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ENVIRONMENTAL COMPARISON OF SOL-GEL *vs*. CONVENTIONAL PAD-DRY-CURE FINISHING PROCESSES FOR ANTIBACTERIAL TEXTILES

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Abstract

In the past decades, increasing environmental concerns have forced the textile industry to search for alternative processes that increase production efficiency while reducing cost, resource consumption, and waste generation. On the other hand, sustainable consumption and production are still problematic issues in the textile value chain. From a life cycle perspective, antibacterial applications are promising due to their potential to reduce environmental impacts of the textiles. In this study, sol-gel and pad-dry-cure application techniques were used to obtain antibacterial cotton textiles and they were compared in terms of water, chemical and energy consumption. Environmental impacts of antibacterial application techniques were evaluated with life cycle analysis method using GaBi 6.0 software and database. The results showed that drying is the most important process that contributes to the overall environmental impact categories. Findings also revealed that chemicals constitute an important part of environmental impacts. The sol-gel method offers a comparatively better environmental profile in most of the impact categories studied and provides a reduction in resource and energy consumption. The findings of this study is important and may help decision-makers to choose alternative sustainable practices for antibacterial applications within the textile industry.

Keywords: antibacterial textiles, life cycle analysis, pad-dry-cure, sol-gel technique, sustainability

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1. Introduction

In recent decades, a massive increase in per capita consumption of textiles was observed globally as a consequence of growing number of the world population and increasing economic prosperity in developing countries (Ekstrom and Salomonson, 2014; Kazan et al., 2020; Manda et al., 2015; Terinte et al., 2014). Retail strategies, such as fast fashion and change of consumer behaviors also have significant influence on increasing consumption tendency (Bhardwaj and Fairhurst, 2010; Fletcher, 2014; Pookulangara and Shephard, 2013; Tokatli, 2008). The fast-fashion system is convenient for cheap, easy, and fast production in high volumes. In this system, fashion goods have quick renewal cycles and quite low prices, which is highly attractive to consumers and encourages them to purchase more goods that they do not need (Fletcher, 2010; Folligne, 2020). This trend is especially criticized due to its great environmental and social impacts emerged from resource consumption and waste production (Bick et al., 2018; Gwozdz et al., 2017; Remy et al., 2016). The fact that conventional textile industry has already been an intensive consumer of chemicals, water and electricity, in addition to production and consumption within such a fast cycle, has led to a higher rate of depletion of natural resources and increased emissions (Piontek and Müller, 2018; Roos et al., 2018). As a result of emerging disposable clothing culture (Bick et al., 2018), the disposal problems exacerbated due to the vast amount of textile waste generation (Birtwistle

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and Moore, 2007; Kozlowski et al., 2012). However, it is important to notice that the rapid replacement of garments is both associated with consumer behaviors and technical and quality issues related to garment texture, which often fails to tolerate multiple washing cycles (Fletcher, 2010; SIFO, 2001).

Today, numerous studies confirmed that use phase is responsible for considerable part of textiles' life cycle impact (Cotton Inc., 2017; Defra 2007; Manda et al., 2015; Walser et al., 2011; Windler et al., 2013). The main reason for that, especially for cotton fabrics, is high amounts of water, chemical and energy consumption during washing (Cotton Inc., 2017; Defra 2007). Damage on textile fibers caused by mechanical action, heat and detergents during the washing process is another factor that has a negative impact on the durability of fabrics (Laitala et al., 2011). In this context, antibacterial applications offer an opportunity to prolong the durability of textiles by reducing washing frequency, and cleaning efforts and costs (Hicks and Theis, 2017; Manda et al., 2015; Periyasamy et al., 2020; Windler et al., 2013). Although it is not applicable to completely replace washing processes with antibacterial applications, it can be one of the possible ways of improving sustainability within the field of textile and clothing.

In recent years, textile and clothing sector have started to focus more on sustainable products and processes to meet environmental and social requirements as well as to acquire competitive advantage in the market. Numerous antibacterial finishing methods were suggested to reduce water, chemical, energy consumption and cost (Aksit et al., 2017; Borda d'Água et al., 2018; MNT-ERANET, 2013). In this respect, the sol-gel method is promising in terms of reducing resource usage and obtaining effective results (Ismail, 2016; Periyasamy et al., 2020). Although it can be considered as a sustainable alternative process to conduct conventional pad-dry-cure practices, the environmental impacts of the sol-gel method have not been discussed widely in the literature (Unvar et al., 2018).

Our study aims to provide inventory data regarding the sol-gel antibacterial finishing process and to compare the environmental performance of sol-gel antibacterial treatment and conventional treatment using life cycle analysis (LCA).

2. Material and methods

LCA, as described in international standard ISO 14040, consists of four sequential steps: goal and scope definition, inventory analysis, impact assessment and interpretation. These four steps are described in relation to the case study. The LCA study was carried out using the GaBi 6 Academy Software (Thinkstep, Germany). The environmental impacts were obtained using factors of Centrum voor Milieukunde Leiden (CML) developed by the Leiden University Centre of Environmental Science, which were updated in 2001.

2.1. Goal and scope definition

The main objective of this LCA study was to conduct a comparative study to measure and evaluate the environmental performance of sol-gel treated antibacterial fabrics and those treated with conventional pad-dry-cure process, and to provide further insight to stakeholders and academics or industry related to the sol-gel process. Scoured and bleached 100% cotton fabric (120 g/m^2), also referred as untreated fabric, was used to obtain antibacterial fabrics. The functional unit (FU) was chosen as "production of 1 m^2 antibacterial cotton fabric from untreated fabric".

Inventory data based on the sol-gel antibacterial recipe were obtained from a previously published study (Aksit et al., 2017) and a project (MNT-ERANET, 2013). The inventory data for the conventional antibacterial method were obtained from existing practices of a company. Nanosol used in the sol-gel antibacterial recipe consists of titanium isopropoxide (TIP) and silver nitrate ($AgNO_3$) as precursors; water and isopropanol (IP) as co-solvents, hexadecyltrimethylammonium bromide (HTDA), hydrazine as reducing agent and acetic acid (AA). The conventional recipe is composed of 3-(tri-methoxysilyl) propyldimethyl octadecyl ammonium chloride, SiQAc a silane quaternary ammonium-based chemical as an antibacterial agent, water as a solvent, softener and acetic acid.

Production processes for certain chemicals were not included due to negligible application rates and lack of process inventory data in the Gabi Academy database. Regarding SiQAc, which is used in the conventional recipe, data obtained from silicone production was used as representative. The data on acetic acid, isopropanol, and silicon production processes were obtained from GaBi 6 Academy database. Flow charts of the both processes involve the following steps; (i) antibacterial application using dip-coating for sol-gel technique and padding for conventional method (ii) drying at 100 °C for sol-gel technique and 130 °C for conventional method and (iii) fixation at 150 °C for both methods. System boundaries for methods were identical and given in Fig. 1.

2.2. Life cycle inventory analysis

In the inventory analysis, calculations were carried out to measure the relevant inputs and outputs of the antibacterial cotton fabric. Mass inputs were calculated considering the pick-up rate, which indicates the percentage of liquid absorbed by untreated fabric based on its dry weight. The total required amount of substance (T_M) for untreated cotton fabric was calculated with Eq. (1) where W_U is the weight of 1 m^2 dry untreated fabric (120 g/m^2) and P is the pick-up rate.

$$T_M = P \times W_U \tag{1}$$

Pick-up rate is determined by adjusting squeezing pressure of foulard machine and velocity that emerges after antibacterial application. According to the data obtained for inventory analysis, antibacterial applications were conducted at pick-up rate of 80% via proper adjustment of squeezing rollers. Following Eq. (1), T_M (96 g/FU) was found identical in both methods using the same fabric and pick-up rate. It should be noted that in both cases, the quantity of total antibacterial solution applied on fabrics was more than T_M however, excessive amounts of antibacterial solutions were recycled after the squeezing process to reuse in the next batch. Consequently, in the inventory analysis, only the total required amount of substance was considered for calculations. Inventory data for the required amount of chemicals and water input per functional unit for each antibacterial application were given in Table 1.

Due to lack of data on specific energy use for conventional pad-dry-cure antibacterial treatment performed in the connected production line, the inventory data on energy use for both methods were calculated theoretically. Energy consumption regarding drying and fixation processes were included in the inventory analysis, taking into consideration the domination of heat treatments over the whole process flow. The Eqs. (2-5) were used to calculate energy requirements of drying and fixation processes for both antibacterial treatments.

$$Q_{textile-drying} = m_c \times C_{p,t} \times \left(T_{out,druing} - T_{in,drying}\right) \quad (2)$$

$$Q_{solvent} = m_s \times C_{p,s\,25^0\,C} \times \left(T_{vaporization} - T_{in,drying}\right) \quad (3)$$

$$Q_{solvent\_evap} = m_s \times \Delta h_s \quad (4)$$

$$Q_{textile-fixation} = m_t \times C_{p,t} \times \left(T_{out,fixation} - T_{in,fixation}\right) \quad (5)$$

The equations refer to required energy amount calculations throughout the drying and fixation processes in the given order: (2) Heating the textile at 25°C ($T_{in,\ drying}$) until it reaches drying temperature ($T_{out,\ drying}$); (3) Heating the solvent in the textile at $T_{in,\ drying}$ until its vaporization temperature ($T_{vaporization}$); (4) Evaporation of solvent; (5) Heating the textile at $T_{out,\ drying}$ (which equals to the temperature of $T_{in,\ fixation}$) until it reaches fixation temperature ($T_{out,\ fixation}$). The mass of textile and solvent (water or water and isopropanol depending on the method) were denoted as m_t and m_s, respectively. Specific heat capacities for textile ($C_{p,t}$), and solvent ($C_{p,s}$) and enthalpy changes for solvents (Δh_s) at given equations were obtained from the literature (Andon et al., 1963; Majer et al., 1985; Matthews, 1947; Wagner and Kretzschmar, 2007) and given in Table 2.

According to the obtained data, the drying temperature was 100 °C for the sol-gel process and 130 °C for the conventional pad-dry-cure method, while the fixation temperature was 150 °C for both methods.

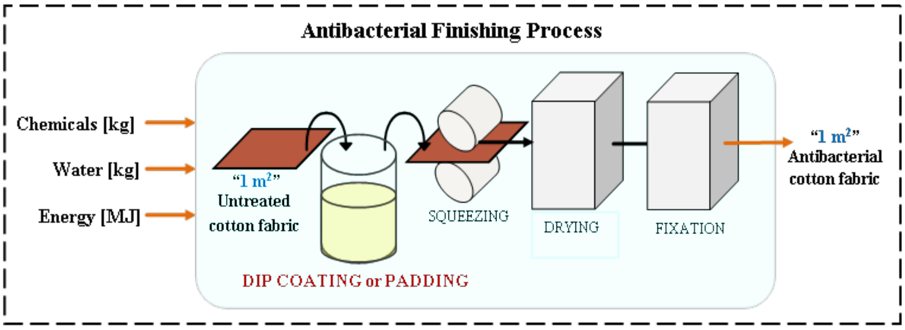

Fig. 1. System boundaries for sol-gel (dip coating) and conventional pad-dry-cure (padding) antibacterial fabric production methods

Table 1. Inventory data for sol-gel and conventional pad-dry-cure method per functional unit

Sol-gel treatment				*Conventional pad-dry-cure treatment*			
	T_M (g/FU)	*Amount (g)*	*Mass (%)*		*T_M (g/FU)*	*Amount (g)*	*Mass (%)*
Water	67.8	30.0	70.6	**Water**	88.9	1000	92.6
Chemicals				**Chemicals**			
$AgNO_3$	0.2	0.1	0.2	SiQAc	4.5	50	4.6
IP	23.1	10.2	24.1	Softener	2.2	25	2.3
TIP	1.7	0.8	1.8	AA	0.4	5	0.5
AA	1.2	0.5	1.2				
HDTA	1.6	0.7	1.7				
Hydrazine	0.3	0.1	0.3				
Total	**96.0**	**42.5**	**100**	**Total**	**96.0**	**1080**	**100**

Table 2. Specific heat capacities and enthalpy changes at (2) to (5) equations

Specific heat capacities C_p (kJ/kg.K)			*Enthalpy changes Δh_s (kJ/kg)*	
$C_{p,t}$ (Textile)	$C_{p,s\ 25°C}$ (Water)	$C_{p,s\ 25°C}$ (Isopropanol)	Δh_s (Water)	Δh_s (Isopropanol)
1.3356	4.1822	2.5697	2256.54	663.12

It was assumed that the evaporation process of solvents was completed during the drying process due to a very slight change in the mass of fabric before and after fixation. On the other hand, Eqs. (3) and (4) were used for complete evaporation of solvents during the drying process of both treatments and the vaporization of other species was neglected. Consequently, energy data for the processes were calculated as given in Table 3 and it was assumed to be supplied by Turkey's electricity grid mix.

In industrial operations, heat treatments mostly result in higher energy demands due to heat loss, machinery efficiency, etc. However, the ratios between methods in terms of energy requirements will presumably oscillate in a narrow range. Thus, considering the relative contributions of methods, it is important to note that outputs of this study may be assessed on a common ground and adapted for commercial or different kinds of scales.

3. Life cycle impact assessment results and discussion

The life cycle impact assessment was carried out up to characterization step. The potential environmental impacts were calculated using CML 2001 characterization factors. The impact categories considered in the impact assessment phase included Abiotic Depletion Potential (ADP), Acidification Potential (AP), Eutrophication Potential (EP), Freshwater Aquatic Ecotoxicity Potential (FAETP), Global Warming Potential (GWP, excl. Biogenic carbon, 100 years), Human Toxicity Potential (HTP), Ozone Layer Depletion Potential (ODP), Photochemical Ozone Creation Potential (POCP) and Terrestric Ecotoxicity Potential (TETP).

The impact assessment results for each impact category regarding the materials and sub-processes involved in both sol-gel (SG) and conventional pad-dry-cure (C) antibacterial application systems were given in Fig. 2 and Fig. 3.

The contribution of the drying process to most of the impact categories was similar in both systems and each system was influenced considerably by energy consumption in this process (Figs. 2a, 2b, and Fig. 3). The highest contribution to acidification potential derived from electricity consumption, and consequently all processes involved in heat treatments showed higher contribution to this category (Fig. 3a).

On the other hand, the share of electricity consumption in drying and fixation processes provided the highest share in acidification potential in both systems as 89% for the sol-gel process and 83% for the conventional pad-dry-cure process (Fig. 4), and a similar tendency was also observed in eutrophication potential. The usage of electricity for heat treatments also has a considerable influence on environmental impacts, which is consistent with the previously published data (Manda et al., 2015). Regarding other categories such as GWP, HTP, POCP, and TETP, the contribution of electricity usage varies between 50-80% for sol-gel and 30-75% for the conventional method (Fig. 4).

These findings reveal that electricity usage has a considerable contribution to a wide range of impact categories in both systems, mainly due to electricity production processes. It is known that environmental impacts of electricity usage vary depending on natural resources, material and technology used in electricity generation, geography, import-export of electricity etc. (Colett et al., 2016; Hertwich et al., 2015; Masanet et al., 2013; Rusu et al., 2018).

Table 3. Heating energy data for the sol-gel method and conventional pad-dry-cure method

	Drying (MJ)			*Fixation (MJ)*	*Total (MJ)*
	$Q_{textile}$	Q_{water}	$Q_{isopropanol}$	$Q_{textile}$	
Sol-gel Method	0.0120	0.1743	0.0187	0.0080	**0.2130**
Conventional pad-dry-cure treatment	0.0168	0.2285	-	0.0032	**0.2485**

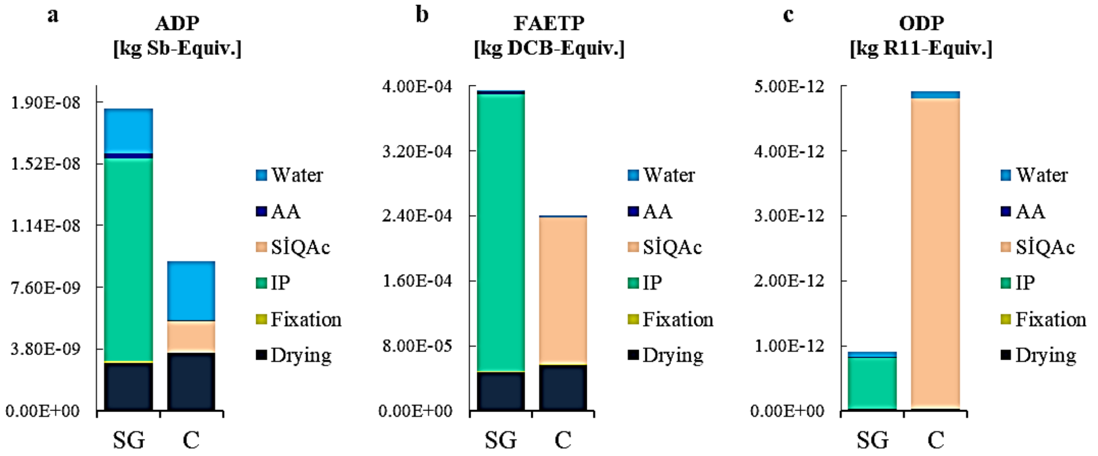

Fig. 2. Impact assessment of sol-gel (SG) and conventional pad-dry-cure (C) processes for antibacterial finishing of cotton fabrics: (a) ADP, (b) FAETP and (c) ODP

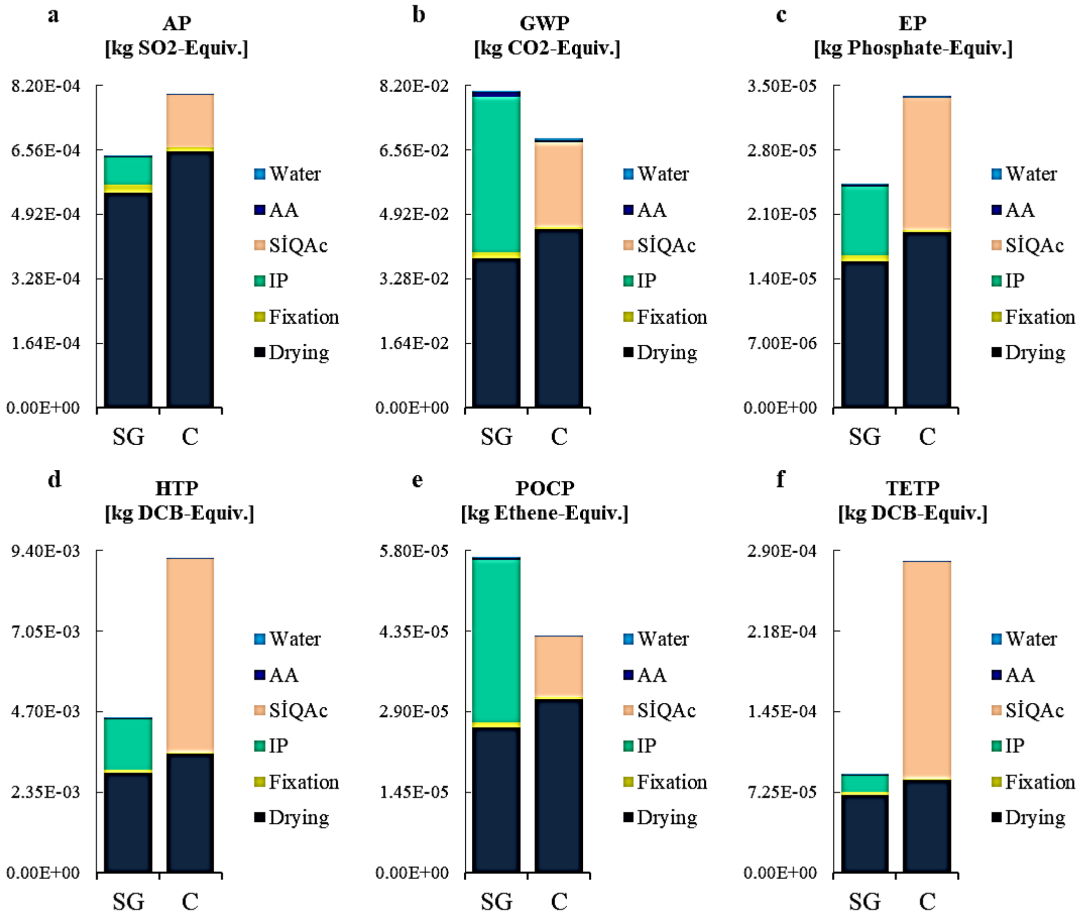

Fig. 3. Impact assessment of sol-gel (SG) and conventional pad-dry-cure (C) processes for antibacterial finishing of cotton fabrics: (a) AP, (b) GWP, (c) EP, (d) HTP, (e) POCP and (f) TETP

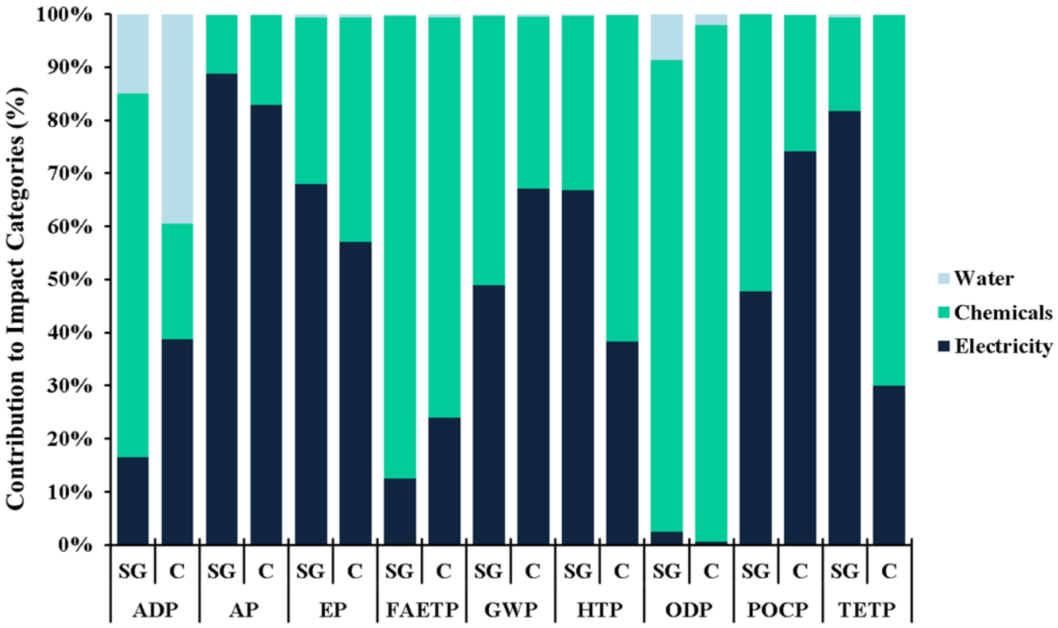

Fig. 4. The relative contribution of process inputs to the impact categories

Turkey's electricity grid mix is based on fossil fuels such as hard coal and natural gas (Kılıç et al., 2018), which results in generation of a variety of emissions and has a considerable contribution to environmental impact categories (Atilgan and Azapagic, 2015, 2016a, 2016b; Günkaya et al., 2016). The highest contribution of water usage is 40% and 15% to the ADP impact category in conventional pad-dry-cure and sol-gel treatment, respectively (Fig. 4). The sol-gel process provided a 24% reduction in this category due to lower water usage compared to conventional pad-dry-cure finishing. As seen in Fig. 4, it was observed that chemicals used in conventional pad-dry-cure treatment contributed to HTP (62%), TETP (70%), FAETP (76%), and ODP (97%) mainly due to silicon production processes.

Production processes of acetic acid did not result in substantial impact in both systems and represented a 2% overall contribution at most.

In sol-gel process, chemicals appeared to be the major contributor in GWP (51%), POCP (52%), ADP (69%), FAETP (87%) and ODP (89%) categories. The contribution to ADP impact category, which is related to the depletion of natural resources, was dominated by the production process of isopropanol used in the sol-gel process. FAETP, GWP, and POCP were the other categories, where the sol-gel system had higher impacts, mainly due to isopropanol use. Isopropanol has greater upstream production demands, due to complication and number of its production processes (Tsang et al., 2016).

Based on the utilized database, production of isopropanol was carried out with propene via indirect hydrogenation process, which results in wastewater discharge with sulfuric acid and caustic soda (Panjapakkul and El-Halwagi, 2018). Consequently, the remarkable contribution of isopropanol use in ADP (67%) and FAETP (86%) may be attributed to high resource and energy use in the production phase and disposal phases of this solvent (Hellweg et al., 2004). The impact of the conventional pad-dry-cure system is higher in AP, EP, HTP, ODP and TETP categories in comparison to the sol-gel system. This is mainly due to silicon production processes, which demand high electricity in its production processes (Tveit et al., 2004), and high electricity use in the drying process. In the conventional system, the highest contribution of silicon production processes to impact categories include ODP, FAETP and TETP account for 97%, 75%, and 70%, respectively.

The solvents affect both systems in terms of material input and their functions within the system, which could be supportive or aggravative. Although isopropanol, as a chemical input, exacerbated the impacts of the sol-gel system at the ADP, FAETP, GWP, and POCP categories, it also provided a reduction in water use compared to the conventional system. In addition, the sol-gel process has enabled a 16% reduction in energy consumption during drying in comparison to conventional finishing (depending on less amount of water needed to be evaporated from the fabric). Consequently, the sol-gel system presented a better environmental profile by contributing 14% less than the conventional system in all impacts caused by the use of energy.

The comparative assessment of sol-gel and the conventional process was summarized in Fig. 5 covering all impact categories. Sol-gel process has comparatively higher impacts than conventional process in ADP (102%), FAETP (64%), GWP (18%) and POCP (33%). The main reason for this outcome is the usage of isopropanol in sol-gel antibacterial treatment. On the other hand, the sol-gel process presented a better environmental performance and decreased contribution to AP, EP, HTP, ODP and TETP by 20%, 28%, 51%, 81% and 68%, respectively, in comparison to conventional finishing. The reason for this improvement is mainly due to the production processes for the silicon (considered as SiQAc representative), and higher energy demand for drying in the conventional system.

Consequently, chemicals were found to constitute a remarkable environmental load, due to their production processes. Regarding heat treatments, improving energy efficiency in the drying process, utilization of alternative renewable energy sources, and reduction of the amount of solvent would be reasonable improvement options to reduce the energy-related environmental impacts in these production stages.

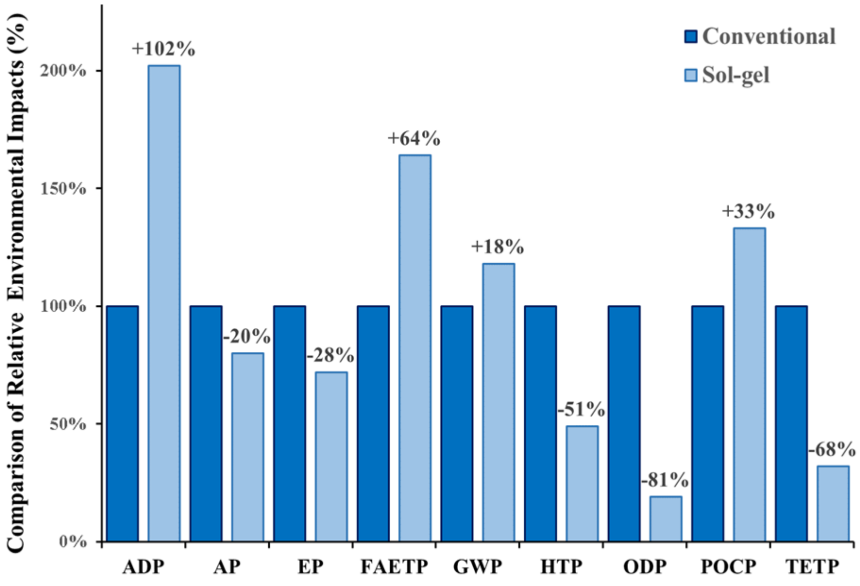

Fig. 5. Comparison of relative environmental impacts between conventional pad-dry-cure and sol-gel antibacterial finishing

Considering identified hotspots regarding chemicals within this study, replacement of organic solvents with greener alternatives and improving the efficiency of the process, which is one of the significant contributors to environmental profile of production system (Petraru and Gavrilescu, 2010), is strongly recommended to avoid environmental impacts derived from production processes of conventional solvents.

4. Conclusions

In this study, the results obtained from comparative environmental assessment of sol-gel and conventional pad-dry-cure antimicrobial finishing process were presented. Results reveal that energy consumption at drying processes and chemicals are the environmental hotspots of the antibacterial finishing processes and have a great contribution to the total environmental performance of both systems.

The sol-gel method showed better environmental performance in more than half of the environmental impact categories and provided a reduction up to 81% at related categories in comparison to the conventional pad-dry-cure process. Isopropanol supported the sol-gel system as co-solvent and led to the reduction of water and energy consumption. However, due to the production processes of this chemical, it increased environmental contribution of sol-gel process to overall impact categories.

The sol-gel method enables the production of antibacterial cotton fabric with less energy, water, and chemical consumption, and this, in turn, benefits to environmental performance and efficiency of the process. Although we observe promising improvements in sol-gel technology, there are some issues that should be taken into consideration. For instance, cost is the foremost issue, which limits the expansion of sol-gel method to mass production due to mainly expensive precursor materials. On the other hand, industrial viability of sol-gel technology is more feasible thanks to low-temperature treatment, easy application in textile mills and no requirement for special equipment. Today, industrial acceptance for advanced technologies is tightly coupled with cost and viability as well as environmental performance. Therefore, in near future, sol-gel technology will find its way in textile industry, through further research on reduction of its cost by optimized process conditions, and search for precursors with moderate prices due to its lower environmental impacts.

The results obtained from this study, highlight improvement opportunities for researchers dealing with sol-gel process and further life cycle assessment practitioners. Although the recent paper contributes to the goal of providing inventory data for the sol-gel process applied for antibacterial textile for the first time, data collection should be expanded in future studies by gathering data on energy consumption and production process for the specific chemicals used in sol-gel method.

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