one part of polypropylene was mixed with another equal part of polyethylene (PE50PP50), then a hybrid structure of both

flakes and a web-like structure was represented here as a mesh-like structure. When one part of polyethylene was added to three parts of polypropylene (PE25PP75), we observed a hybrid-web-like structure, leaning towards the dominant polypropylene. Likewise, when one part of polypropylene was added to three parts of polyethylene (PE75PP25), we observed a hybrid-flake-like structure, leaning toward dominant polyethylene morphology. Moreover, we used the dissolution method to mix two different polymers, resulting in a homogenous polymer blend. If these polymers were not mixed well and incompatibility remained, we would have observed a combination of flakes and web-like structures.

Moreover, the annealing step during synthesis facilitated the healing of the internal fractures of the polymer at the nano-level. As shown in Fig. 5, a mesh-like structure was observed with some rough edges. The edges smoothened with further heating, and the polymer started aggregating as it melted, resulting in micropore formation. With additional heating beyond the melting point, the structure started to melt, and the micropores began to shrink.

Likewise, for oil sorption apart from macropores for oil diffusion, we have introduced swellable 3D cavities (Figs. 6 and 7), which further increased the uptake capacity and retention volume. Inorganic filler (sodium chloride in our case) was used to create swellable cavities within the thin film. The inorganic filler was dispersed in the polymer solution. The solvent that dissolved the polymer should not dissolve the inorganic filler. After the spin coating process, the thin film was heated to enhance its strength, and then the thin film was washed in water to remove the inorganic filler resulting in hollow swellable cavities. The size of the cavities is determined by the particle size of the inorganic filler (in our case, 150–200 μm).

The thin film sorbents showed cavities adjoined with macropores that facilitate the oil penetration and high oil retention volume. This increased oil retention capacity was due to the cohesive forces between oil molecules present inside the sorbent's structure and at the surface and the adhesive forces between the sorbent and oil.

Furthermore, the profilometry images were captured that reflected the 3D-image projection of the cavities (Fig. 7). It also provided surface roughness, a synergistic point for oil sorption and retention. The cavities played a significant role in enhancing oil retention and high uptake capacity. When the sorbent was squeezed mechanically or washed with the solvent, the oil from the cavities was collected, and the sorbent was reused for another cycle. On squeezing, the structure of cavities shrinks to the size of the thickness of the thin film, which upon reusing and resorption, retains its original swollen structure. Thus, these cavities act as swellable reservoirs. We checked the uptake capacities of thin films made from different polymer compositions to verify whether all compositions would be effective in oil sorption.

We observed the following key observations before and during the spin coating stage: (a) The polymer solution must be in solution form to facilitate spin coating, which is possible by mixing and keeping the solution at a high temperature. Also, it is not necessary to keep and maintain the spin coating chamber at high temperatures; (b) On spinning the polymer with inorganic filler, allowing the chamber at ambient temperature and the solution at high temperature, with a lapse in time, the temperature of the polymer solution decreases, and it entraps the inorganic filler from expulsion, resulting in the formation of cavities. Due to its higher density, the inorganic filler settles on the glass plate first, and then the polymer forms a coat over it, forming cavities and macropores. (see Fig. 6a-b); (c) The temperature of the glass substrate plays an essential role in the size of the openings of the cavities. If the temperature of the glass substrate is kept high but below the solvent's boiling temperature, the cavities' openings are wide (see Fig. 6c). If the glass substrate and solution are heated at high temperature, the transfer of heat is slow, the time taken for the polymer to cool down is more, and the polymer remains at high temperature for a relatively long time, which facilitates the inorganic filler to settle to the bottom quickly and allowing to have more surface contact area, resulting in wide openings. Contrarily, when the glass substrate was kept at room temperature, and

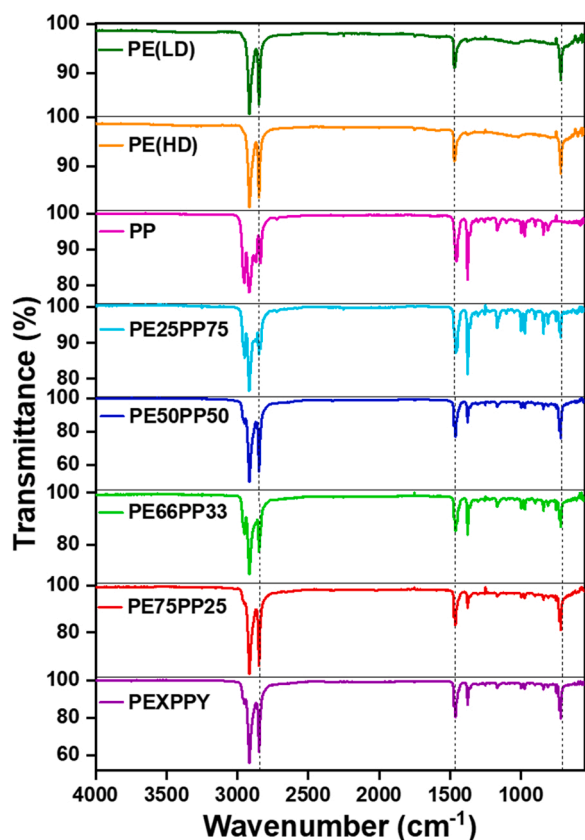


Fig. 3. FTIR spectra of pure polyolefins and the different compositions of polyolefins.

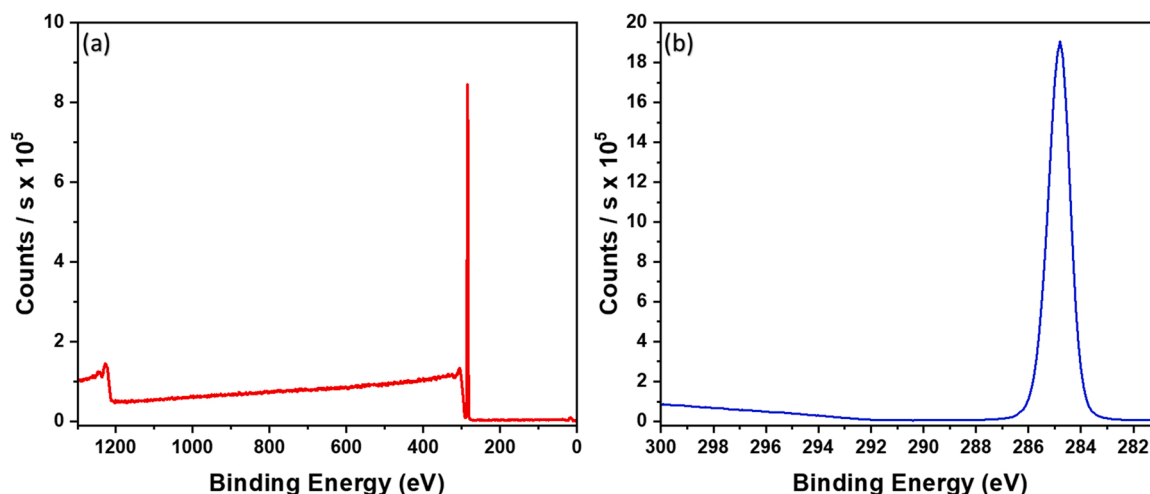


Fig. 4. (a) XPS Survey spectra (b) C1s spectra of PE50PP50 polyolefin composition.

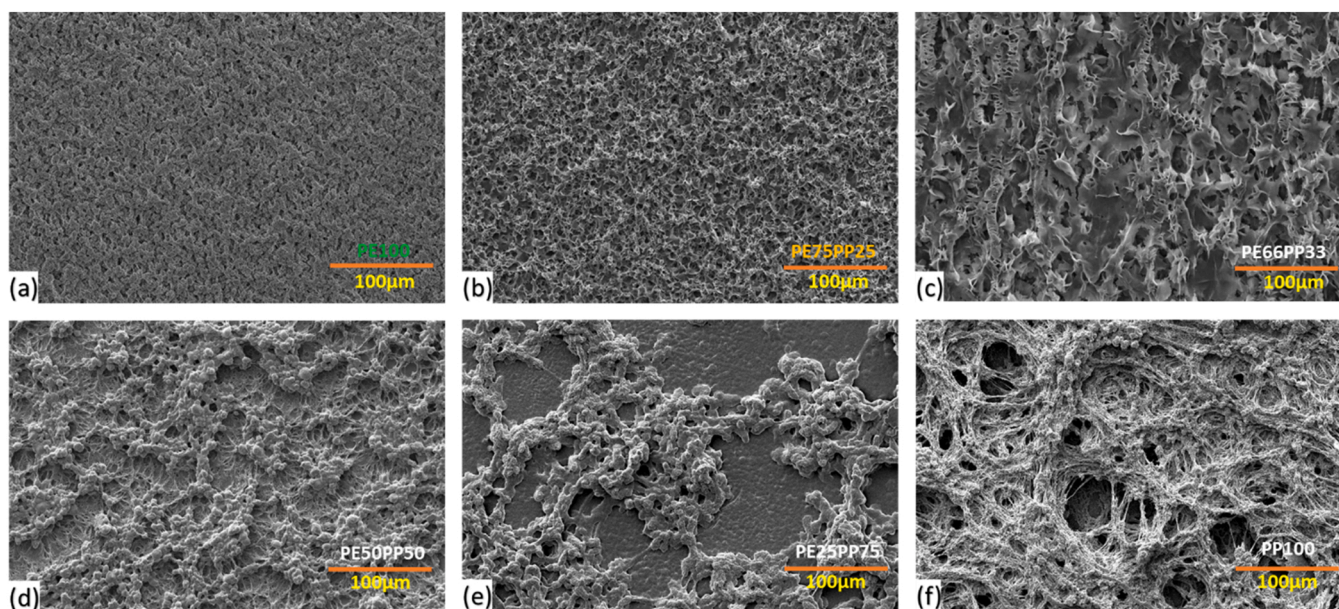


Fig. 5. SEM images of polyolefin compositions before annealing (a) PE100 (b) PE75PP25 (c) PE66PP33 (d) PE50PP50 (e) PE25PP75 (f) PP100.

the hot solution was poured on it, then a quick transfer of heat took place, allowing the polymer to precipitate swiftly by fast cooling, which does not aid the inorganic filler in settling to the bottom and allowing to have low surface contact area, resulting in small openings (see Fig. 6d).

Furthermore, the profilometry images were captured that reflected the 3D-image projection of the cavities (Fig. 7). It also provided surface roughness, a synergistic point for oil sorption and retention. The cavities played a significant role in enhancing oil retention and high uptake capacity. When the sorbent was squeezed mechanically or washed with the solvent, the oil from the cavities was collected, and the sorbent was reused for another cycle. On squeezing, the structure of cavities shrinks to the size of the thickness of the thin film, which upon reusing and resorption, retains its original swollen structure. Thus, these cavities act as swellable reservoirs. We checked the uptake capacities of thin films made from different polymer compositions to verify whether all compositions would be effective in oil sorption.

3.4. Tensile Strength and Modulus

To provide sufficient strength to the microporous thin film with

swellable cavities, annealing was performed. The freestanding thin films were difficult to synthesize without annealing as the structure collapsed because of weak strength. So, we refer to the thin films that are not annealed here as "before-anneal", and the thin films that are annealed are termed "after-anneal". In the before-anneal thin film, the polymer chains were amorphous, possessed weak intermolecular interactions, and were loosely connected. In the after-anneal thin film, polymer chains become soft upon heating, rearranging, and realigning the chains to a more compact and denser polymer with enhanced crystallinity. This realigning crosslinking of polymer results in increased mechanical strength because of increased intermolecular dispersion forces. Also, this heating facilitated the easy separation of the film from the substrate. The resultant thin film possesses sufficient strength to hold its structural integrity and can be used as a freestanding thin film.

In addition, the modulus and strength of the thin film are affected by the change in concentration of the inorganic filler, as shown in Fig. 8a-b. Initially, the effect of filler concentration on the strength of the sorbent was studied. As the filler concentration increases, cavities increase, thereby increasing the cavity count per area. Potentially, the uptake capacity and retention capacity will increase. On the other hand, when

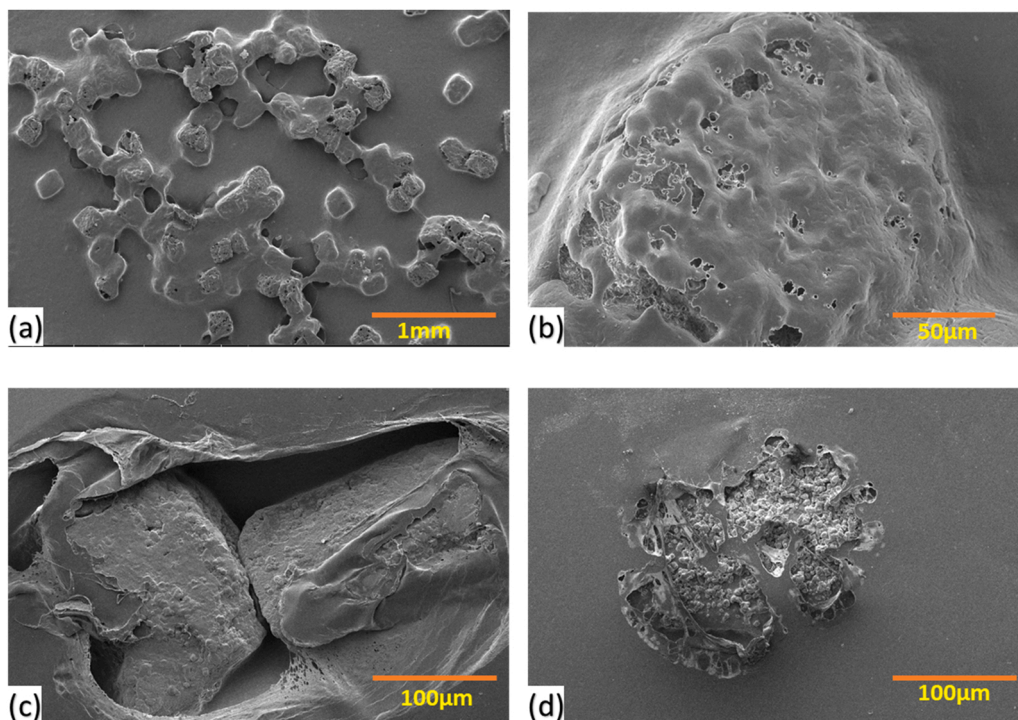


Fig. 6. SEM of PE50PP50 showing cavities (a) overview (top view), (b) close-view of a cavity showing macropores (top view), (c) wide opening (bottom view), and (d) narrow openings (bottom view).

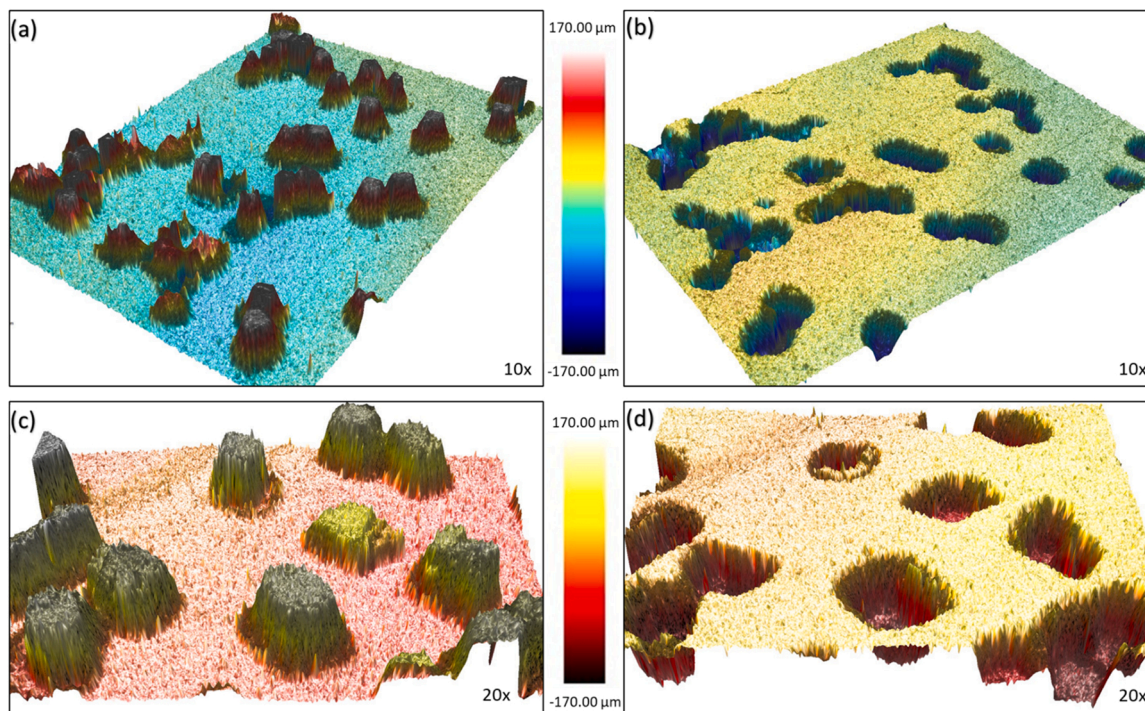


Fig. 7. Profilometry 3D-imaging of the thin film sorbent made from PE50PP50 polymer composition showing cavities (a) top view at 10x resolution (b) bottom view at 10x resolution (c) top view at 20x resolution (d) bottom view at 20x resolution.

the number of cavities per area increases, the breakpoints increase, increasing the tearing of the sorbent and leading to a decrease in its strength. This prone to tearing will reduce the recyclability. However, the cavities cause the sorbent film to break suddenly, whereas fewer cavities cause it to elongate; thus, an increase in the filler concentration increases the tensile modulus. Consequently, we have optimized the

filler concentration to polymer in a 2:1 (w/w%) ratio with an inorganic filler size of 150 – 200 μm (Fig. 8c). We also attained sufficient mechanical strength of ~ 10 MPa for all polymer compositions, with an average of 600 cavities per cm^2 . Thus, we can upcycle the polyolefins into one blend without further separation. The physical properties at varying parameters for PE50PP50 are shown in Table 2.

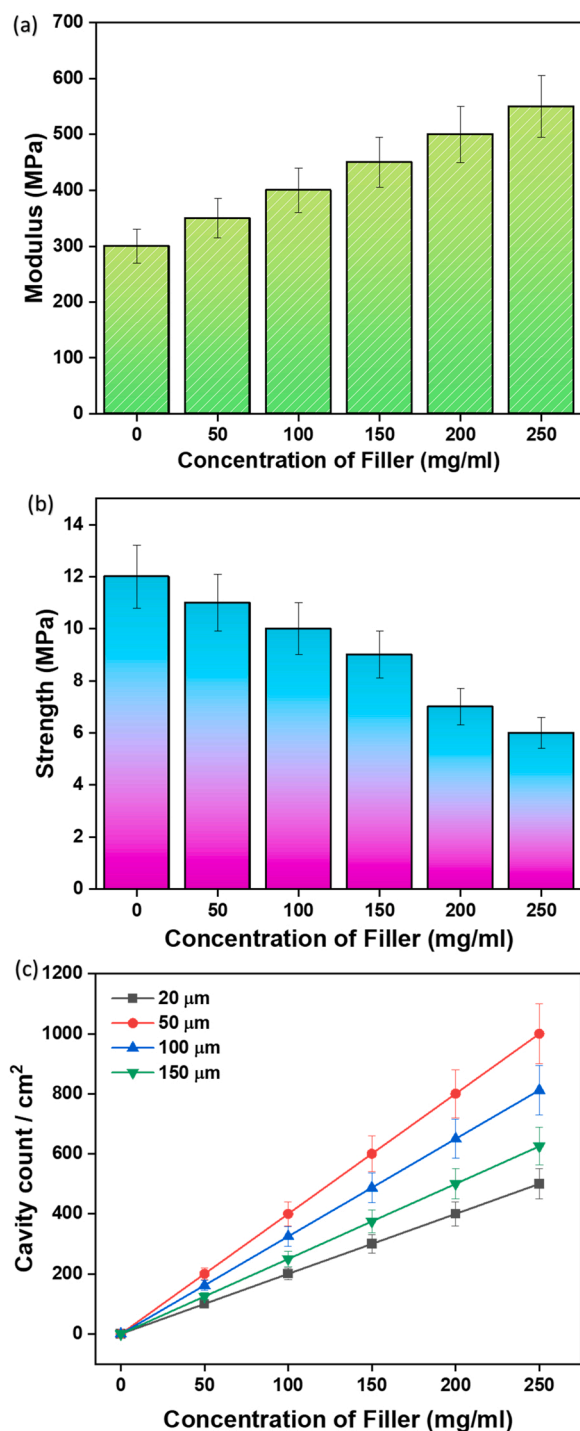


Fig. 8. The effect of concentration of filler on (a) modulus (b) strength, and (c) cavity count per area.

3.5. Oil sorption application

Fig. 9 explains the process of oil sorption. Thin film sorbents, which are easily pressable, have been created with cavities and macropores (part i & ii). These cavities swell when exposed to oil (part iii). In the next step, these thin films are squeezed, which releases absorbed oil (part iv). Nevertheless, some oil remains entrapped in macropores, which are later removed using an organic solvent. (part v).

The saturation kinetics of the oil sorbent was carried out to determine how quickly the sorbent reached its maximum uptake capacity. We have observed that the change in compositions of PE and PP did not alter

Table 2

Physical parameters of thin-film sorbent PE50PP50.

SN	Porosity (%)	Thickness (μm)	Temp (°C)	Time (min)	Strength (MPa)
1	79	20	25	0	ND
2	74	18	155	5	ND
3	68	16	155	10	1
4	62	14	155	15	4
5	35	7	155	20	8
6	8	5	155	25	11
7	< 1	5	165	5	12

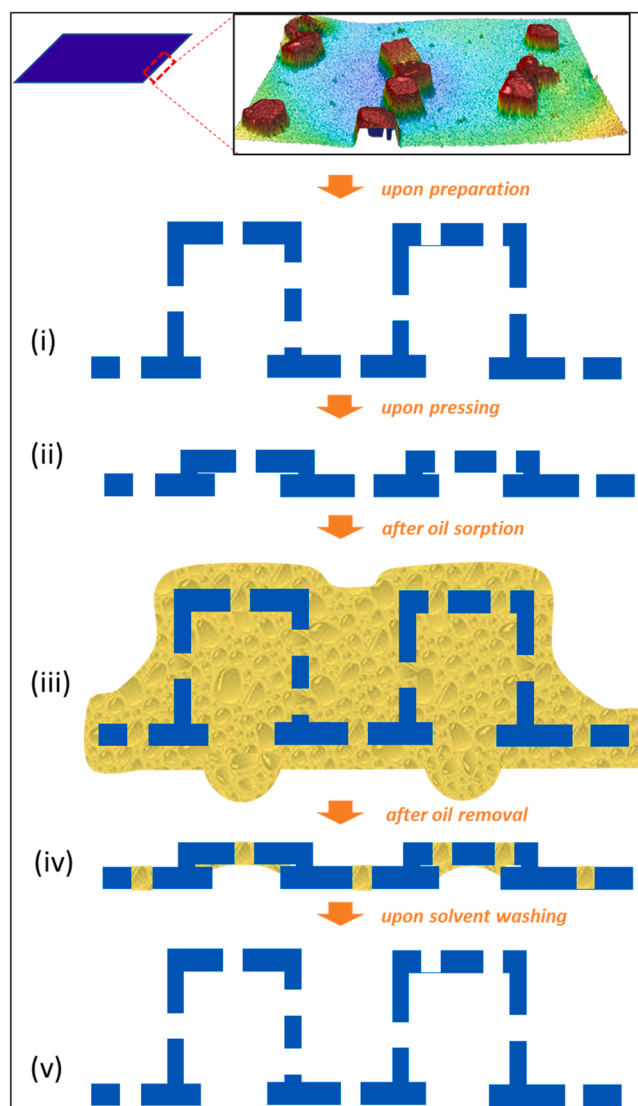


Fig. 9. Process of oil sorption.

the uptake capacity because both PE and PP are hydrophobic and oleophilic in nature. They are inert and do not decompose when dipped in oil; thus, they can be recycled and reused. Further, the PE-dominant thin films showed a relatively more uptake capacity because of the internal structure of the cavities. At a given constant polymer percentage, polypropylene is less viscous than polyethylene; thus, polypropylene forms a rounded structure around the inorganic filler, whereas polyethylene forms a cuboidal structure. Once the thin film sorbents were placed on the oil surface, the oil was immediately adsorbed onto the surface. With time, the oil penetrated the sorbent and the cavities, increasing the uptake capacity. The adhesive forces between the rough

surface of the cavities and the oil molecules increase the retention capacity, in addition to the cohesive forces between the oil molecules inside the cavities and on the surface, resulting in high uptake capacities of 91–148 g/g for engine oil. For different compositions, when the saturation uptake capacity was evaluated immediately, it was measured to be 142–148 g/g (see Fig. 10a). After reaching equilibrium, the uptake capacity for engine oil was estimated to be 91–95 g/g (see Fig. 10a). The equilibrium hanging time for the saturation kinetics was 5 min.

The dripping kinetic was also studied for the different sorbents made from different compositions. Similar to the saturation kinetics, we observed no significant change in the dripping kinetic capacities for different compositions of the polymers, as both the polyethylene and polypropylene were oleophilic and possessed uniform cavities, which helped in retaining high amounts of oil. As explained earlier, the oil in the cavities creates strong cohesive forces between the oil molecules inside the cavities and on the surface. The synergistic effect of the adhesive and cohesive forces resulted in high retention even after allowing the sorbent to drip for 15 min. (see Fig. 10b).

Further, to widen the usage of the sorbent to organic solvents and oils, we have investigated the uptake capacities of different polymer sorbents with diverse oils, including sunflower oil, paraffin oil, crude oil, and engine oil, along with organic solvents like toluene. Here too, we observed no substantial change in the uptake capacities using different polymer compositions. However, we observed a change in uptake volumes while using different oils because of the varying viscosities of the oils and their affinity towards the sorbents. Moreover, organic solvents like toluene showed good uptake capacity despite their lower viscosity, which is attributed to the oleophilic nature of the sorbents and the 3D-structural design of the cavities. This widens the scope of usage of these

sorbents to a variety of oils and organic solvents. The immediate and equilibrium uptake capacities are shown in Fig. 10c-d, respectively.

Furthermore, we investigated oil-water separation studies to understand the efficiency of the sorbents in real-time applications because oil spillage on the water surface makes it challenging to collect only oil by repelling water. This selective separation study showed high oil selectivity over water and can be used in oil sorption and separation from the water. These films are super oleophilic and hydrophobic, so they have an increased tendency towards oil compared to water. When they were placed on thin oil films placed on water, they absorbed only oil and repelled water. A 25 cm² thin film sorbent was placed in 100 ml water with differing oil percentages. With the increase in oil percentage in water, the sorbent uptake capacity was limited. A larger-sized sorbent can overcome this problem. However, the water retention was less than 0.5%, and the oil selectivity was over 99.5%. We separated oil from water with all the compositions of polymers, which opens the scope of upcycling the polyolefins as a single blend without further separation (see Fig. 11).

Additionally, the recyclability was carried out to examine the reusability of the sorbent. For recyclability, the sorbents should possess sufficient strength to be used multiple times without losing their uptake capacity and without collapsing the structure. For oil-sorbent thin films, we annealed them after spin coating. The annealing process helped in enhancing the mechanical strength of the thin film, thus making the sorbent a reusable oil sorbent, especially after the sorption and desorption of oil from the sorbent structure.

We have successfully reused the sorbent films ten times, which can be further reused. Sorbent films can be recycled in two ways: one by mechanical squeezing and the other by solvent washing. The mechanical

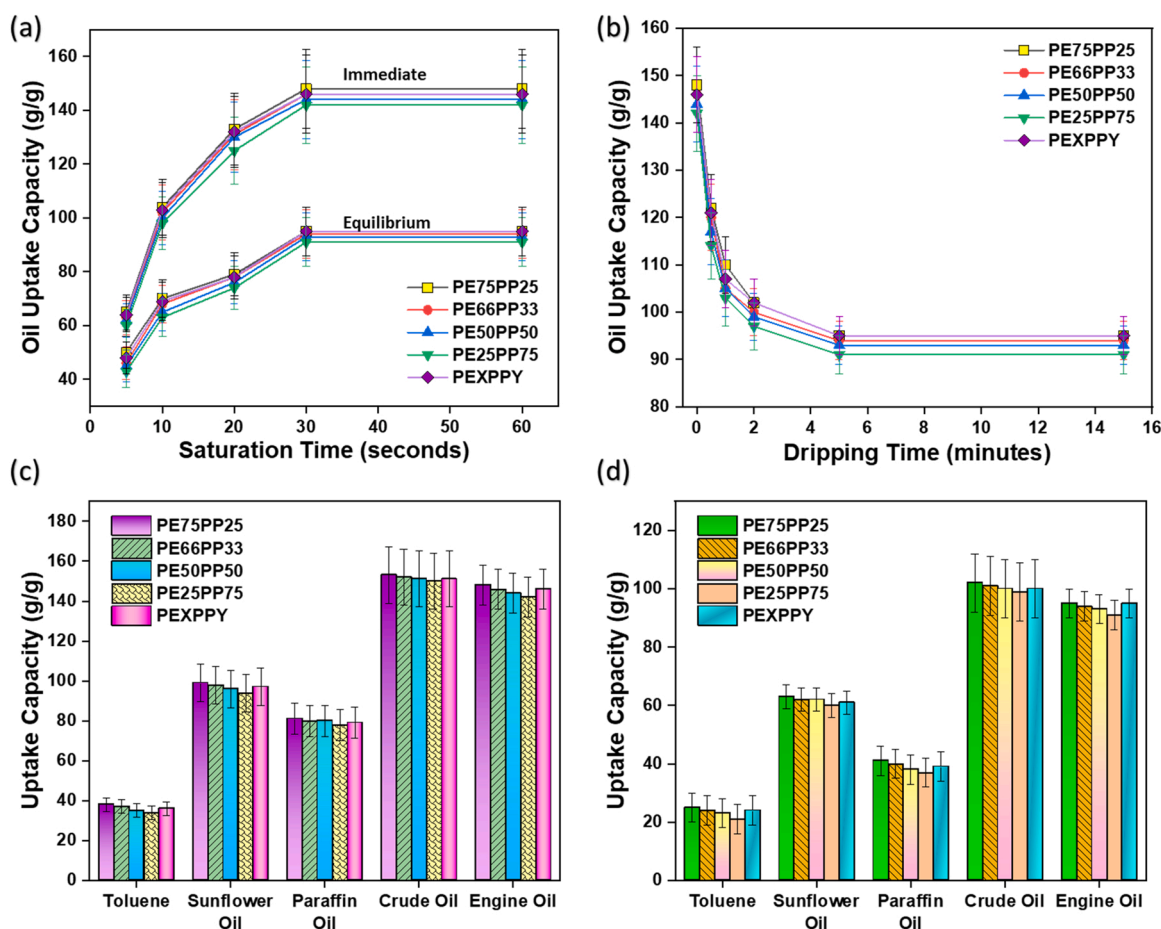


Fig. 10. (a) saturation kinetics showing immediate and equilibrium uptake capacities using engine oil (b) dripping kinetics to know immediate and equilibrium uptake capacities using engine oil, (c) immediate uptake capacity by comparison with different oils (d) equilibrium uptake capacity by comparison with different oils.

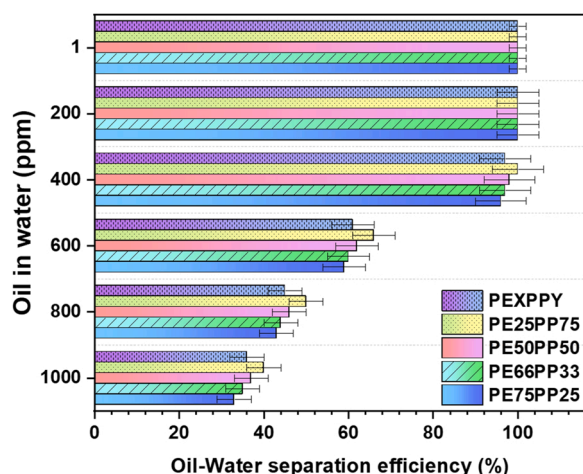


Fig. 11. Oil-Water (engine oil) separation efficiency using different polymer composite oil sorbents.

squeezing method is the simplest in reusing the sorbent; however, a small portion of oil is trapped inside the sorbent because the pressing will not be sufficient to remove oil from the pores. On reusing the sorbent, the thin film cavities absorb the oil to its maximum uptake capacity by retaining its structure. Thus, mechanical squeezing helps collect oil easily and quickly with an efficiency of 97% (see Fig. 12).

On the other hand, the solvent washing method is time-consuming but 100% efficient in oil recyclability. In this method, the oil was squeezed after oil sorption, and the sorbent was placed in hexane, which dissolved the oil and cleaned the sorbent thoroughly. Further, the sorbent film was effectively used to its maximum uptake capacity upon reusing. Then finally, the hexane solvent was evaporated in a rotary evaporator to separate the oil and hexane. Thus, achieving 100% efficiency with solvent washing method.

Moreover, the contact angle measurements suggest the sorbents be

hydrophobic or hydrophilic. We have measured contact angles for engine oil, paraffin oil, toluene, and water. We observed that the rate of oil sorption increases with time because of the oil penetration into the cavities. This fast penetration of oil inside the sorbent film depicts the effectiveness of the sorption mechanism. For instance, engine and paraffin oil showed initial contact angles of 14.9° and 11.0° , respectively; however, the contact angles reduced to 4.4° and 2.9° within 10 s (Fig. 13). Moreover, we could not measure the angle with toluene because as soon as it is dropped on the surface of the sorbent, it spreads through the sorbent without retaining any angle. It was due to the superoleophilic nature of the sorbent. Besides, the contact angle with water remains the same concerning time, showing the sorbent's hydrophobic nature.

3.6. Acid and base treatment studies

Moreover, we have treated the sorbent PE50PP50 in 1 N HCl and 1 N NaOH solution, to investigate any chemical changes in the structure or the composition of the sorbent. We placed the sorbents in the acid and basic solution overnight and submitted them for FTIR and XRD characterizations. As anticipated, we did not observe any significant change in the composition or structural change. The XRD and FTIR spectra of untreated, acid-treated and base-treated sorbents (PE50PP50) are shown in Fig. 14.

3.7. Comparison with commercial sorbents and reported Studies

In addition, the as-prepared sorbent (PEXPPY) was compared with the commercial sorbents, namely Corksorb, BP9W, and HP-255 (Fig. 15, Table 3). Prepared sorbent (PEXPPY) was developed with inherent macropores and swellable 3D cavities that have enhanced oil absorption capacity. The design and structure of the cavities are what contribute to the increased uptake capacity.

In both the immediate and equilibrium states, the saturation uptake capacity of produced sorbent (PEXPPY) was compared with that of commercial solvent. Immediate values are lower than equilibrium state

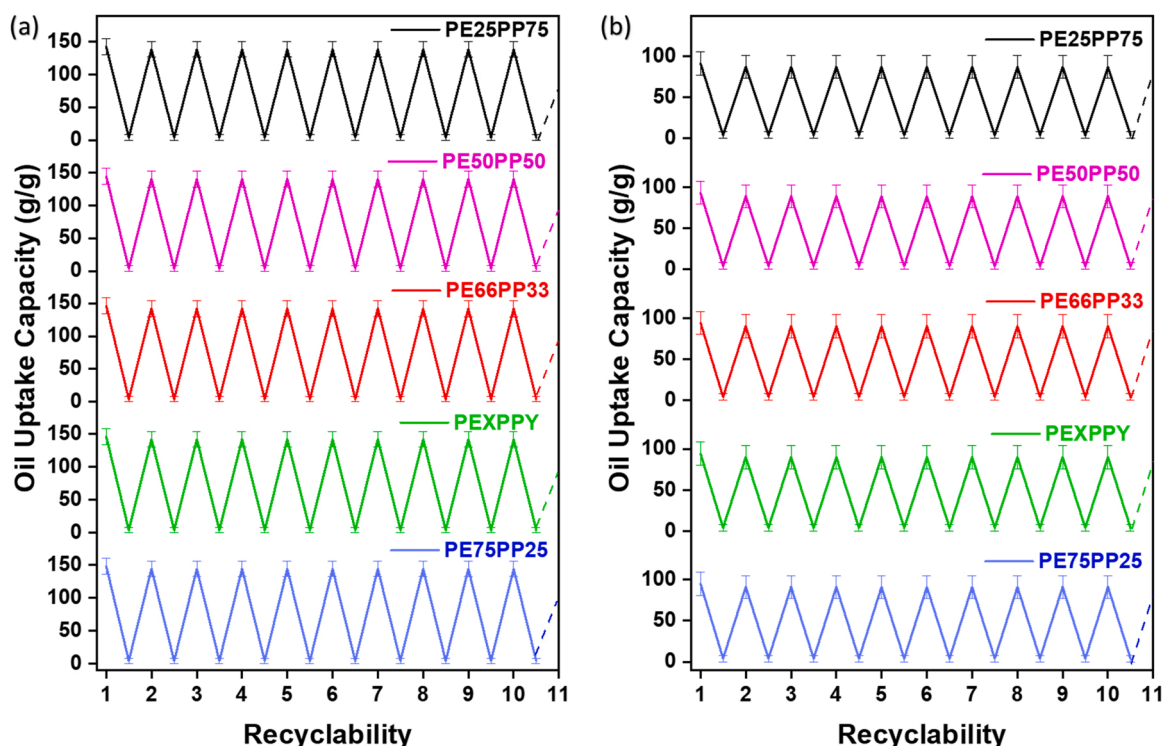


Fig. 12. Recyclability of different polymer composite sorbents using engine oil at (a) immediate uptake capacities and (b) equilibrium uptake capacities.

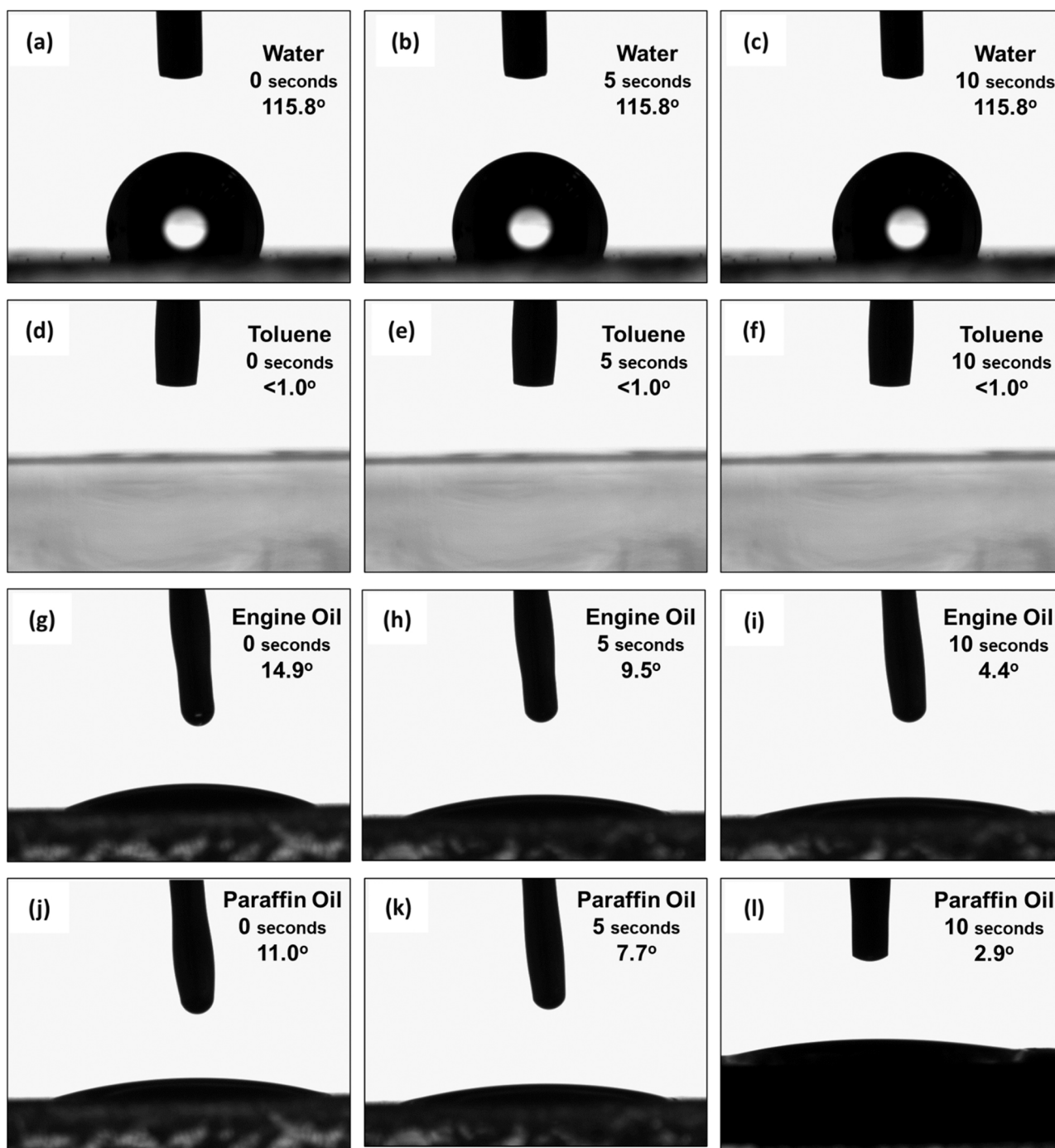


Fig. 13. Contact angle measurement at different intervals of PE50PP50 sorbent using (a)-(c) Water, (d)-(f) Toluene, (g)-(i) Engine Oil, and (j)-(l) Paraffin Oil.

values since surface oil that was not absorbed in macropores, and cavities is gradually removed from the surface. Commercially available sorbents could provide no simple method of recycling. During development, they didn't consume any waste.

In recent years, there has been increasing concern about the enormous amount of polyethylene and polypropylene waste found in landfills and awaiting decomposition for decades. Therefore, it is crucial to devise new methods for recycling plastic waste that could result in products with added value, such as oil sorbent sheets for oil spill control and prevention. The development of high-efficiency absorbent materials has been explored. Still, there has been little success in identifying a significant solution that combines the utilization of plastic waste with noticeable oil absorption capacity, as shown in Table 4.

4. Conclusion

This work provides a process concept to convert mixed plastic waste into reusable super oil sorbent. Initially, isolation and quantification of polyolefins (PP and PE) were carried out using float-sink, differential scanning calorimetry, and X-ray diffraction. The quantified polyolefins were mixed with NaCl and organic solvent, followed by spin coating and annealing. NaCl was chosen due to the following reasons: (a) as a cavity-forming agent which was later removed by washing with water, (b) it can be easily washed out using water hence organic solvents can be avoided, and (c) the targeted size of the cavities was 150–200 μm ; hence commercial grade NaCl was found to be the cheapest cavity-forming agent. Annealing enhanced crystallinity and facilitated chain

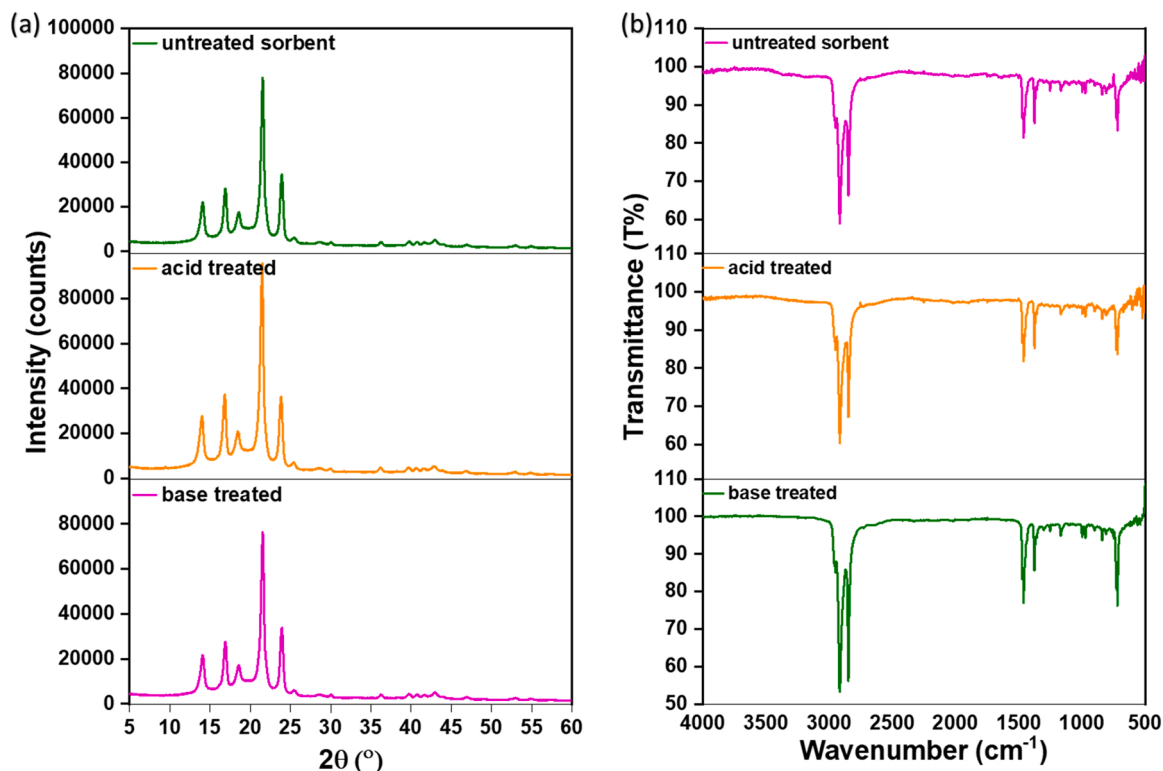


Fig. 14. XRD and FTIR spectra of untreated, acid-treated and base-treated sorbent (PE50PP50).

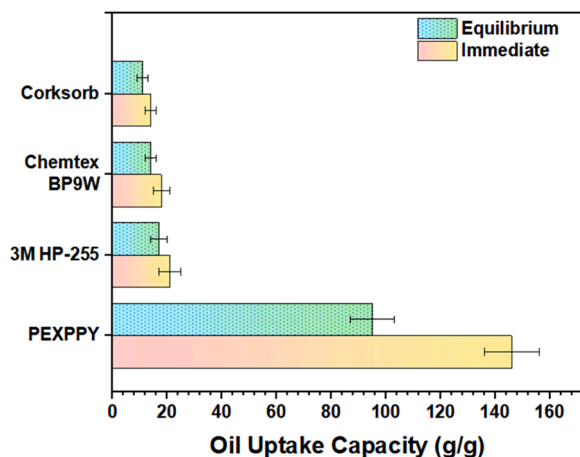


Fig. 15. Performance comparison with commercial sorbents using engine oil.

realignment within the polymer, resulting in improved mechanical strength. As a result, the film could be easily separated from the substrate.

The composition of different polymers in mixed plastic waste was determined using DSC. Analyzing FT-IR spectra, the molecular

interactions between PE and PP chains were used to identify their qualitative composition. Aside from that, XPS was performed to confirm that the solvent had been completely removed from the polymer surface. Thin film sorbents, inspected under a scanning electron microscope, contained macropores and cavities that allowed high oil uptake. Additionally, 3D representations of these cavities were projected onto profilometry images. These cavities significantly contributed to improved oil retention and enhanced absorption capacity as they offered surface roughness, a synergistic point for oil sorption and retention.

Developed super-sorbents contain pores and cavities which can expand twenty times their thickness, acting as sponges. The sorbent exhibited an oil uptake capacity of 70–140 g/g, depending upon the type of sorbate and dripping time. The results were compared with three commercial sorbents, namely Corksorb, 3 M-HP-255, 3 M-156, and Chemtex-BP-9 W, and found to be far superior. A mechanical or manual squeezing method can be used to recover the sorbed oil, ensuring that the sorbent can be reused up to 97% of the time. In contrast, the solvent extraction method recovered 100% of the oil.

The widespread use of plastics, coupled with their low-end and one-time uses, makes them one of the most polluting materials. A strategic and sustainable approach to managing plastics at the end of their lives is essential, given their importance to the global economy. In contrast to traditional recycling methods such as mechanical, chemical, or thermal, upcycling polymers offers a paradigm shift by creating higher-value

Table 3

Performance comparison with commercial sorbents.

	Oil Uptake capacity (g/g)		Type of material	Recyclability	Mode of recyclability	Waste utilization
	Equilibrium	Immediate				
Corksorb	10	13	special cork granules	No	N/A	N/A
Chemtex BP9W	14	18	Polypropylene	No	N/A	N/A
3MHP-255	17	21	Polypropylene	No	N/A	N/A
PEXPPY (our sorbent)	95	142	Plastic waste PE and PP	10 Cycles	Solvent extraction & manual squeezing	Utilize plastic waste

Table 4

A literature survey of waste utilization for oil absorption.

SN	Waste utilization	Oil uptake capacity g/g	Mode of recyclability	Recyclability/ Reusability	Ref.
1	Polyethylene Terephthalate (PET)	Crude Oil 22.9 Gasoline 11.1 Diesel 19.3 Pump Oil 19.6	Recycled by squeezing	5 times	[37]
2	Polyethylene Terephthalate (PET)	Chloroform 39.30 Soybean 27.65	Hand squeezing	10 times	[38]
3	PP Aerogel	6.0	Recycled by solvent	10 times	[39]
4	Unsaturated Polyester Resins	18.29 g/g	Solvent extractions and compression	10 cycles	[40]
5	Polyethylene (PE) and Polypropylene (PP)	NR	NR	NR	[41]
6	HDPE HD-75	Marine Diesel 100 Corn Oil 45 Mineral Oil 30	NR	NR	[42]
7	Recycled Polyethylene Terephthalate) (PET)	Crude oil 40 Toluene 35	NR	5 cycles	[43]

products from them. It is anticipated that the integrated approach demonstrated in this work could lead to more utilization of plastic waste as an abundant source of value-added materials.

Environmental implication

The widespread use of plastics, combined with their low-end and one-time usage, makes them one of the most polluting materials. A strategic and sustainable approach to managing plastics at the end of their lives is essential, given their importance to the global economy. In contrast to traditional recycling methods such as mechanical, chemical, or thermal, upcycling plastics offers a paradigm shift by creating higher-value products from them. This work attempts to present how the plastic industry could transform into a sustainable and circular sector by providing an alternate way of reusing plastic waste through upcycling into a reusable super-sorbent for oil and organics removal from water.

CRedit authorship contribution statement

Junaid Saleem: Conceptualization, Supervision, Methodology, Investigation, Writing – original draft. **Zubair Khalid Baig Moghal:** Methodology, Investigation, Formal analysis, Data curation, Visualization. **Gordon McKay:** Supervision, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability

No data was used for the research described in the article.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at [doi:10.1016/j.jhazmat.2023.131356](https://doi.org/10.1016/j.jhazmat.2023.131356).

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