Life Cycle Energy Analysis and Environmental Life Cycle Assessment of Carbon Nanofibers Production

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Abstract-*Life Cycle Assessment of Nanotechnology has been suggested to evaluate claims about the potential benefits of this emerging technology. This paper presents one of the first LCAs and Life Cycle Energy Analysis of Vapor Grown Carbon Nanofibers (CNF) synthesis. Life Cycle inventory data is compiled with data reported in the open literature. CNFs are compared with traditional materials on an equal mass basis to quantify the life cycle energy intensity and environmental burden. The results of the study indicate significantly higher life cycle energy requirements and higher environmental impact of CNFs as compared to traditional materials like aluminum, steel and polypropylene. Savings in life cycle energy consumption and possibly a reduction in environmental burden are envisaged if higher process yields of these fibers can be achieved in continuous operations. Since the comparisons are performed on an equal mass basis, these results cannot be generalized for CNF based nanoproducts and quantity of their use may decide their cradle to grave impact. Specific CNF based applications need to be studied to evaluate their environmental performance and are the topics of future work.*

I. INTRODUCTION

Carbon Nanofibers (CNFs) belong to a new class of materials that have exceptional mechanical properties like high tensile strength (12000 MPa) that is approximately 10 times that of steel [1-2]. Besides mechanical strength, CNFs possess novel electrical properties like high electrical conductivity. These properties of CNFs are being explored in a variety of ways by imparting functionalities in various intermediate and final value-added consumer products. Applications include the use of CNFs as polymer additives for high strength Polymer Nanocomposites, use of CNFs in carbon-lithium batteries, start capacitors for electronic devices and electrically conducting polymers [2]. High specific surface area of these fibers is an additional attribute that has been investigated for use of these Engineered Nanoparticles as catalyst support material especially for liquid phase reactions [1]. Although these properties make CNF based nanoproducts commercial attractive, they also raise concerns and pose difficult questions about the human and ecosystem impact of these materials. The bitter lessons of the unfounded technological exuberance of some of the earlier technologies such as chlorofluorocarbons, asbestos, insecticides etc. should be remembered before reaching any concrete conclusions about the impact of nanoproducts. The point here is not to discount the benefits of these technologies for human welfare. However, the underlying message is that the costs of failure to see beyond the early hype into the broader implications of any technology can be enormous.

In the case of nanotechnology, this entails following a proactive approach to ensure smart and sensible research and development thereby leading to a sustainable nanotechnology industry. There is an immediate need to study the toxicity, fate and transport, and mechanism of damage of the products and byproducts of nanotechnology [3]. However, focusing attention on one process without sufficient attention to others in a product's supply chain can lead to the unintended environmental tradeoff of one problem for another. The use of a holistic and systems approach can help avert such problems. One such methodology is Life Cycle Assessment (LCA) that considers environmental impact of products or processes over their entire life cycle.

Researchers have identified the need for the LCA of potential nanoproducts [4-6]. However, LCA of nanotechnology poses several formidable challenges. Firstly, the existing life cycle inventory databases are limited in scope and are useful for evaluating only common products and processes. Secondly, there is very little quantifiable data available on the human health and ecosystem impacts of products and byproducts of nanomanufacturing. Besides these, nanotechnology is still in its infancy and nanomanufacturing processes are evolving rapidly. In the light of these challenges, LCA of Nanotechnology has received very little attention. Lloyd and Lave employed the EIOLCA framework to study nanotechnology applications for automobile body panels and catalytic converters [7-8]. Osterwalder and coworkers [9] compared the wet and dry synthesis methods for oxide nanoparticle production with respect to energy consumption. Details such as release and impact of emissions during nanomanufacturing are not accounted in these studies possibly due to lack of information. Further, equipment level information that is both energy and emissions, for the synthesis of nanoparticles is missing. Although these studies represent first important step towards LCA of nanomanufacturing, their accuracy is limited for guiding selection among alternatives. No *nanoparticle specific LCAs exist in the literature that uses detailed process level information and combines that with details from other steps*

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in the life cycle to quantify the energy and environmental burden associated with the production of nanomaterials. Such studies will be especially useful since they will lay the basis for compiling the life cycle inventory for nanoparticle synthesis and hence pave the way for LCA of specific nanoproducts. This is especially important at an early stage of research to evaluate the economic-environmental trade-offs among manufacturing processes and among alternative products to aid in sensible engineering decision-making.

This paper presents one of the first LCAs for synthesis of Carbon Nanofibers along with an analysis of Life Cycle Energy requirements. Section II describes the details of the process considered for the synthesis of CNFs along with a description of the data sources and assumptions. Section III of the paper briefly introduces the concept of life cycle assessment and the tools and techniques used in this study. Section IV presents the results of the Life Cycle Energy Analysis of CNF synthesis. A comparison of CNFs with traditional materials is presented to get insights into the life cycle energy intensity of CNF production. Section V describes the results of the "cradle-to-gate" Environmental LCA of CNFs and a comparison of CNFs with traditional materials on an equal mass basis. Both midpoint and end-point indicators are presented. Finally, Section VI briefly summarizes the conclusions of this study and the directions for future work.

II. CARBON NANOFIBERS- PROCESS DESCRIPTION

The avoidance of formation of carbon deposits has been of utmost importance in refinery applications like steam reforming of hydrocarbons, hydrocracking, hydrotreating etc. primarily because these carbon filaments leads to deactivation of the catalyst surface, blockage in reactor systems and reduction in heat transfer [1,10]. However, in recent years, CNFs have received a great deal of attention both from the research and industrial community because of their novel properties that can be realized in a variety of ways in various applications. Several academic and industrial research groups have directed their efforts towards synthesizing and optimizing the growth of these CNFs. In its simplest form the catalytic synthesis of CNFs consists of formation of these fibers on metallic catalysts in the form of powders, foils, gauzes or supported particles. The process consists of reduction of the catalyst sample in a hydrogen-inert gas stream at a somewhat lower temperature followed by heating the catalyst up to the reaction temperature subsequent to which the reaction mixture consisting of hydrocarbon, hydrogen and inert gas is introduced into the system [11-13]. Several different metallic and bimetallic catalysts can be used. Most commonly used catalysts are iron, cobalt, nickel and copper both in bulk and supported form. Lower hydrocarbons like methane, ethylene, acetylene or benzene and CO are the common source of carbon material.

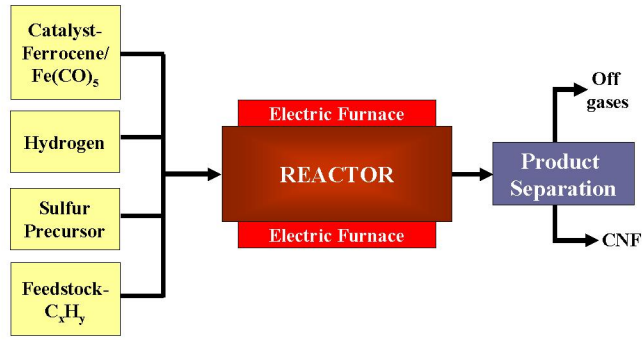

Fig. 1. Typical Process Schematic of VGCNF Synthesis

However, despite great advances in synthesis methods and efforts at understanding the mechanism of nucleation and growth of CNFs, continuous production of these fibers has proven to be challenging and several issues need to be addressed. In recent years, a new method capable of synthesizing these Carbon Nanofibers on a continuous scale namely the Vapor Grown Carbon Nanofibers (VGCNF) has emerged [14-17]. A schematic of the VGCNF synthesis is shown in Figure 1. VGCNFs are produced by catalytic pyrolysis of hydrocarbons in the presence of a transition metal acting as the catalyst. Trace amounts of Sulfur is added to the feed to promote the formation of CNFs. Ferrocene dissolved in a suitable solvent or Iron Pentacarbonyl, $Fe(CO)_5$ are commonly used catalyst sources. The organometallic catalyst decomposes forming clusters of Fe particles that act as nuclei for the formation and further growth of CNFs. The fibers coming out of the reactor along with the off-gases are trapped or separated using a series of cyclone separators or trap mechanisms located downstream of the reactor. The entire reactor assembly is enclosed within an electric furnace to supply sufficient heat to maintain the pyrolysis temperature of 1100-1200^{0}C.

III. TOOLS AND TECHNIQUES

Traditional engineering approaches tend to focus on the process level details without caring about other steps in a product's life cycle. Such reductionist thinking can lead to a shifting of the problem outside the analysis boundary. This however, can be resolved by the use of a more holistic life cycle approach which is the underlying theme of this study. Process energy requirements are first calculated by applying an overall energy balance around the CNF synthesis reactor. The indirect life cycle energy requirements due to the consumption of other inputs like hydrocarbon feedstock, carrier gas and sulfur source are obtained directly using the SimaPro software. The above two are combined to get the overall life cycle energy requirements for the synthesis of CNFs. Here "*life cycle energy consumption*" primarily refers to the cumulative fossil energy requirements for the synthesis of CNFs and does not include the contribution of ecosystem goods and services. Thus, it quantifies the non-renewable energy requirements along the supply chain of CNFs.

A natural follow up and a relevant question to the life cycle energy analysis is the Environmental burden associated with CNF production. This is estimated using the Process

LCA. Life Cycle Assessment is a methodology to quantify the environmental impact of a product or a process over its entire life cycle. In its simplest form, LCA methodology involves four basic steps: Goal and Scope Definition, Inventory Analysis, Impact Assessment, and Interpretation.

Goal and Scope Definition- In general, the scope of a LCA study comprises of the resource extraction, inputs synthesis, product manufacture, waste treatment, packaging, use phase and finally ends when the product is finally disposed off or recycled back. Such an analysis is termed as "Cradle-to-grave". This study however, is a "Cradle-to-gate" analysis of CNF synthesis since no specific nanoproducts are being investigated at this stage. The aim is to quantify and compare the environmental burden of CNF synthesis with traditional materials on an equal mass basis. In the case of CNF synthesis, since a lot of data is either missing or proprietary information, certain assumptions have to be made to carefully define the process boundary. For example, emissions from CNF synthesis are not included due to lack of information. In addition, emissions corresponding to the catalyst life cycle are not included again due to the lack of data. These omissions are likely to introduce some error and uncertainties in the final results, but will still represent and yield a first set of "*conservative*" estimates for the LCA of CNFs. Emissions corresponding to the production of all other inputs required for CNF synthesis are incorporated in the study.

Inventory Analysis- As discussed in Section I, performing LCA of nanotechnology is a daunting task because of the severe lack of data about the inputs and outputs of nanomanufacturing. In this study, this challenge is addressed by compiling life cycle inventory for VGCNF synthesis based on laboratory experience and data available in the open literature [15-17]. Missing data is reconciled based on suitable assumptions and ensuring the conservation laws of mass and energy. Life cycle inventory data for common inputs is taken directly from "SimaPro 6.0 Demo version". SimaPro is a widely used LCA software and includes Life Cycle Inventory data for several minerals and chemicals. SimaPro consists of "cradle-to-gate" life cycle inventory data for Western European technologies. Inventory data for traditional materials like aluminum, steel and polypropylene is obtained directly from SimaPro too.

Impact Assessment- This phase involves classifying and characterizing emissions into various impact categories to provide several indicators for analyzing the potential contributions of the resource extractions and wastes/emissions in an inventory to a number of potential impacts. Several Life Cycle Impact Assessment (LCIA) approaches exist and these have been described and critically assessed in detail in the literature [19,20]. In this study, a hierarchical approach as described by Bare et.al. [20] is used. The emissions are classified and characterized based on their impact into various impact categories often described as Midpoint Indicators. Midpoint indicators can be further aggregated to obtain Endpoint Indicators. Endpoint indicators are often combine together to provide more meaningful and interpretable damage indicators. These damage indicators are described in detail along with the results in Section V. It is important to mention here that the level of uncertainty increases as we move from midpoints to the endpoints. Therefore, a hierarchical approach is used to provide a set of metrics at various levels.

In this study, the CML approach developed by the Leiden University Insititute of Environmental Sciences is used to obtain midpoint indicators. EcoIndicator 99 methodology is used to obtain Damage Indicators [21]. Under Eco-indicator, only three kind of environmental damages are weighed, namely damage to human health, damage to ecosystem quality and damage to resources. Damage to Human Health is expressed in terms of DALYs (Disability Adjusted Life Years). DALYs scale originally developed by Murray et.al. for WHO and World Bank incorporates damage to human health in the form of Years of life lost (YLL) and years of lives disabled (YLD) as a result of emissions of substances along the supply chain of a product. Damage to ecosystem quality refers to the damage caused to non-human species that is plant and animal species. Under Eco-indicator 99, Damage to Ecosystem is expressed in terms of PDF times area times year ($PDF*m^2*yr$), where PDF stands for potentially disappeared fraction and includes percentage of species that are threatened or that disappear from a given area in a given time.

IV. LIFE CYCLE ENERGY ANALYSIS

VGCNF synthesis from hydrocarbons is considered for quantifying the Life Cycle Energy Requirements. Process energy requirements are first calculated by applying an overall energy balance around the reactor. The boundaries for the process scale energy requirements are limited to the reactor. Feed is introduced at 22^0C with the cracking of hydrocarbons occurring at 1100^0C. An yield value of 0.5 as reported by Tibbetts et.al. [15] is assumed though Fan et.al. [17] reports yield values ranging from 15% to a maximum of 30 % by weight. Material inputs are compiled based on data available in the published literature [15-17]. A purification efficiency of 90% is considered for the CNFs. The indirect life cycle energy requirements are obtained directly using the SimaPro software. The above two are combined to get the overall life cycle energy requirements for the synthesis of CNFs. It is important to point out that the energy requirements for separation and purification of the produced CNFs from the fiber mixture are not accounted for thus resulting in conservative estimates of the life cycle energy requirements.

Three different feed stocks i.e. methane, ethylene and benzene are considered to quantify the life cycle energy intensities of VGCNFs. Different scenarios for catalyst and carrier gas recycle rates are evaluated to address uncertainty and evaluate the bounds on the life cycle energy consumption and emissions. Figure 2 represents the life cycle energy requirements for CNF synthesis for different feedstocks. Figure 2 presents a direct comparison of the life cycle energy requirements for CNF synthesis with those of traditional materials namely Aluminum, Steel and Polypropyl-

ene. The plot shows that the life cycle energy requirements for CNF synthesis ranges from 654 MJ/kg for Benzene feedstock to around 1807 MJ/kg for Methane with Hydrogen as the carrier gas. A further comparison with aluminum, steel and polypropylene reveals that the life cycle of CNFs is energy intensive with the life cycle energy requirements being 86, 30 and 119 MJ/kg for aluminum, steel and polypropylene respectively.

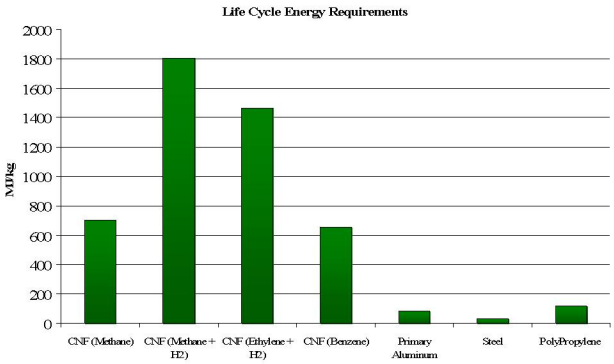

Fig. 2. Life Cycle Energy Requirements- Carbon Nanofibers vs. Traditional Materials

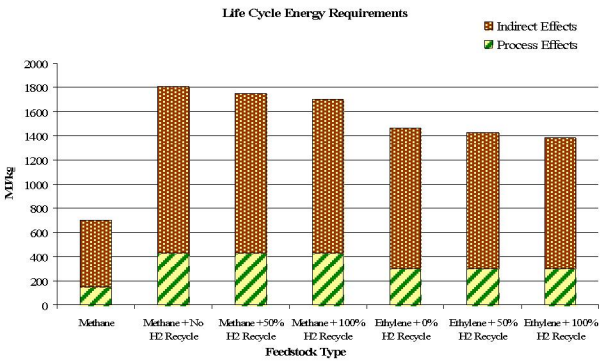

Fig. 3. Life Cycle Energy Requirements- Effect of Carrier Gas Recycle

The catalyst life cycle is not accounted for in the absence of life cycle inventory data for the catalyst thus resulting in lower bounds on the CNF life cycle energy requirements. The plot in Figure 2 indicates the lowest energy requirements for the benzene followed by methane feedstock alone, however most industrial synthesis is carried out in the presence of a carrier gas that is generally H_2, NH_3 or some other inert gas. A sensitivity analysis is further carried out to study the influence of carrier gas recycle rates on the life cycle energy requirements. The results are displayed in Figure 3. Considering the methane feedstock, it is observed that even with complete recycle of the hydrogen stream that is only theoretical in nature, the total energy requirements decreased from 1807 MJ/kg to 1702 MJ/kg. This corresponds to a 6% decrease in the overall life cycle energy requirements. A similar trend is observed for ethylene feedstock where a complete recycle of the hydrogen stream corresponds to a 5.6% reduction in the overall energy requirements. It is noteworthy to mention here that the energy required for separating H_2 from the off-gases coming out of the reactor is not accounted here. Therefore, the numbers presented in Figure 3 represents an upper bound on the savings in energy consumption that can be achieved when a carrier gas separation and recycle system is employed. A comparison of the CNFs with traditional materials like aluminum, steel and polypropylene reveals that life cycle energy requirements for CNFs is 6-60 times that required by these traditional materials. Several reasons can be attributed to this enormous difference. Firstly, processes required for manufacturing traditional materials are well-studied and have been optimized with respect to material and energy consumption over the last century or so. On the other hand, although researchers have been synthesizing CNFs on a laboratory scale, industrial synthesis of these nanoparticles remains a challenge. Secondly, industrial yields of these fibers are low even for continuous operations, i.e. in the range of 10-30 % by weight of the feedstock. This again entails research efforts for process improvement to increase the yield of these nanoparticles to make them competitive with alternatives for a given application. In addition, economies of scale have still not been realized for the production of CNFs. Unless, these materials are used in a widespread way especially in large volume applications like nanocomposites, it will be challenging to exploit the economies of scale and hence make these materials cost competitive with other alternatives. Researchers are also exploring the possibility of other low temperature processes like the Plasma enhanced CVD for the synthesis of CNFs [18]. These processes are expected to be operating at room temperatures thus eliminating the need for process heating. However, converting these laboratory scale processes to full-fledged continuous industrial operation will be challenging. Needless to mention, LCA of these new processes should be an integral component of research efforts in order to ensure that savings in energy and material resources are not offset by increased consumption in other phases of the life cycle.

V. ENVIRONMENTAL LCA OF CNFs

Environmental Impact Assessment is carried out for the synthesis of CNFs and both midpoint and endpoint indicators are obtained. Since the scope of this study is restricted to a "cradle-to-gate" LCA of CNFs alone and no specific nanoproducts are evaluated, the results of CNF LCIA is compared with traditional materials on an equal mass basis. Figures 4 and 5 present a comparison of some of the midpoint indicators for CNFs with aluminum, steel, and polypropylene on a per kg basis. Two base cases are evaluated for CNF synthesis, one with Methane and the other with Ethylene as the feedstock. Both cases are considered to have hydrogen as the carrier gas in accordance with the current industrial schemes.

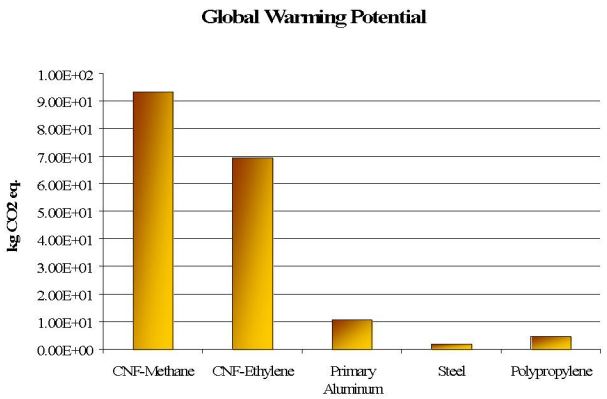

Fig. 4. Midpoint Indicator- Global Warming Potential

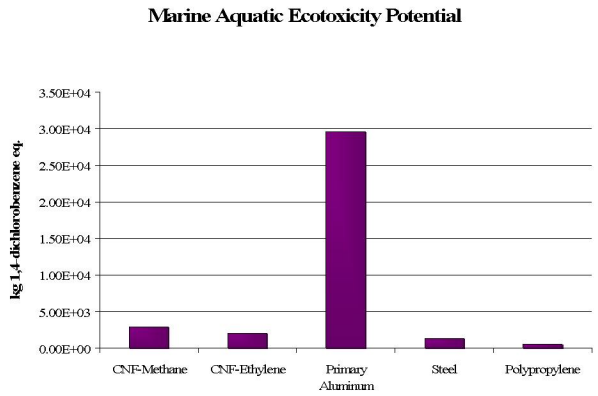

Fig. 5. Midpoint Indicator- Marine Aquatic Ecotoxicity Potential

Figure 4 shows a higher Global Warming Potential (GWP) for both methane and ethylene based CNFs when compared with aluminum, steel and polypropylene on an equal mass basis. Similar trends are observed for Acidification Potential, Eutrophication Potential, Human toxicity Potential and Photochemical Smog Formation Potentials. One argument that is often given in favor of the potential impact of nanomaterials is the relative small quantities in which these materials are required in various end product applications. However, the estimated production rates are 58000 metric tonnes of ENPs in the period from 2011-2020 [4,22]. This can have an impact equivalent of 5 million and 50 billion metric tones of conventional materials when accounted for their size [22]. Figure 5 presents the results of midpoint indicators for the impact category of marine aquatic ecotoxicity potential. It is observed that traditional materials especially aluminum seems to have a higher environmental burden in this category when compared with CNFs. Higher ecotoxicity of aluminum can be attributed to the emissions of a variety of heavy metals with higher ecotoxicity during ore extraction and processing. However, the impact numbers for CNFs represents only lower bounds whereas those for traditional materials are more precise since detailed and relatively more complete inventory data is available in the literature for traditional materials. On the other hand, the emissions and hence their impact from the CNF synthesis step are not accounted. In addition, the emissions from the catalyst life cycle have been ignored due to lack of data. More importantly, *release and impact of CNFs* on humans and ecosystem species during manufacturing are not accounted. Studies exist in the literature that present evidence of size-dependent hazardous and toxic effect of CNFs [23].

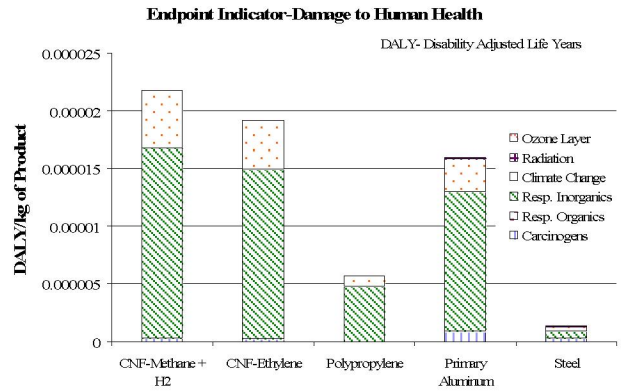

Fig. 6. Endpoint Indicator- Damage to Human Health

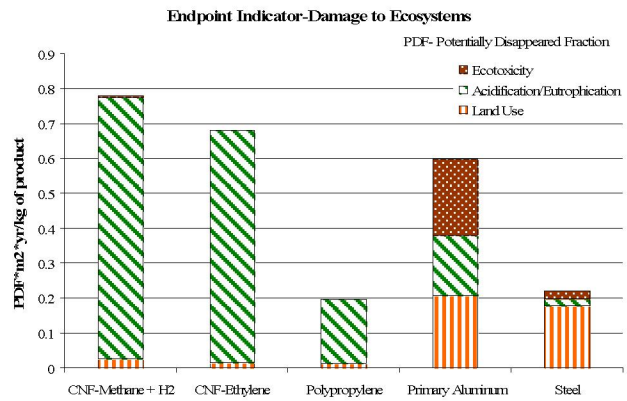

Fig. 7. Endpoint Indicator- Damage to Ecosystems

Figure 6 and 7 present the Endpoint Indicators that are obtained based on Eco-indicator 99. These are obtained directly from SimaPro. These figures indicate higher impact of CNFs in the category of damage to human health and damage to resource consumption compared with traditional materials on an equal mass basis. As observed for midpoint indicator, Figure 7 shows a higher ecotoxicity impact of aluminum on ecosystems as compared to CNFs. However, the overall impact of CNFs in the category of Damage to ecosystems is higher based on both methane and ethylene feedstocks. This is because the damage caused to ecosystems because of higher acidification and eutrophication potential for CNFs outweighs the higher ecotoxicity impact because of aluminum. It is important to reiterate that human and ecosystem impact of CNFs emitted to the environment is not accounted due to lack of information about the human and ecotoxicological impact of these engineered nanoparticles. Detailed knowledge and quantifiable data about possible fate, transport and mechanism of damage of CNFs is not available. Thus, the impact numbers presented

here reflect only the material and energy use during the synthesis of CNFs.

VI. CONCLUSIONS AND FUTURE WORK

A Life Cycle Energy Analysis of CNF synthesis is presented and compared with traditional materials on an equal mass basis. Preliminary results indicate 6-60 times higher life cycle energy requirements for CNFs when compared with aluminum, steel and polypropylene. High energy requirements and hence the resulting high cost of these engineered nanomaterials will tend to restrict their use only in very niche applications and hinder their use for large volume applications like polymer composites. Research efforts are required to increase the process yields and optimize the growth of CNFs so that the novel properties of these ENPs can be realized to yield products with superior properties. A preliminary "cradle-to-gate" LCA of Carbon Nanofiber synthesis is also presented. Midpoint and Endpoint Impact Assessment methods suggest that CNFs may impose a higher environmental burden as compared to traditional materials per kg of product. Results indicate a lower ecotoxicity impact of CNFs compared with traditional materials. However, the human and ecotoxicological impact of CNFs themselves are not accounted for due to the lack of information about the release and impact of CNFs. The comparisons of CNFs with traditional materials are presented on an equal mass basis with traditional material to get insight into the life cycle energy intensities and environmental burden. However, products based on CNFs might be greener than alternatives for a given application. Quantity will be the deciding factor and hence specific nanoproducts and applications need to be evaluated.

Future Work is expected to use these results to evaluate claims about the potential benefits of specific CNF based nanoproducts and nanoprocesses. One of the potential applications is the use of Carbon Nanofiber (CNF) Reinforced Polymer Composites in the body panels of automobiles as an alternative to traditional materials like aluminum and steel. Statistical techniques will also be useful for addressing the uncertainty in the Life Cycle Inventory data. These studies also ignore the possible emissions and impact of nanoparticles, and thermodynamic LCA methods are also being explored as proxy indicators in the absence of emissions and impact information. In the long term, integration of this work with the detailed fate and transport studies of these nanomaterials can provide useful insights about the ecotoxicity of these nanomaterials.

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