APPLICATION OF THE PRINCIPLES OF GREEN CHEMISTRY IN THE PLASTIC RECYCLING INDUSTRY: A CASE STUDY

ORIGINAL SCIENTIFIC PAPER

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ABSTRACT:
The subject of this research was to verify the feasibility of implementing green chemistry principles within the business company „Omorika Reciklaža“ Ltd. situated in Johovac near Doboj, Bosnia and Herzegovina. The objective of this study was to conduct a detailed assessment of the company's facilities and operations, using environmental audits, to identify technological processes (production lines), energy and waste flows, capacities, product range, and other pertinent factors crucial for the application of green chemistry principles. Special emphasis was placed on analyzing each substance that constitutes a raw material, whether used individually or as part of mixtures. The assessment of safety data sheets involved utilizing the CAS registration numbers of substances from the Chemical Abstracts Service, cross-referenced with the ECHA database (European Chemicals Agency). The outcomes, attained through an exhaustive analysis of each substance, were presented as a "chemical inspection" of the company. Through the analysis of all substances and mixtures in the technological process (chemical inspection), as well as the capacity of production flows, energy and resource flows, wastewater, and waste, the potential for enhancing the technological process was identified. This involved reducing dust levels in the workspace, decreasing electricity consumption (utilizing renewable sources), and substituting particularly hazardous chemicals used in the technological process.

KEYWORDS: green chemistry; chemical safety; SVHC; environmental audit

INTRODUCTION

Green chemistry, according to the definition popularized by Paul T. Anastas, one of the pioneers in the field of green chemistry, is the design of chemical products and processes that reduce or eliminate the use and generation of hazardous substances [1]. Green chemistry applies across the life cycle of a chemical product, including its design, manufacture, use, and ultimate disposal. Green chemistry, or the 12 principles of green chemistry, encompasses techniques from chemistry, engineering, and many other scientific fields, and has spurred entirely new research [2]. The principles of green chemistry can be applied in various fields and industries to enhance sustainability and reduce negative impacts on the environment.

According to the European Association, Plastics Europe AISBL, in 2021, 90.2% of the world's plastic production was fossil-based. Post-consumer recycled plastics and bioplastics accounted for 8.3% and 1.5% of the world's plastic production, respectively [3]. From the mentioned data, the importance of plastic recycling and its positive impact on the environment is evident. However, the industrial plastic recycling process itself can be further improved to have a lesser environmental impact. There are many ways in which the principles of green chemistry can be applied in industry to reduce the negative impact on the environment and human health while simultaneously improving the efficiency of production processes.

On the other hand, according to the World Health Organization (WHO), chemical safety is an approach that encompasses all activities aimed at protecting people and the environment from the harmful effects of chemicals, whether of natural origin or manufactured [4]. This includes measures to reduce human and animal exposure to harmful chemicals, as well as measures to mitigate the damage that can be caused by incidents involving hazardous materials. The goal of chemical safety is to reduce the risk of adverse effects of chemicals on human health and the environment. To apply the principles of green chemistry in an industrial facility, it is necessary to be familiar with the basic principles of green chemistry. In addition to the principles of green chemistry, knowledge in the field of chemical management (chemical safety) is also essential.

Chemical safety in Bosnia and Herzegovina (BiH) is under the jurisdiction of the entities (Republika Srpska and the Federation of Bosnia and Herzegovina) and the Brčko District of Bosnia and Herzegovina, except in the area of reporting to the European Commission (in accordance with the obligations of the Stabilization and Association Agreement) and customs policies, including the control of the import and export of goods subject to special regimes in trade. Regulations governing chemical safety in BiH are not harmonized, and the adoption of EU regulations in this area is not coordinated. As a consequence, different regulations are applied in BiH, and legal entities operating in the entire country do not have equal status.

In October 2020, the Federation of BiH adopted the Law on chemicals [5], which, among other things, aligns the Federation of BiH with EU regulations governing the field of chemicals. However, to this point, Law on biocidal products has not been adopted, and the subordinate regulations foreseen by the Law on chemicals have not been adopted. Therefore, the transposition of the mentioned EU legislation and conventions is at a low level, which has been recognized as the biggest challenge for the institutions of the Federation of Bosnia and Herzegovina in this area. In the Republika Srpska, there is clear legislation regarding chemical management, stemming from the Law on chemicals [6] and the Law on biocidal products [7], under the jurisdiction of the Ministry of Health and Social Welfare of Republika Srpska. The existing regulations governing chemical safety in Republika Srpska are generally, to a greater or lesser extent, aligned with certain conventions (of which BiH is a signatory) or directives and regulations of the European Union (EU).

EU legislation defines centralized procedures within the EU, which are applicable only to member states. Such provisions cannot be transposed into the legislation of entities and the Brčko District of BiH; instead, adjustments and preparatory actions have been undertaken for their future implementation. In 2006, the EU introduced the Registration, Evaluation, Authorization and Restriction of Chemicals (REACH) regulation [8], which requires companies to provide data showing that their products are safe. This regulation ensures not only the assessment of the hazards of chemicals as well as the risks during their use, but also includes measures to ban or restrict/authorize the use of certain substances [9]. The REACH chemicals regulation is considered a very powerful promoter of sustainable innovation and green chemistry. REACH favors innovative new materials and processes by allowing exemptions from registration for five years for substances used in research and development. REACH is a tool used by the EU to encourage the transition to "green chemistry" in line with environmental policies. The Law on chemicals of the Republika Srpska [6] and the Law on chemicals of the Federation of Bosnia and Herzegovina [5] are harmonized with the REACH Regulation. REACH is currently in the process of revision and changes to the existing REACH regulation are expected. One of the most significant results of REACH is the establishment of the European Chemicals Agency (ECHA).

From the above, it is clear that green chemistry and chemical safety are interconnected as they both ensure human health and environmental protection. Legislation in the European Union (EU) plays a crucial role in this field.

The aim of this research was to analyze the production process, applied technologies, waste streams (wastewater, solid waste, waste gases, etc.), raw materials, chemicals and energy flows. A special aspect is devoted to the analysis of raw materials and chemicals, which includes a detailed analysis of safety data sheets (SDS) to identify hazardous chemicals and assess substances contained in given mixtures (chemicals). The research was conducted at the company "Omorika Reciklaža" Ltd. Johovac, Doboj.

MATERIALS AND METHODS

The application of green chemistry begins with an understanding of the 12 principles of green chemistry [2]. These principles form the basis for the development of green chemistry, which is aimed at designing chemical products and processes that are sustainable, less harmful and contribute to environmental protection. In practice, the production process or its stages can rarely satisfy all 12 principles of green chemistry, at least not completely. Sometimes it is one principle, three or four, but it is important to have them as goals to strive for [2].

The application of some of the principles of green chemistry involves a detailed analysis of the production process, i.e., an environmental audit. An environmental audit is a systematic evaluation of the environmental impact of an organization's operations. In this case, the environmental audit is tailored and conducted to assess the feasibility of implementing green chemistry principles in the company "Omorika Reciklaža" Ltd. An environmental audit can be considered a key tool for implementing green chemistry principles in a business, and by conducting an environmental audit, the company can assess its impact on the environment in accordance with green chemistry principles. The final result of the environmental audit is a report that contains information about the organization's impact on the environment, identified areas for improvement, as well as recommendations (measures) for the implementation of green chemistry principles.

These measures can encompass the establishment of new policies and procedures related to chemical management, recycling, and reducing the use of harmful chemicals and materials, the utilization of renewable energy sources, as well as waste reduction and greenhouse gas emissions reduction. However, this paper will not present measures concerning energy sources and energy flows. "Energy measures" will be presented in a separate research.

As previously mentioned, special attention is devoted to the analysis of raw materials and chemicals. According to the Law on chemicals, a chemical means a substance and a mixture containing a substance. A substance means a chemical element and its compounds in the natural state or obtained by any manufacturing process, including any additive necessary to preserve its stability and any impurity deriving from the process used, but excluding any solvent which may be separated without affecting the stability of the substance or changing its composition. While mixture means a mixture or solution composed of two or more substances and a hazardous chemical is a chemical classified within at least one hazard classes [6].

As part of the environmental audit, a so-called "chemical inspection" was conducted, during which SDSs for twenty-six chemicals (mixtures) were thoroughly analyzed. The assessment of SDSs was conducted using the Chemical Abstracts Service (CAS) numbers of substances that are part of each mixture and the ECHA`s database [11]. Each substance included in the composition of the mixtures used in this company was analyzed. By entering the CAS number or name of the substance into ECHA's database, basic data on the substance (*Substance Infocard*) containing *Substance identity*, *Hazard classification & labeling* and *Properties of concern* were obtained. Also, it is possible to view the *Brief profile* of the substance, where the classification of the substance is specified. ECHA uses a system of chemical classification and labeling based on the EU CLP (Classification, Labelling, and Packaging) regulation [12]. This labeling system utilizes several key elements to clearly and precisely identify the hazards and properties of chemicals.

There are the basic labels used according to the CLP regulation. Pictograms are visual icons that depict hazards associated with chemicals. Examples of pictograms include a flame symbol for flammable chemicals, a skull and crossbones for substances that are harmful or toxic, and others. Hazard statements ("H numbers") include words such as "Flammable," "Carcinogenic," "Toxic," "Irritant," and others, describing specific hazards associated with chemicals. For example, "H225" corresponds to the Hazard Statement "Highly flammable liquid and vapor". Each chemical has an associated Signal Word that emphasizes the severity of the hazard. Examples include "Danger" or "Warning." Precautionary statements provide additional information about hazards and precautionary measures to be taken when handling the chemical. This includes information on safe handling, storage, and disposal (for example, precautionary statement “P264”: Wash hands thoroughly after handling). Chemicals are categorized into various Hazard Classes and Categories based on their properties. Examples of hazard classes include "Flammable," "Health Hazard," and "Environmental Hazard".

ECHA, in accordance with the CLP [13], publishes and regularly updates the list of classified and labeled chemicals - CL Inventory [14]. This database contains information on the classification and labeling of substances that have been submitted by manufacturers and importers in the EU. This information is valuable for chemical users as well as regulatory bodies enforcing chemical regulations to ensure the safe use of chemicals in the EU and protect the human health and the environment. The CL Inventory is implemented in the Republic of Srpska through the Rulebook on Chemical Inventory [15]. As of September 25, 2023, there are 6.782 chemicals listed in the Chemical Inventory [16], while there are 227.204 chemicals listed on the European CL Inventory [17].

Substances that can have serious consequences for human health and the environment can be identified as Substances of Very High Concern (SVHC). The list of SVHC substances is maintained by the ECHA and is regularly updated to include new substances identified as SVHC. Due to their potentially harmful impact on health and the environment, SVHC substances are subject to strict regulation in the EU, and their use may be restricted or prohibited. To this end, REACH introduces, among other instruments, an authorization regime for SVHC substances listed in Annex XIV of the Regulation [8].

In 2008, ECHA recorded its first chemical that could potentially harm human health and the environment. This led to the creation of two lists, the Authorization List [18] and the Candidate List [19], which include all hazardous chemicals. To be placed on the market, specific conditions of use must be prescribed for these chemicals. The goal of these procedures is to reduce the presence of SVHCs in products and transition to safer alternatives. The Candidate List has been transposed into the legislation of the Republic of Srpska as the List of substances that are candidates for inclusion in the list of SVHC (Candidate List) and contains 209 substances (updated on December 22, 2020) [20], while the ECHA Candidate List of SVHC now contains 235 entries [19]. The Law on Chemicals [5, 6] stipulates that the supplier is obliged to provide any distributor or user with information about a particular product if it contains a SVHC in a concentration greater than 0.1%. The information should include the name of the substance that the product contains and should be voluntary for the safe use of that product.

Substances listed on the Candidate List can indeed move to the Authorization List (list for issuing approval) according to Annex XIV of REACH [8]. This means that, after a specified date, companies will not be allowed to place SVHC on the market or use them unless they have been specifically authorized to do so. One of the primary goals of authorization is to gradually phase out SVHCs wherever possible [21]. The Authorization List was transposed into the legislation of the Republic of Srpska through the Rulebook on restrictions and bans of chemicals [22]. The list of substances of particular concern includes 43 substances [23], as of September 25, 2023, while 59 substances are on the Authorization List.

ECHA also provides a database (list) of substances classified as persistent organic pollutants (POPs) – "List of substances subject to the POPs Regulation" and chemicals under preparation for their potential inclusion as POPs – "List of substances proposed as POPs". POPs are organic substances that persist in the environment, accumulate in living organisms and pose a risk to our health and the environment. POPs are regulated worldwide by the Stockholm Convention and the Aarhus Protocol [24]. These international treaties are implemented in the European Union by the POPs Regulation [25].

Companies in the EU are increasingly replacing hazardous chemicals and manufacturing processes with safer chemicals and greener technologies. This business approach can bring significant benefits to companies, the environment, and the health of workers and consumers. Additionally, it can have a substantial positive impact on the implementation of a circular economy [26]. In the company "Omorika Reciklaža" Ltd, a "chemical inspection" was conducted as part of a comprehensive environmental audit to verify whether chemicals listed by ECHA are used in the production process. The company mainly recycles PET packaging and produces several types of semi-finished and finished products made from recycled PET, but also recycles other types of plastic such as polyethylene (PE), polypropylene (PP) and polystyrene (PS).

RESULTS AND DISCUSSION

The audit included a specification of the facility and the processes. Only the block diagrams of the processes for the production of crushed flakes of PET (Figure 1) and the production of PET film for thermoforming (Figure 2) are shown here, i.e. only for those processes in which the implementation of the principles of green chemistry is proposed. Other plant parts or processes as well as energy flows will not be considered here. Therefore, the presented results are the analysis of chemicals ("chemical inspection"), water and wastewater streams, as well as the analysis of the generated waste.

As part of the "chemical inspection", a review and analysis of the contents of 26 SDSs for substances and mixtures used in the company was carried out. Here is a brief overview of the research, where commercial names of substances are not listed for reasons of confidentiality. Table 1 provides an overview of the most commonly used chemicals with the amounts consumed in 2021. In the production process, the largest quantities of chemicals are used in the washing process, specifically sodium hydroxide and detergents. In smaller quantities, appropriate additives are used in the facility to enhance certain product properties. Aerosols in the form of pressurized sprays are used as needed for corrosion removal, lubrication (containing concentrated silicone oil), and surface cleaning. All the mentioned aerosols do not contain persistent, bioaccumulative, and toxic (PBT) substances, very persistent and very bioaccumulative (vPvB) substances, SVHC, or contain them at less than 0.1% w/w in their composition.

Sodium dichloroisocyanurate dihydrate (NaDCC) is used as a disinfectant. This substance can be found in the form of white crystals, powder or tablets. NaDCC is a stable salt that contains chlorine and is soluble in water, forming a solution containing hypochlorous acid and isocyanurate acid. This solution is an effective disinfectant used for water disinfection, including drinking water, swimming pools, public bathrooms, and other facilities where maintaining hygiene is crucial. NaDCC has advantages over other disinfectants like chlorine as it has a milder odor and is less irritating to the skin and mucous membranes. Additionally, NaDCC can be used in smaller quantities compared to other disinfecting agents, which can reduce its environmental impact. Although NaDCC is considered relatively safe, it is essential to handle it with care as it can be irritating to the eyes and skin and harmful to health in larger quantities. NaDCC is harmful if swallowed (H302), causes serious eye irritation (H319), may cause respiratory irritation (H335), and is very toxic to aquatic life with long-lasting effects (H410). Therefore, special precautions are necessary when handling it, following the SDS.

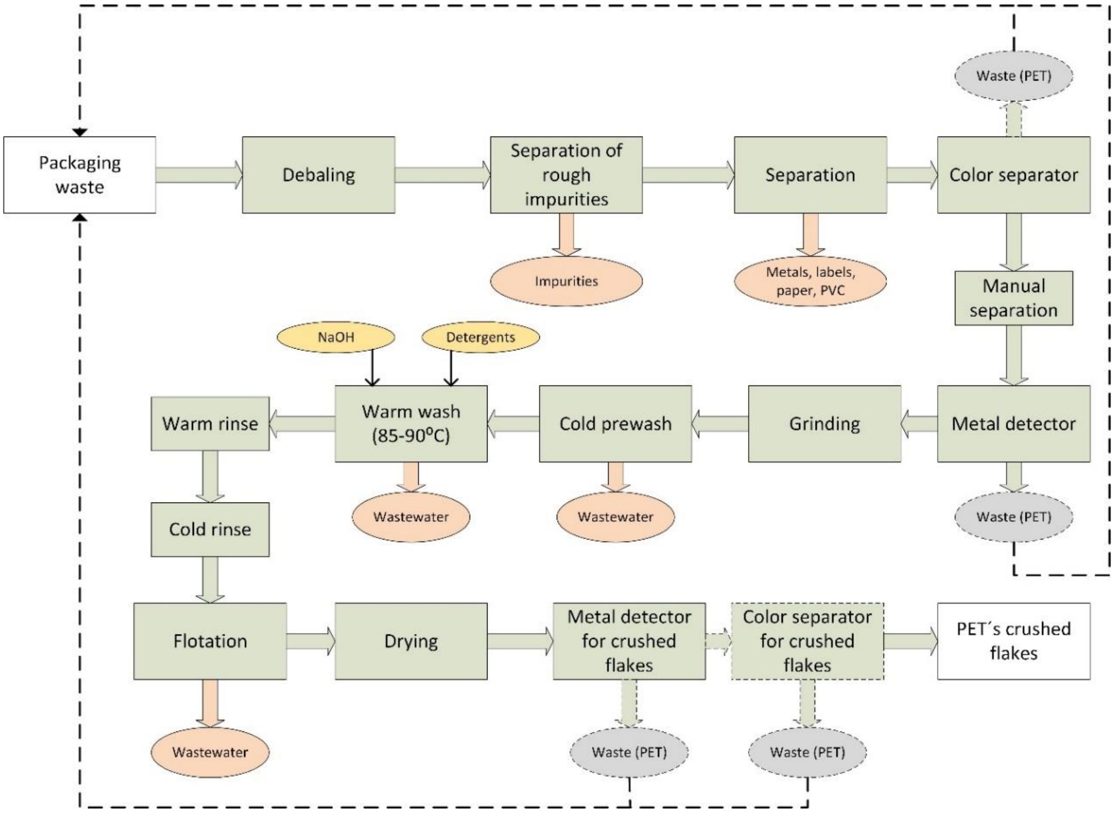

Figure 1. Block diagram of the process for the production of PET´s crushed flakes

To prevent the occurrence of electrostatic electricity in plastic packaging and consumer goods, an effective antistatic agent is used (safe for food contact). It neutralizes the static charge that polymeric materials acquire during production and processing. The antistatic agent can cause severe eye damage (H318) and is toxic to aquatic life (H411). The active ingredient is registered on the positive list for "food grade materials" in accordance with Regulation (EC) No 1935/2004 [27]. To reduce adhesion between surfaces, another additive (antiblock) is used, which does not contain hazardous substances and is safe for use.

The mixture used as an antifoaming agent and flotation aid is a combination of highly efficient substances (nonionic and anionic surfactants) that suppress foam formation and additives that assist in separating PET and HDPE or PP through the flotation process. This mixture is classified as hazardous because it can cause severe eye irritation (H319) and is harmful to aquatic life with long-lasting effects (H412). However, the mixture contains (<0.25% w/w) octamethylcyclotetrasiloxane (D4) (CAS 556-67-2), which is classified as an SVHC according to ECHA due to its PBT and vPvB properties. For this reason, it has been included in the Candidate List for authorization, which means a complete ban on its use can be expected very soon. The same mixture contains (<1% w/w) decamethylcyclopentasiloxane (CAS 541-02-6), known as D5, which is classified as an SVHC substance due to its PBT and vPvB properties and is included on the Candidate List for authorization.

D4 and D5 are also on the List of substances proposed as POPs, which means that they are being prepared for their potential inclusion in the Stockholm Convention. In the Republic of Srpska, D4 and D5 are on the List of substances that are candidates for inclusion in the list of SVHC by Ministry of Health and Social Welfare [20].

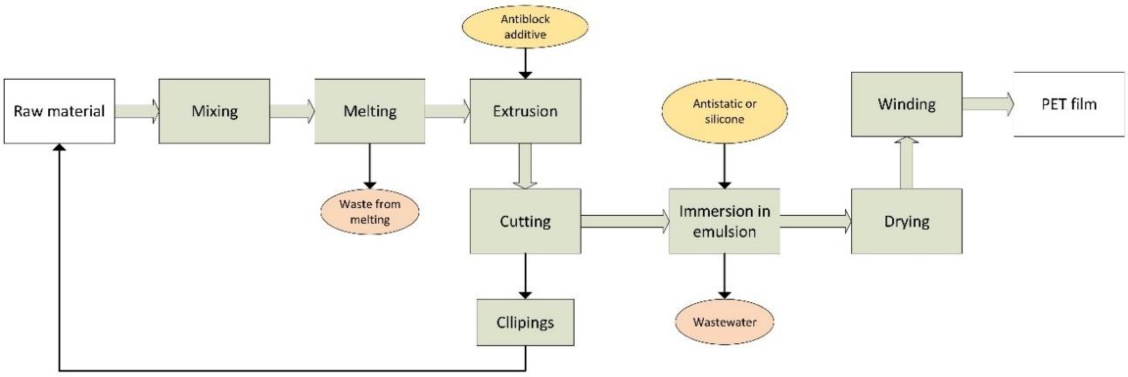

Figure 2. Block diagram of the process for the production of PET film for thermoforming

Table 1. Data on the type and consumption of chemicals

Chemical name	Location of use	Hazard Statement	Consumption in 2021.
Caustic soda	Warm wash process of PET´s crushed flakes (Fig. 1)	Corrosive. Hazards: May be corrosive to metals (H290). Causes severe skin burns and eye damage (H314)	10 - 12 t
Detergent and cleaning agent 1	Warm wash process of PET´s crushed flakes (Fig. 1)	May cause eye damage (H318)	1.44 m^3
Detergent and cleaning agent 2	Warm wash process of PET´s crushed flakes (Fig. 1)	Irritating to skin (H315). Causes eye damage (H318). Harmful to aquatic life (H412)	2 m^3
Antifoam and flotation agent	Flotation (Fig. 1)	Causes severe eye irritation (H319). Harmful to aquatic life (H412). Contains SVHC substances	700 kg
Antistatic	Production of PET foil (Fig. 2)	Causes severe eye damage (H318). Toxic to aquatic life with long-term effects (H411)	0.1 m^3
Antiblock	Production of PET foil (Fig. 2)	It is not a dangerous substance.	100 kg

The cleaning agent is a mixture of nonionic surfactants (ethoxylated isododecanol) and solvents (alcohol) and can cause severe eye damage (H318). Isotridecanolethoxylate (ITO) is a surfactant commonly used in cleaning products and detergents. It is an alcohol with a single side methyl group and a total alkyl chain length of 13 carbon atoms, along with 1-2 polyethylene oxide groups. ITO is a type of surfactant that enhances the detergent's ability to remove impurities from surfaces. This surfactant can be found in various cleaning products and detergents, including laundry, dishwashing, and floor cleaning products. Although it is considered relatively safe, isotridecanolethoxylate can cause eye and skin irritation in concentrations exceeding the recommended dose.

Isotridecanolethoxylates (CAS 69011-36-5) found in the mixture in an amount < 40% m/m are very toxic substances for aquatic life with long-term consequences. They are not on some of the lists that imply imminent authorization. Despite this, some chemical and cleaning product manufacturers are making efforts to replace them with less harmful surfactants. In addition to the mentioned isotridecanolethoxylate, another washing and cleaning agent is also used, which is a mixture of ethoxylates of fatty alcohols, complexing and dispersing agents. It also contains isotridecanolethoxylates in an amount < 40% m/m. The mixture causes skin irritation (H315) and can cause severe eye damage (H318) and is harmful to aquatic life with long-term effects (H412), which requires special care when handling and treating wastewater.

The highest consumption of water is in production of PET's crushed flakes, where water is mainly used for the process of cold and warm washing, rinsing and in the flotation process. By analyzing the wastewater and water streams, special attention was paid to the washing process in production of PET's crushed flakes (Fig. 1). Table 2 provides an overview of water consumption and the amount of wastewater generated in 2021. Raw water is used in the washing process, without prior treatment. The hardness of raw water and possibly present suspended matter and water turbidity cause inefficient use of chemicals for the washing process, which is why the consumption of chemicals is higher. The proposal is to provide water treatment (demineralization) for the washing process, where a washing mixture containing surfactants is used. In the process of warm washing, caustic soda is added to the water (the final concentration of NaOH is 2%) and three types of detergents (in the final concentration of 1, 3, 5 mg/L). If the water has a high total hardness at a temperature of 85 – 90 °C and this concentration of NaOH, insoluble $CaCO_3$ can precipitate (scale formation) on the walls of the reactor and heaters, which affects the reduction of the efficiency of the heaters and increases energy consumption.

Table 2. Water consumption and the amount of wastewater generated in 2021.

Process	Water consumption	Water type	Wastewater quantity
Washing and rinsing of PET´s crushed flakes (Fig. 1)	5 390 m^3	Raw water	< 5 405 m^3
Flotation (Fig. 1)	1 348 m^3	Raw water	< 1 348 m^3
Immersion of PET foil in emulsion (Fig. 2)	4 m^3	Deionized water	< 4 m^3

During the grinding of waste PET in the production of PET crushed flakes (Fig. 1), there is an emission of PET dust, which is a useful by-product. A part of this dust is collected and sold in the market (approximately 40 tons of PET dust annually), and the rest is waste due to the low efficiency of the dust collection system. Dust collection is carried out through ventilation and bag filters, but this method is not entirely efficient. The audit determined that there is a possibility to improve dust collection by installing new filters on the PET's crushed production line, which could increase the amount of collected dust and reduce the proportion of generated waste.

PROPOSED MEASURES FOR IMPLEMENTING THE PRINCIPLES OF GREEN CHEMISTRY

After the audit was completed, measures were proposed for the implementation of the principles of green chemistry (without measures related to energy flows):

1. Replacement of the mixture used as an antifoam and flotation agent (contains SVHC and substances proposed as POPs),
2. Water treatment (demineralization) for the process of washing PET´s crushed flakes,
3. Improve the dust collection system.

Measure no. 1 represents the fifth principle of green chemistry: Safer solvents and auxiliaries. The average annual consumption of this chemical is 700 kg. By replacing it with a safer alternative, the use of hazards chemicals would be completely avoided, thus protecting human health and the environment. Measure no. 2 represents the first and sixth principle of green chemistry: Waste prevention and Design for energy efficiency. The implementation would reduce the amount of wastewater and the consumption of chemicals for the washing process. The same washing effect would be achieved with a smaller amount of chemicals. Due to the use of NaOH in heated water that has high hardness (raw water), scale deposits occur, which reduces the efficiency of the heater and increases the energy consumption for heating water (85-90 ºC). Measure no. 3 represents the first principle of green chemistry: Waste prevention. It is expected that the emission of waste PET dust into the work area will be reduced by ≈ 50% and the share of dust sold on the market will increase.

The report of the environmental audit was presented to the management of the company Omorika Reciklaža Ltd. who accepted the implementation of the proposed measures. Currently, measures 1 and 3 have been implemented, and measure 2 will be implemented later. With the implementation of measure 1, the antifoam and flotation agent was taken out of use without affecting the quality of the product. The company's staff tested the possibility of performing flotation without this agent and the dangerous mixture was completely phased out by December 2022. This reduced the cost of purchasing the given chemical. By installing new bag filters in

January 2023, measure 3 was implemented. Filters (diameter 20 cm, length 4 m) are installed on the line for the production of PET´s crushed flakes, after the drying process (Fig. 1), where a large amount of PET dust appears. Bag filters contain antistatic, suitable for working in certain explosive zones. So far, it has been observed that the amount of dust in the production facility is lower compared to the period when the old dust collection system was used. Monitoring of this measure continues.

CONCLUSIONS

Green chemistry and the EU REACH regulation, as the main drivers of implementing green chemistry principles in the industry, promote sustainability and the reduction of the harmful impact of chemicals on the environment and human health. Through an environmental audit, the possibility of improving the technological process for this case study in the plastic recycling industry has been identified in terms of applying the principles of green chemistry, with the aim of protecting human health and the environment. The proposed 3 measures result in:

- Ceasing the use of a mixture containing two substances (organosiloxanes) classified as SVHC and substances proposed as POPs.
- Reducing the amount of waste PET dust while simultaneously increasing the production of PET dust for the market (byproduct).
- Reducing the consumption of chemicals in the warm washing process by water demineralization treatment while increasing the efficiency of the heater and increasing energy consumption.

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