Technical performance assessment of large scale process concepts for the recycling of end-of-life photovoltaic panels

Szabolcs Fogarasi[1], Florica Imre-Lucaci[2], Ana-Maria Cormoş[1], Árpád Imre-Lucaci[1]

[1]Department of Chemical Engineering, Faculty of Chemistry and Chemical Engineering, Babeş-Bolyai University, Cluj Napoca, RO-400028, Romania
[2]Interdisciplinary Research Institute on Bio Nano Sciences, Babeş-Bolyai University, Cluj Napoca, RO-400271, Romania
Corresponding author email: arpad.imre@ubbcluj.ro

Abstract

Among the key challenges of modern society to achieve an equilibrium between global development and conservation of biosphere and other valuable resources is to define novel and innovative approaches that can transform waste and by products into recyclable raw materials. In attempt to provide a useful contribution to sustainable development, the current study presents novel large scale process concepts for the comprehensive recycling of end-of-life photovoltaic panels (PhVP). To identify the optimal recycling plant configuration, the technical performance indicators were evaluated and compared for the operation with and without CO_2 capture and with/without thermal integration. All case studies assessed the dependency of process performance on the type of reducing system, glucose+NaOH+Na_2CO_3 or Al, used in the case of silver recovery. It was found that in the best operating conditions the total production of recovered materials is 8800 kg/h which represents an average recovery yield of 89% and apart from Si and Ag, the individual recovery yields are over 98% which reveals a high performance for the overall process. The results revealed that the thermally integrated process concept using Al in the silver recovery subsystem achieves higher performance for both energy generation and material consumption even with post-combustion CO_2 capture and storage.

1. Introduction

The industrial and building sectors are the largest energy consumers and emitters of CO_2 accounting for more than 300 EJ of the total final energy consumption and 90% of the global electricity consumption in 2022 (IEA 2023). Current trends clearly indicate that both sectors of the economy face a significant rise in electricity demand due to different applications and polices, projected to increase twice by 2050 in the STEPS Scenario and even higher in more ambitious scenarios. In consequence, global electricity demand is set to increase by 80% between 2020 and 2050, with emerging markets and developing economies driving more than 85% of this growth (IEA 2023). But there is no economic growth without providing sustainable energy production which is one of the key components of sustainable development and it has to be indispensable to ensure a climate neutral economy (Golroudbary, et al. 2024; Rabaia, et al. 2022).

Photovoltaic (PV) power systems have emerged as one of the most promising alternatives to reduce the impact of the energy sector on climate change and to improve the energy security of countries all over the world (Chowdhury, et al. 2020; Mahmoudi, et al. 2020). Thanks to process improvements and a switch to low-emissions power generation, the emissions intensity of solar PV manufacturing has decreased by 40% in the last decade, driving an 80% cost decline and positioning solar PV as the most affordable electricity generation technology in many parts of the world (IEA 2022a; IEA 2022b). However, this surge in electricity demand necessitates the increase of capacity additions for PV systems with more than 600 GW per year between 2020 and 2030 leading to 20 times higher contribution to global electricity generation by 2050 than 2020 (IEA 2021; IEA 2022a; Romel, et al. 2024). By 2050, renewable are expected to provide nearly 70% of electricity generation, with PV systems accounting for about 30% of total generation in comparison to roughly 3% in 2021 (IEA 2021; Liu, et al. 2020). But the rapid expansion in PV installations comes with its own set of challenges, notably concerning the demand for critical minerals and the management of end-of-life PhVP. In 2021, solar PV panel production already claimed a significant share of global resources, accounting for 10% of the demand for silver and over 40% of global tellurium usage (IEA 2022a; IEA 2022b). However, looking ahead, the PV industry's demand for critical minerals is projected to surge dramatically, with estimates ranging from 150% to 400% between 2021 and 2030, raising concerns about supply chain stability and environmental impact (IEA 2022a). By 2030, forecasts suggest that the demand for critical minerals could skyrocket to 4,000 kt, a substantial increase from the 1,000 kt recorded in 2021. Moreover, the demand for silver in solar PV manufacturing is anticipated to climb to over 30% of total global production by 2030 and 70% by 2050 (Briand, et al. 2023; IEA 2022a; IEA 2022b).

This growth driven high energy and raw material consumption cannot be economically sustained by existing supplies without effective recycling measures in place. For the above reasons the European Union revised the WEEE Directive (2012/19/EU) making mandatory for all producers recycling rates 85%/80% mass recovery rate/recycling rate of end-of-life PhVP from 2018 (Deng, et al. 2019; Wang, et al. 2022). Similar approach was made by the South Korea's Ministry of Environment but with and effective application from 2023 (Deng, et al. 2019). According to literature data there are estimates for the accumulation of 1.7–8 million tons by 2030 and 60–78 million tons by 2050 of end-of-life PhVP (Mahmoudi, et al. 2021; Nazar, et al. 2024). If these amounts of end-of-life PhVP are processed at 85% recovery rate, imposed by the WEEE directive, the recycled materials could cover 3-7% of the solar PV industry's demand for Al, Cu, glass, Si and Ag required during 2031-2040, and over 20% for Al, Cu, glass, Si and almost 70% for Ag in the period of 2041-2050 (IEA 2022b). Huang et al. and Shao et al. estimate that the recycling process of 78 million tons of end-of-life PhVP could generate a minimum USD 15 billion in revenue and maximum USD 60 billion in revenue if solargrade quality materials are obtained (Huang, et al. 2017; Shao, et al. 2023). Nevertheless, recycling must be applied not only to provide resources for the manufacturing of new photovoltaic panels (PhVP) but to process and valorize the large amounts of end-of-life PhVP (Li, et al. 2023). Besides the economic advantages related to the reuse of recycled materials in the manufacturing of new PhVP, Wang et al. reveal that greenhouse gas emissions are cut down by 42% (Wang, et al. 2022). An additional benefit of end-of-life PhVP recycling relies on the fact that it involves processing a single material containing all essential metals for PhVP production, in concentrations surpassing those found in any ore and is more uniformly distributed worldwide (Jose-Luis, et al. 2019; Yue, et al. 2022).

To manage such a large quantity of waste material goes beyond just enforcing policies, it requires rethinking and redesigning some key steps of existing e-waste treatment technologies in order to apply them cost effectively for end-of-life PhVP recycling (Choi and Fthenakis 2014; Heiho, et al. 2023). Unfortunately, many studies try to solve the problem only at laboratory scale and performing experimental assessments on different (high-value) material fractions present in end-of-life PhVP or targeting the recovery of some valuable components (Deng, et al. 2022; Pagnanelli, et al. 2017). Other publications offer a larger focus evaluating the process from technical, economic and environmental point of view but disregarding important details and not applying a comprehensive industrial scale approach (Ardente, et al. 2019). Given that end-of-life PhVP recycling is in its infancy and the increasing demand for large-scale industrial recycling technologies, this study outlines a comprehensive process and assesses technical capabilities not on a subsystem-by-subsystem basis at laboratory scale, but within an integrated large-scale recycling facility encompassing all critical subsystems. This methodology yields overall conclusions on technical performance variations for different process concepts of the industrial-scale recycling plant and reveals the influence of post-combustion CO_2 capture on recycling efficiency.

Keywords: spent Li-ion batteries, recycling, modeling, simulation.

2. Plant configuration and model assumptions

2.1. Description of the mathematical models developed for the end-of-life PhVP recycling plant

The mathematical models developed for the recycling process of end-of-life PhVP with a processing capacity of 10 t/h are presented in Fig.1. The technical performances of the end-of-life PhVP recycling plant were evaluated for the following base scenarios considering two different reducing systems for the recovery of Ag:

- **Case 1** - the reducing system is glucose + NaOH + Na_2CO_3
- **Case 2** - the reducing agent is Al.

In addition, for both base scenarios the following sub-cases were defined in order to find the optimal technological option:

(I) - non thermal integration (additional amounts of CH_4 were considered to cover the thermal energy consumption);

(II a) - thermally integrated recycling process;

(II b) - thermally integrated recycling process with post-combustion CO_2 capture process.

According to the mathematical models the recycling plant involves a sequential processing of end-of-life PhVP that leads to the recovery of glass, organic material (TEDLAR) and silicon respectively silver that can be reintroduced in the production of new PhVP. For both case studies defined in Fig. 1. the end-of-life PhVP recycling plant is divided into the following subsystems:

1. **Processing and mechanical separation of end-of-life PhVP** is the first stage of the process and has the main purpose of disassembling and sorting used PhVP into different material fractions. In this step, the Al frame and the fine/intermediate glass fractions are separated from the stream of the coarse fraction which is represented by the photovoltaic cells whose components are held together thanks to the EVA polymer.
2. **The processing of the coarse fraction** begins with the dissolution in C_6H_{12} of the organic material (TEDLAR) which by filtration is separated from the solid materials and by centrifugation leads to the obtaining of TEDLAR and the recirculation of C_6H_{12} in the process. Photovoltaic cells are separated by flotation from the solid fractions and the electrostatic separator leads to the separation of glass from Al and Cu respectively later Al from Cu.
3. **Ag and Si release** is achieved by pyrolysis at a temperature of 400 °C. The gaseous phase resulting from the decomposition of the EVA polymer is processed in a column to recover CH_3COOH at the base of the column and the gaseous stream at the top of the column, which is subjected to a combustion process.
4. **Dissolution and purification.** The solid material from the previous step is subjected to a leaching process with HNO_3 which allows the separation of silver, in the form of $AgNO_3$ solution, by filtration from silicon. AgCl is precipitated from the $AgNO_3$ solution, in the presence of NaCl, which is filtered and further processed in the last stage of the process.
5. **Silver is obtained by reducing AgCl** to Ag using one of the two reducing systems shown in Fig. 1. In case 1, AgCl is reduced using the reducing system glucose+NaOH+Na_2CO_3 and in case 2 the reducing agent is Al obtained in the previous steps. The separation of Ag from $AlCl_3$ and other secondary products is carried out by melting and filtering at 500 °C.

2.2. Description of the mathematical model developed for the CO_2 capture process

The recycling plant for end-of-life PhVP was integrated with a post-combustion CO_2 capture system based on amine absorption. The CO_2 capture process, as illustrated in Figure 2, comprises three main stages:

(i) CO_2 absorption into a lean amine solution at temperatures ranging from 35 to 55°C and a pressure of 1.05 bar.

(ii) The rich amine-CO_2 stream undergoes pumping and heating to temperatures of approximately 100 to 120°C, utilizing heat from the solvent mass at the bottom of the desorption column. Following preheating, the CO_2-rich stream proceeds to the desorption column, where solvent regeneration occurs with thermal energy provided by the recycling plant.

(iii) The CO_2 stream is subsequently dried and compressed in four stages, reaching a storage pressure of 122 bar.

2.3. Methodology and basic assumption

The evaluation of the technical performances for the recovery processes of metals from end-of-life PhVP based on the defined mathematical models were carried out by simulating and optimizing the processes using process flow modeling software CHEMCAD specific to chemical engineering. Considering the operating conditions and the physico-chemical properties of the compounds involved, the Soave–Redlich–Kwong (SRK) thermodynamic model with Boston–Mathias modifications was used. In the simulations, chemical and phase equilibrium conditions based on the Gibbs free energy minimization model were considered. The pressure loss in the heat exchangers was considered 1%, the minimum temperature difference for the thermal integration of 10 °C and the pressure drop in the barometric condenser of 46 mbar.

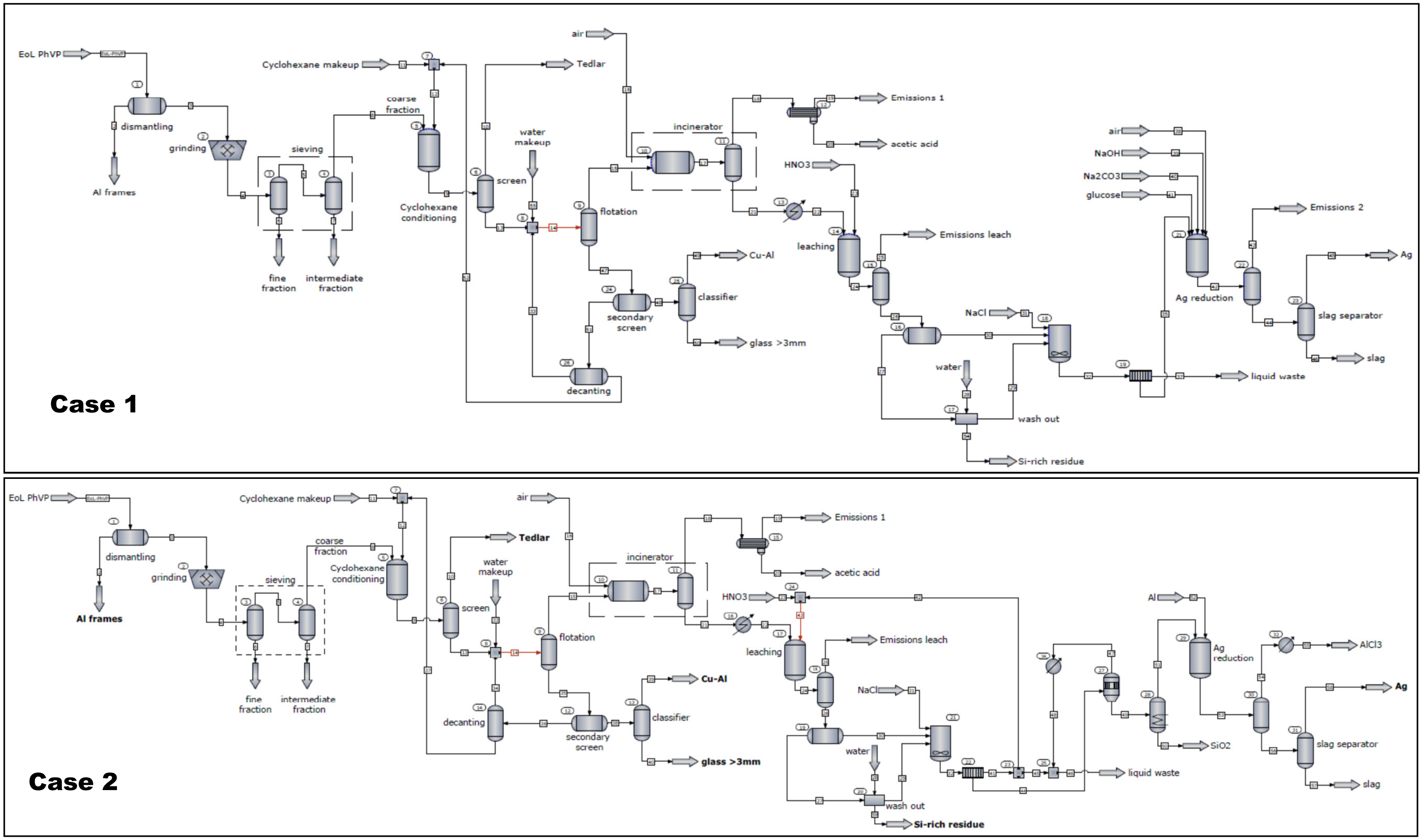

Fig. 1. Process flow diagram of the recycling plant of end-of-life photovoltaic panels for the two case studies.

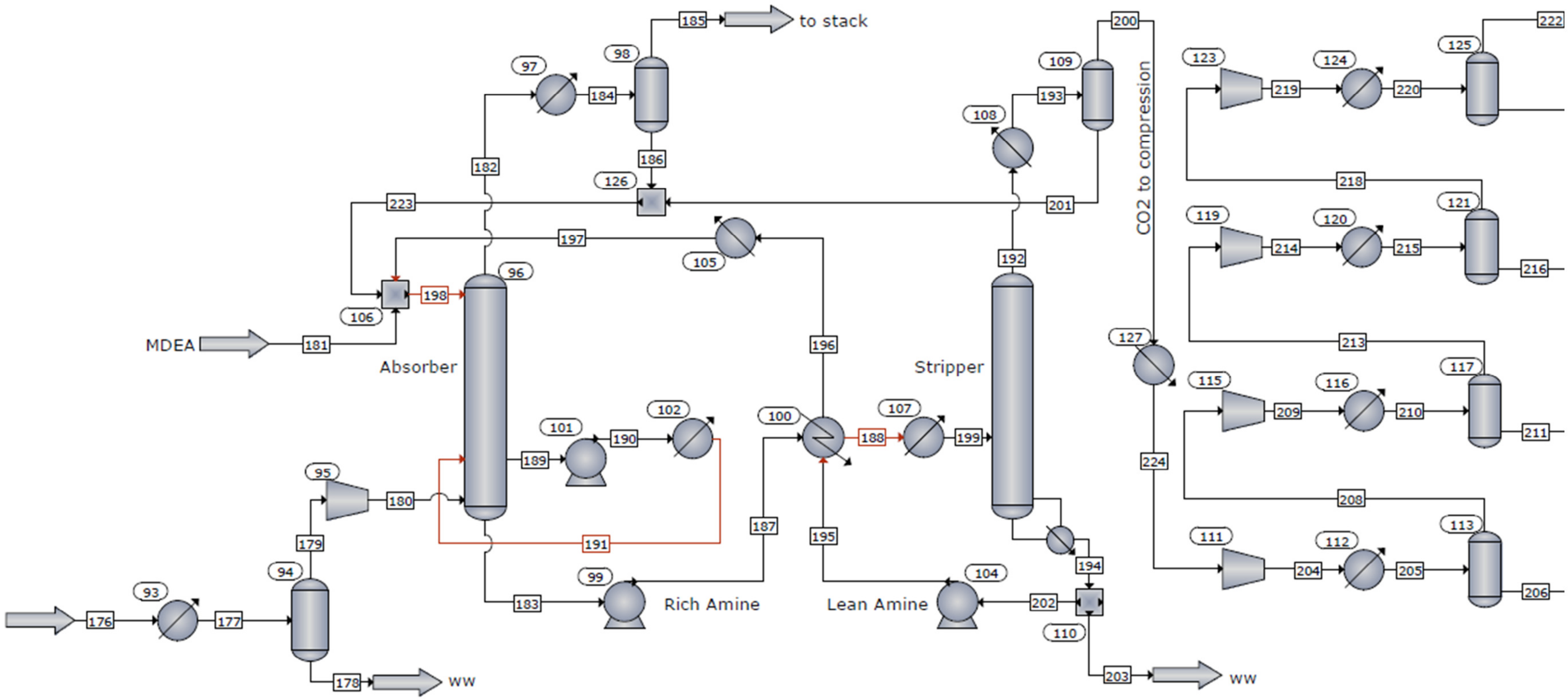

Fig. 1. Process flow diagram for the CO_2 capture process.

Results & Discussion

The mathematical models developed and presented in Fig. 1. for the recovery process of metals from end-of-life PhVP were simulated in order to find the optimal technological option. The production rate and recovery yield for the main products of the end-of-life PhVP recycling process were calculated based on the material balance data. Considering the obtained results present similar values for the performance indicators defined for case 1 and case 2, only the average values are presented in Table 1.

Table 1. Average production rate and recovery yield for the main products of the end-of-life PhVP recycling process.

Process type	Product	Glass	Tedlar	Si	Ag	Al	Cu
Case 1 and Case 2	**Production rate, kg/h**	6950	430	330.2	4.45	1000	108
	Recovery yield,%	97	98	89.23	89.01	98.83	99.99

The total production rate of recovered materials is 8800 kg/h which represents an average recovery yield of 89%. Apart from Si and Ag, the individual recovery yields are over 98% which reveals a high performance for the overall process. The high values for the production of Al and glass are also related to the fact that these two materials are present in the highest proportion in end-of-life PhVP. Regarding the specific consumption of raw materials, relative to the processing capacity, it can be observed based on the data in Table 2 that case 2 provides a slightly higher performance than case 1. This difference is determined by the much lower consumption of HNO_3 for the dissolution and purification process in subsystem 4 for case 2.

Table 2. Specific consumption of raw materials in kg/kg PhVP to obtain the main products of the recycling process of end-of-life PhVP.

Process type	Raw material		H_2O	HNO_3	NaCl	NaOH	Na_2CO_3	$C_6H_{12}O_6$	Air	C_6H_{12}	TOTAL
Case 1	**Consumption**	**kg/h**	919	1713	26.39	2.42	1.17	1.12	16004	203.1	-
		kg/kg PhVP	0.09	0.17			~ 0		2.75	0.02	**3.03**
Case 2	**Consumption**	**kg/h**	919	210	2.5	-	-	-	16000	203.1	**-**
		kg/kg PhVP	0.09	0.02	~ 0	-	-	-	2.75	0.02	**2.88**

This aspect becomes even more obvious if we analyze the specific consumption of raw materials in kg/kg product for different subsystems of the end-of-life PhVP recycling process. From the data presented in Table 3, it can be seen that subsystem 4 has the highest specific consumption of raw materials for case 1 and is followed by subsystem 1, which is the most material-intensive subsystem even for case 2. Moreover, summing up the specific consumption of raw materials, of the different subsystems in Table 3, leads to a total that is 40% higher for case 1 than for case 2, which reveals the superiority of the technological variant defined in case 2.

Table 3. Specific consumption of raw materials in kg/kg product for different subsystems of the spent PhVP recycling process.

Process type	No. subsystem	1	2	3	4	5	TOTAL
Case 1	**TOTAL, kg/h**	10203	266	16000	2392	9.6	9.2
	W, kg/kg	2.91	0.05	0.91	4.81	0.52	
Case2	**TOTAL, kg/h**	10203	266.1	16000	864.9	0.38	

W, kg/kg	2.91	0.05	0.91	1.74	0.06	5.67

In contrast, case 1 is the more efficient variant from the energy point of view if both processes are not thermally integrated. As the data in Table 4 shows that subsystem 4 has a consumption almost 3 times higher for case 2 than for case 1, being the most energy-intensive subsystem and the one that differentiates the two case studies the most. However, it is worth noting that in both case studies subsystem 3, where the pyrolysis and combustion of organic materials takes place, provides an enormous amount of energy at a relatively high potential.

Table 4. Thermal energy balance for different subsystems of the end-of-life PhVP recycling process for the thermally non integrated case studies.

	No. subsystem	Parameters of energy flows	Thermal energy, MJ/h Generated			Thermal energy, MJ/h Consumed	TOTAL consumption, MJ/h	Specific consumption, kJ/kg	Equivalent consumption, kg CH_4/h
			1	**2**	**3**	**1**			
Case1	**1**	**T, °C**				35			
		Q, MJ/h				68	**68**	**6.80**	**1.36**
	2	**T, °C**				34			
		Q, MJ/h				61	**61**	**12.05**	**1.21**
	3	**T, °C**	400	400	400				
		Q, MJ/h	-4900	-51581	-89				
	4	**T, °C**	0			60			
		Q, MJ/h	0			204	**204**	**411**	**4.08**
	5	**T, °C**	600						
		Q, MJ/h	-1.40						
Case2	**1**	**T, °C**				35			
		Q, MJ/h				39	**39**	**3.90**	**0.78**
	2	**T, °C**				34			
		Q, MJ/h				61	**61**	**12.04**	**1.21**
	3	**T, °C**	400	400	400				
		Q, MJ/h	-4900	-53120	-89				
	4	**T, °C**	40			60			
		Q, MJ/h	-491			532	**532**	**1070**	**10.64**
	5	**T, °C**	500						
		Q, MJ/h	-7						

The amount of thermal energy generated in the process eclipses the energy consumption for both case studies, and because it is released at a much higher temperature than the temperature at which energy must be supplied to the process, it can be used for the thermal integration of the process. Thus, the energy consumption is reduced to zero (Table 5) for both thermally integrated case studies (IIa) and the generated thermal energy decreases by only 0.3%, maintaining at over an equivalent of 1100 kg CH_4/h.

Table 5. Overall thermal energy balance for the recycling process of end-of-life PhVP.

Process type		**Parameters**	**Thermal energy** Generated, MJ/h			**Thermal energy** Consumed, MJ/h	**Equivalent CH_4, kg CH_4/h** Generated	**Equivalent CH_4, kg CH_4/h** Consumed
Case 1	**I**	**T, °C**	600	400	600	60		
		Q, MJ/h	-1.4	-56570	-1	333	**1131**	**6.66**
	IIa	**T, °C**		400				

		Q, MJ/h		-56365			**1127**	
	IIb	**T, °C**		400				
		Q, MJ/h		-46487			**930**	
	I	**T, °C**	40	400	500	60		
		Q, MJ/h	-491	-58109	-7	631	**1162**	**12.63**
Case 2	**IIa**	**T, °C**		400				
		Q, MJ/h		-57577			**1152**	
	IIb	**T, °C**		400				
		Q, MJ/h		-47699			**954**	

The generation of such a high flow of energy, by burning organic materials in subsystem 3, also leads to high total and specific CO_2 emissions (Table 6). By using the CO_2 capture system (Fig. 2.) the obtained results indicate a 95% decrease in total and specific CO_2 emissions, for both case studies, regardless of whether it is the thermally non-integrated (I) or integrated (IIa) process. Of course, CO_2 capture requires additional thermal energy consumption that can be covered from the energy generated in subsystem 3, reducing the energy generated in the process by 20% (IIb). Even in this situation, the case studies remain thermal energy producers with the capacity to provide an equivalent of over 900 kg CH_4/h.

Table 6. Total and specific CO_2 emissions, respectively energy consumption of the CO_2 capture process for the recycling of end-of-life PhVP.

Process type		**No capture**		**With capture**		**Capture energy consumption CO_2, GJ/h**
		Total, $kgCO_2$/h	**$kgCO_2$/t PhVP**	**Total, $kgCO_2$/h**	**kg CO_2/t PhVP**	
Case1	**I**	2989	298.92	149	14.95	9.94
	IIa	2971	297.09	149	14.85	9.88
Case2	**I**	3006	300.55	150	15.03	9.99
	IIa	2971	297.07	149	14.85	9.88

Conclusions

The technical assessment of the designed, modeled and simulated recycling plant revealed its potential application for the comprehensive treatment of end-of-life PhVP. It was found that in the best operating conditions the average recovery rate of valuable materials is over 89% and the purity of the obtained products was more than 99 % which makes them appropriate for new PhVP production. The results proved that in both case studies the pyrolysis and combustion of organic fractions of the waste material provides the necessary amount of energy for the thermal integration of the process. According to the results it can be stated that both case studies remain net thermal energy producers with the capacity to provide an equivalent of over 900 kg CH_4/h even in the situation with CO_2 capture. Comparing the specific raw material consumptions of the two case studies it can be concluded that the one using aluminum for silver reduction requires 50% less raw materials for the processing of 1 kg of end-of-life PhVP. Further studies should complete the assessment of the developed conceptual recycling plant by evaluating its economic viability and environmental impact, creating the basis for its comparison to other approaches presented in the literature.

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Data availability
All data generated or analyzed during this study are included in this publish article.
Declarations
Ethical approval: Not applicable.
Consent to participate: Not applicable.
Consent for publication: Not applicable.
Competing interests: The authors declare no competing interests.

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