

Ref.: R. E. Allred and J. M. Gosau, "Plant Requirements for Large-scale Chemical Recycling of Composite Materials," *Proc. of Composites and Advanced Materials Expo (CAMX)*, Anaheim, CA, September 26-29, 2016.

Plant Requirements for Large-Scale Chemical Recycling of Composite Materials

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ABSTRACT

Current methods of manufacturing composites produce large quantities of scrap. The manufacturing scrap primarily consists of uncured prepreg cut-offs and cured trim and cutouts. End-of-life components and full-scale test articles must also be considered in the scrap mix. Recycling and reuse of scrap composite materials is greatly needed; however, little of the scrap being produced currently is being recycled. It is recognized that technologies are needed for the collection, processing, and reuse of composite materials. A need exists for a practical, economical, scalable recycling process for reinforced thermosetting polymeric materials that does not substantially degrade the reinforcement properties and provides them in a clean, reusable condition. Our approach is to chemically degrade thermosetting polymers to release the reinforcement material for reuse. In this process, thermosetting polymers are broken down into a low molecular weight hydrocarbon mixture, using heat, pressure, and catalyst in a suitable reaction vessel containing a liquid heat transfer fluid. Recovered fibers, nanomaterials, whiskers, and fillers have near original properties for subsequent reuse. The process is rapid and readily scalable to handle thousands of tons of composite scrap. Both cured and uncured scrap sources can be readily processed. The recycling process involves collection and size reduction of scrap, processing in a reaction vessel, separation of heat transfer fluid, catalyst, and hydrocarbons from reinforcements, purifying recovered solids, separation of hydrocarbons, surface chemical treatment of solids, production of non-woven mat, and packaging of solids and hydrocarbons for reuse. Equipment needs and workflow through a large-scale recycling facility are discussed.

1. INTRODUCTION

There is growing concern by society regarding depletion of the earth's resources and energy. Current methods of manufacturing composites produce large quantities of scrap. The manufacturing scrap primarily consists of uncured prepreg cut-offs and cured trim and cutouts. End-of-life components and full-scale test articles must also be considered in the scrap mix. Recycling and reuse of scrap composite materials is greatly needed to save energy and protect the environment; however, little of the scrap being produced currently is being recycled. It is well recognized that technologies are needed for the collection, processing, and reuse of composite materials. As such, research and development of such technologies and their commercialization is being extensively pursued.

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To meet this need, there is a substantial amount of research being conducted around the world on recycling composite materials. Technologies being addressed include chemical [1-8], thermal [9-17], and mechanical [18-20], processes. There is also considerable research regarding the use of fluidized beds [21-25] and solvolysis using supercritical fluids [26-31]. There are also several reviews available on the state of the art [32-39].

The patent literature mirrors the technical literature. Most patented processes for recycling composite materials use thermal (pyrolysis [40-46]), (catalytic cracking [47]), or (microwave [48]), mechanical grinding [49,50], solvent cracking [51], or matrix resin modification [52] methods. The pyrolysis process, thermal cracking in a low oxygen environment, is being pursued commercially, particularly in Europe. Thermal processes are limited to composite scrap with thermally stable reinforcements, leave an undesirable char layer on the surface of the recovered solids, and degrade the mechanical properties of the recovered reinforcements. Mechanical grinding does not make good economic use of the expensive reinforcement materials. The solvent cracking process kinetics do not allow for a practical means of scaling up to handle large volumes of scrap.

There is still a need for a practical, economical, scalable recycling process for reinforced thermosetting polymeric materials that does not substantially degrade the reinforcement properties and provides them in a clean, reusable condition.

2. EXPERIMENTATION

Adherent Technologies, Inc. (ATI) has been exploring recycling technologies for organic-based materials for the last two decades. Those studies began using a vacuum pyrolysis technique on plastics and composites [53-57]. While most thermoplastics were successfully reduced to hydrocarbon species for reuse, carbon reinforcements from thermoset matrices such as epoxy had a residual surface char layer that was deemed undesirable for fabricating new composites with the reclaimed fibers. We then switched to a new high temperature, high pressure process using a liquid heat transfer fluid and basic catalyst. That process produced very clean, undamaged carbon fibers from all types of composite matrices [58-60]. In pursuing scale up options for producing large quantities of composite scrap, it became clear that the equipment for the high temperature process was too expensive. We then sought means to effectively recycle thermoset matrix composites at lower temperatures and pressures. After some years of development, a low temperature, low pressure process was developed and proven out [6-8, 61]. While all three processes in the portfolio are in use, depending upon the particular feedstock, the low temperature process has become the primary technology for commercial scale plants.

This paper describes the low temperature process, including subunit operations, and work flow for a large scale recycling facility. A process flow diagram for the low temperature process is given in Figure 1 for two reactors. As seen in Figure 1, during the process, size reduced scrap composite feedstock material is fed into a reaction vessel, where the feedstock is converted at low temperature and pressure into a low molecular weight hydrocarbon mixture and separated from the reinforcement solids in the presence of a liquid heat transfer fluid and catalyst. The

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resultant mixture is then separated into solids and liquids using a centrifuge. The liquid fraction is then used to process additional composite feedstock while the solid reinforcement phase is washed and dried for reuse. A suitable sizing or finish coating can be applied to the

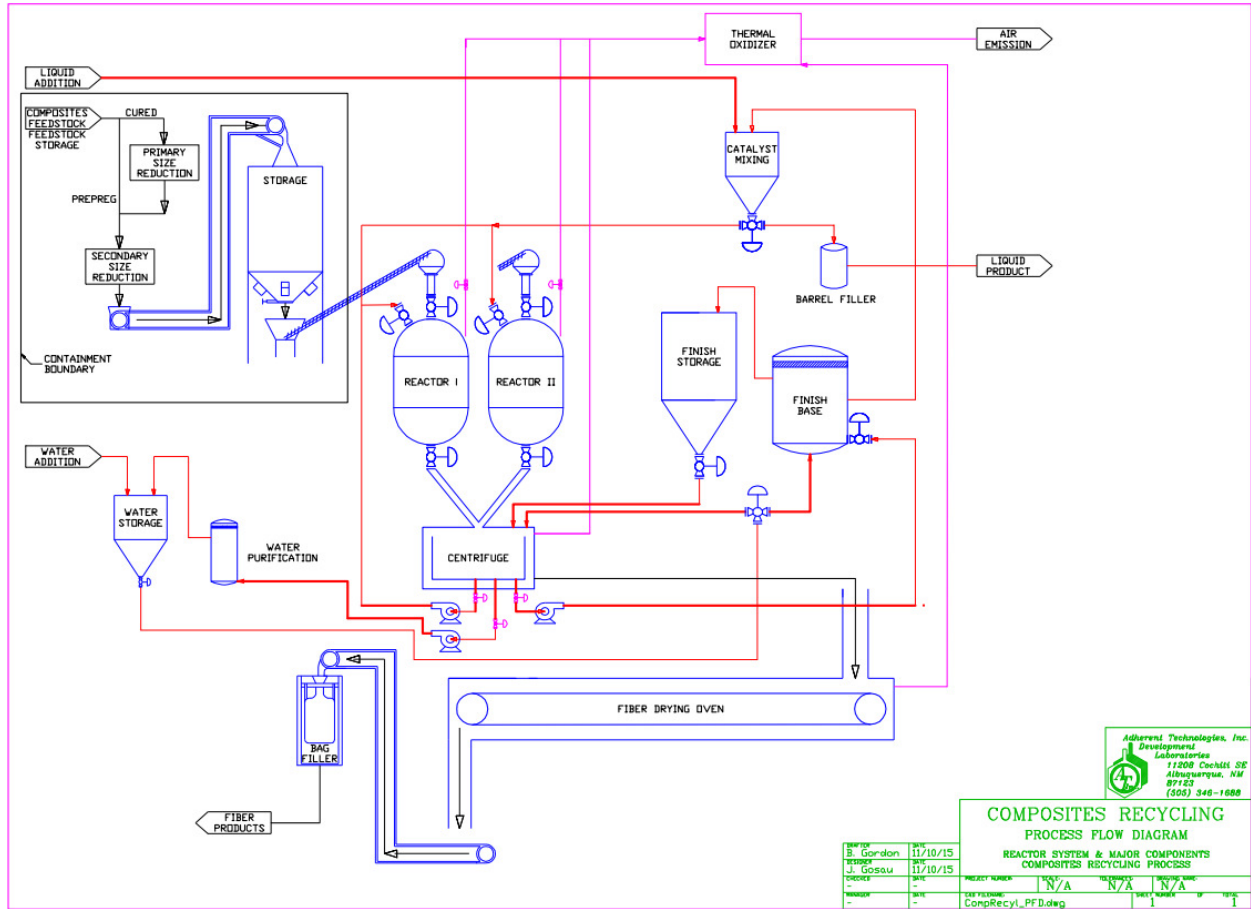


Figure 1. Process flow diagram for low temperature, low pressure chemical recycling process

reinforcements to enhance compatibility when producing new reinforced polymer matrix composite materials. Over time, fractions of the liquid volume are removed to capture a liquid hydrocarbon product.

Details of each step in the process shown in Figure 1 are presented in the Results section.

3. RESULTS

The first step in the recycling process involves collection and size reduction of scrap. Each of the composite waste streams must be size reduced for introduction into the reaction vessel. Size reduction of composite scrap is important because it provides a feedstock that the reactor can

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physically transport, affects the overall reaction rate, and affects the fiber length for the reclaimed fiber product.

The process requires that the composite scrap be size reduced into the range from fines (powder) up to 100 cm or more in length. The size-reduction unit operation will be one of the first in the process after collection and may be applied at collection centers to improve the economics of transporting feedstock to the processing site. By size reducing the composite scrap at the source, the bulk density of the material is increased, thus increasing the amount of material transported per unit volume. Size reduction also increases the surface area available for reaction, thereby increasing the rate of reaction in the process reactor. Very large pieces such as cargo door cutouts may need to be cut up using diamond blade circular saws before size reduction. Suitable size-reduction methods include shredders, hammer mills, grinders, and granulators, among others. Each method produces a unique product distribution, depending upon the feedstock type. Size reduction considerations are discussed in Ref. 62. It is likely that some combination of these methods will be needed in a large-scale facility.

The chemical recycling reaction is performed in a suitable reaction vessel, such as a chemical stirred tank reactor, containing one or more openings for the introduction and removal of chemicals and feedstock and equipped with a suitable mechanical stirrer, such as an anchor or "H" stirrer. The bottom portion of the vessel can be a flat surface or on inverted truncated cone. The volume of the reaction vessel can range between 200 liters (50 gallon) and 15,000 liters (4000 gallon), depending on the amount of scrap to be processed in one reaction. Our preferred layout consists of a module of six each 4000 liter (1000 gallon) reactors arranged around a central centrifuge. Depending upon the amount of scrap to be processed, as few as two of the six reactors in the module may be used as shown in Figure 1, with additional reactors added as additional scrap volumes become available.

The reaction vessel is a glass lined steel reactor heated with a jacket, bottom heaters, and/or internal heaters. The reaction vessel may also be constructed of highly corrosion resistant alloys, such as Hastelloy 256, Carpenter 20, or other suitable alloy. The temperature, stir rate or agitation, pressure, mass of feedstock, pH, and liquid to solids ratio are adjusted to maximize the reaction rate and resultant composite throughput. The preferred reaction temperature is in the range between 120 and 180°C, pressure is in the range between 0 and 1 MPa (150psi), stir rate is in the range between 0 and 50 rpm, and the solids to liquids ratio is less than 25 percent.

During the reaction, the low molecular weight hydrocarbon fraction formed by breaking down the thermosetting resin becomes part of the liquid heat transfer fluid. The process works equally well with uncured prepreg and cured laminate. The process is for tertiary recycling of thermoset polymer-based composites, including fiber, filler, whisker, and nanomaterial-reinforced materials. The reinforcements are recovered with their structural and mechanical properties largely intact.

Common thermosetting polymers, such as epoxies, vinyl esters, polyesters, phenolics, polyimides, and other types, are thermochemically broken down into a mixture of low molecular

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weight hydrocarbons and separated from the reinforcements. The thermochemical reactions include chain cleavage, depolymerization, dehydrogenation, hydrocracking, and hydrolysis. The process is applicable to most types of composites and takes place under mild conditions and has virtually no environmental impact.

At reaction completion, the hot heat transfer fluid, low molecular weight hydrocarbons, catalyst, and the solid reinforcements are pumped, drained, or dumped into the centrifuge to separate the liquid fraction from the solid fraction. A scrubber is operatively connected to a port on the reaction vessel or centrifuge to condense any vapors formed in the process. The scrubber can be directed into another separator device.

The liquid fraction containing the heat transfer fluid and reaction catalyst is then pumped into a separate reaction vessel that was previously charged with composite scrap to begin a new recycling reaction. Periodically, the low molecular weight hydrocarbon fraction of the liquid phase must be removed by distillation or other suitable separation process to maintain the appropriate amount of liquid volume in the system. The removed hydrocarbon fraction functions as a chemical feedstock or fuel, as appropriate. We have previously shown that the hydrocarbon products are excellent starting materials for making phenol-formaldehyde adhesives for bonding wood products, such as plywood, particle board, and pressboard [60]. They may also be effectively used to make phenolic finishes for carbon fibers [63].

One or more refining units downstream of the separation unit are required, including distillation, hydroprocessing, vacuum pyrolysis, or drying. The fiber mass is washed and purified in the separation device and introduced into a drying oven held near 100°C. The wash water is purified in an oil-water separator for reuse. Where solvent is used to purify the solid fraction, the solvent is reclaimed through distillation or other suitable separation process. Once dried, the fibers or other reinforcements are introduced into a suitable medium for storage, processing, and/or shipping.

The chemical recycling reaction produces mostly a low energy, dead surface that does not form a strong interfacial bond. A strong interface is necessary in short fiber-reinforced composites to maximize strength according to shear lag theory. To improve the interfacial bonding, ATI's reactive finishes may be added as an aqueous emulsion after the fiber washing step. These finishes have been shown to improve wetting and adhesion with numerous matrix resins [63-67]. Suitable finish (sizing) carrier resins include epoxy, phenolic, vinyl ester, polyester, bismaleimide, polyimide, thermoplastic, silane, and other compatibilizing coatings for composite processing. Once the reactive finish is applied to the reclaimed fibers, they are subjected to a drying/reaction heating step as shown in Figure 1.

After the finishing step, one preferred form for reusing the reclaimed fibers is as a non-woven fiber mat, wherein the fiber mass is combed or carded to separate tangled material and formed into a roll of random in-plane fibers with areal weights ranging from 100 g/m² to 1000 g/m². There are numerous advantages of using aligned non-woven fabrics of short fibers, the biggest of which is achieving much higher volume fractions in a fabricated composite. Alternative means

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for handling the reclaimed fibers include pelletizing, milling, and bulk packaging in storage sacks.

4. CONCLUSIONS

A need exists for a practical, economical, scalable recycling process for reinforced thermosetting polymeric materials that does not substantially degrade the reinforcement properties and provides them in a clean, reusable condition. Our approach is to chemically degrade thermosetting polymers to release the reinforcement material for reuse. In this process, thermosetting polymers are broken down into a low molecular weight hydrocarbon mixture, using heat, pressure, and catalyst in a suitable reaction vessel containing a liquid heat transfer fluid. Recovered fibers, nanomaterials, whiskers, and fillers have near original properties for subsequent reuse. The process is rapid and readily scalable to handle thousands of tons of composite scrap. This thermochemical process is a marked advance over thermal degradation, supercritical solvent, or solvent cracking techniques.

Both cured and uncured scrap sources can be readily processed. The recycling process involves collection and size reduction of scrap, processing in a reaction vessel, separation of heat transfer fluid, catalyst, and hydrocarbons from reinforcements, purifying recovered solids, separation of hydrocarbons, surface chemical treatment of solids, production of non-woven mat, and packaging of solids and hydrocarbons for reuse. Equipment needs and workflow through a large-scale recycling facility have been identified.

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