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A case for closed-loop recycling of post-consumer PET for automotive foams

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ABSTRACT

Striving to utilize sustainable material sources, polyester polyols made via glycolysis and esterification of recycled polyethylene terephthalate (rPET) scrap were used to synthesize flexible polyurethane (PU) foams typically found in automotive interior applications. The objective of this endeavor was to ascertain if a closed-loop model could be established with the discarded PET feedstock. In five separate formulations, up to 50% of the total polyol content (traditionally derived from petroleum-based feedstock) was replaced with the afore-mentioned sustainable recycled polyols. These foams underwent mechanical, thermal, morphological, and physical characterization testing to determine feasibility for use in an automotive interior. Young's modulus, tensile stress at maximum load, tear resistance, and compression modulus all increased by combined averages of 121%, 67%, 32%, and 150% over the control petroleum-based formulation, respectively, in foams possessing 50% rPET polyol content. Thermal stability also increased with sustainable polyol content; thermogravimetric analysis showed that 50% mass loss temperature increased by an average of 20 °C in foams containing 30% recycled polyol. Properties of density and SAG factor remained within 5% of the control petroleum-based reference foams. After comparing these findings to traditional polyols, a compelling argument can be made for the use of post-consumer automotive and industrial feedstocks in developing high-performing interior automotive PU foams.

1. Introduction

Polyurethane (PU) foams are composed of a polymer that is formed from the reaction of polyols and poly-isocyanates, which yields a complex structure comprising of long flexible groups and rigid PU linkages. Manipulation of the polyol's characteristics (the degree of cross-linking, molecular weight, viscosity, hydroxyl number, etc.) in conjunction with the blowing agent, catalysts, and other reaction characteristics determine the final properties of the foam, while giving rise to the numerous different types of PU-based polymers in industrial production today. Therefore, PUs possess great versatility, and have applications in diverse markets, including furniture, bedding, carpet, packaging, paints & coatings, and automotive, depending on their formulation and properties (Herrington and Hock, 1997). Even inside a single vehicle, there are multiple kinds of foams that would benefit from improvements to the overall PU synthesis footprint, for example, in car seats, sound insulation, interior trim components, and carpet underlay. To stay competitive, modern car manufacturers continue to work with their suppliers to reformulate and refine their choices of foam as they focus on achieving high customer comfort and satisfaction. PU foam is a key component in many areas critical to a vehicle's user experience, including energy and noise absorbing insulation and supportiveness and feel of the seat cushioning (Maxwell, 1994).

Traditionally, most PU products are created from petroleum-based polyols (Randal and Lee, 2003). Polyethers and polyesters are the most common polyol types, with polyethers dominating 80%–90% of industrial polyol use (Randal and Lee, 2003; Lefebvre et al., 2005). These non-renewable sources carry large environmental impact in their production by causing considerable greenhouse gas emissions (DeBolt et al., 2016). For instance, when considering one kilogram of the most common construction and insulation materials, PU foam carries the highest observed environmental impact, at 4.42797 kg CO_2 eq. (Boemi et al. 2016). To combat this undesired effect, a universal switch to a recycled source or a suitable waste stream would carry an extensive impact, due to the economies of scale when dealing with worldwide PU production. The global PU market is estimated to be 18 million kilo tons in 2016 (Markets and Markets, 2016). Additionally, flexible foams represent close to 50% (~9 million kilo tons) of the total PU market

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(Simon et al., 2014; Zevenhoven, 2004). Thus, if even half of the total used polyol, or approximately 25% of the final foam composition (~2.25 million kilo tons), could be sourced from waste streams/recycled material thereby eliminating the need for additional production of chemicals, it could reduce greenhouse gas emissions by millions of kilo tons CO_2 eq. per year for PU production worldwide (~4.43 kg CO_2/kg PU foam) (Boemi et al., 2016).

The recycled material used in this research is polyethylene terephthalate (PET). Also known by its common name "polyester", PET is the fourth most produced industrial polymer in the world. It is used heavily in textile and as clothing fibers, soft drink bottles, food packaging, as well as in headliners, carpets, truck liners, underbody shields and seat fabrics of automobiles (NAPCOR, 2015). Due to this large presence in common consumer products, PET is frequently the largest kind of solid plastic waste in many urban environments (de Mello et al., 2009; Matos and Schalch, 2007). The total weight of PET bottles and jars available for recycling in the US in 2014 was 2.7 billion kilograms, and the total amount collected for recycling amounted to 0.82 billion kilograms, resulting in a gross recycling rate of 31.0% in 2014. That rate has been increasing on average for over a decade, making PET a promising prospect for being available for mass-scale application of PET-based polyols (NAPCOR, 2015).

Post-consumer PET material can be incorporated into PU foams physically, by being ground up and used as filler, or chemically, via depolymerization into polyols (Brennan, 1982; Duldner et al., 2013; Dian-Yong et al., 2013; Tabor et al., 2016a; Yeakey and Cuscurdia, 1984; Mukerjee et al., 2015; Trowell, 1988; Shieh, 2013; Tabor et al., 2016b,c; Brennan and Speranza, 1985; Dang et al., 2016). Polyols that have been successfully made from recycled PET (rPET) were obtained employing various methods, with the predominant method being glycolysis (Paszun and Spychaj, 1997; Patel et al., 2005; Paberza et al., 2015; Lee et al., 1994; Cuk et al., 2015; Chaudhary et al., 2013; Ivdre et al. 2016). Many sources have noted these PET polyols have drawbacks, including high viscosity and incompatibility with physical blowing agents (Paberza et al., 2015; Vitkauskienë and Makuðka, 2008; Ionescu, 2008; Beneš et al., 2013). However, the same studies have shown the rPET-based polyols were successfully used to create PU foam with improved mechanical and chemical resistance properties compared to traditional PU foams (Paberza et al., 2015; Patel et al., 2005). Others have found an increase in the concentration of rPET polyol in the foam formula was associated with an increase in compression strength, retention of a low, insulating thermal conductivity over the foam's lifetime, and increased thermal stability (Paberza et al., 2015; Lee et al., 1994). This paper represents the first attempt to unify these known benefits of rPET polyols with an application suited for a supportive, thermally resistant flexible PU foam; namely, automotive engine covers or interior headliners. Ideally, these and future experiments focused on achieving engineering material specifications will bridge the gap between research of this technology and its transition to mass production methods for insulating automotive PU foams.

Beyond the benefit already gained from using recycled materials, there is additional opportunity in the use of PET from recycled automotive components. One study found that there was approximately 2.2 kg of PET contained in an average passenger vehicle, and when taking into account that greater than 15 million vehicles have been sold annually in the US each year since 2013, that scales up to roughly 30 million kg of PET used for automotive components alone (Staudinger and Keoleian, 2001; Auto Alliance, 2016). By replacing petroleum derived polyols with polyols sourced specifically from this vast amount of automotive waste, a 'closed loop' automotive foam composed partially of a previous waste stream can be produced, allowing for progress towards replacing the PU foam entirely with more sustainable materials with lower environmental impact. Many patents have indicated that synthesizing a functioning polyester polyol can be achieved with a wide range of PET feedstocks (Brennan, 1982; Duldner et al., 2013; Dian-Yong et al., 2013; Tabor et al., 2016a; Yeakey and Cuscurdia, 1984; Mukerjee et al., 2015; Trowell, 1988; Shieh, 2013; Tabor et al., 2016b,c; Brennan and Speranza, 1985). Therefore, if automotive PET components and scrap were used in this technology, similarly viable polyols would be produced, and a 'closed loop' recycling system could be realized. The purpose of these experiments was to determine the stability and viability of rPET polyols in production of PU flexible foams for automotive applications. Physical, mechanical, and thermal properties were measured and compared to control samples purely composed of petroleum-based polyol.

2. Materials and experimental methods

2.1. Materials

Flexible PU foams were formed from the traditional method of reacting polyols with polyisocyanates. The petroleum-based polyol Voranol 4701 was sourced from Dow Chemical Co. (Midland, MI), and served as a reference for property comparisons to commercial foam products. Recycled PET-based polyols FFP1000-1.2, FFP1000-2.1, FFP1000-2.2, and FFP1000-2.3 was generously provided by Resinate Materials Group, Inc. (Plymouth, MI). Rubinate 7304, a liquid blend of MDI series polyisocyanates, was used as the isocyanate component, provided by Hunstman International LLC (Auburn Hills, MI). The breakdown of the amount of recycled PET and bio-based content in each polyol's overall composition was provided by Resinate Materials Group, Inc. in Table 1.

It should be noted that subsequent data in this paper's graphs are clustered in ascending order of increasing recycled content, left to right, to highlight how the recycled content factors into the mechanical and thermal properties, as the use of rPET materials was the focus of these experiments. Additionally, in the following figures displaying results from foams made with the four polyols, darker shading represents a relatively higher amount of recycled content in that particular polyol. A table of each reactants individual properties, including hydroxyl number, is shown in Table 2.

The following foaming additives were used in foam production: deionized water - blowing agent; Tegostab B4690 – silicone-based surfactant (Evonik, Hopewell, VA); Lumulse POE (26) GLYC – reactive cell opener (Lambent Corporation, Gurnee, IL); Diethanolamine – co-catalyst agent with cross-linking functionality (Sigma-Aldrich Co, St. Louis, MO); Niax A300 and Niax A1 – catalysts (Momentive Inc., Waterford, KY). These agents were added consistent with the quantities listed in the formulations given in Table 3 below. Note that the isocyanate component was added relative to the overall –OH number of the polyol-side blend, which varied on the rPET polyol in a particular formulation (see Table 2). A release agent, Chem-Trend PU-11331, was used with the laboratory mold system for ease of foam removal. Based on preliminary results, formulations were kept below 50% rPET polyol due to high compression set, which is not favorable for certain flexible PU foam applications, including headliner and seating applications.

2.2. Polyol synthesis

The synthesis of the polyester polyol employing rPET scrap procured from water bottle and other PET waste streams was conducted using Resinate's proprietary glycolysis process technology. The experimental polyester polyol was targeted for flexible foam applications. The polyester polyol includes up to 95% sustainable content. Resinate Materials Group defines sustainable content as

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