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Environmental analysis of novel sorbents for mercury sorption

ABSTRACT: The reduction of mercury emissions in currently existing coal-based power plant solutions by each method i.e. preliminary, primary and secondary (consisting of introducing coal into the combustion chamber and then removing mercury from the combustion gases arising from the combustion process) does not solve the problem of achieving the required limits by power plants. Therefore, the need has arisen to look for new, effective solutions.

The results presented in the work concern the analysis of environmental benefits for the use of zeolites obtained from by-products of coal combustion such as fly ash (from hard coal and lignite) in technologies for removing gaseous forms of mercury. The tested zeolites were silver-modified X-type structures. The reference material in the considerations was active carbon impregnated with bromine – a commercially available sorbent on the market.

The article considers environmental benefits resulting from the use of tested zeolites taking the product life cycle, sorbent efficiency and the possibility of its regeneration compared to activated carbon (AC/Br) into account. The LCA analysis was performed taking the estimated material and energy balances of the manufacturing processes into account. When comparing the production pro-

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cess of type X zeolite materials on the processing line and activated carbons in the amount necessary to capture 375 g Hg from exhaust gases, the LCA analysis showed that zeolites contribute to a lower potential impact on the environment. The advantage is that 5 times less zeolite sorbent than activated carbons is needed to capture the same amount of mercury. In addition, zeolite materials can be regenerated, which extends their life time.

KEYWORDS: fly ash, zeolites, mercury, LCA

Introduction

Mercury is one of the most hazardous pollutants emitted into the atmosphere. Mercury emissions to the atmospheric air come, among others, from fuel combustion processes for the production of electricity and heat, industrial processes, fuel combustion and as a result of the re-emission of pollutants into the environment. The dominant sources of mercury emissions are energy production processes and industrial processes (Wdowin et al. 2014).

Mercury emissions from industrial processes (by-product emission) are emissions that accompany production processes, where mercury is a by-product of these processes or is a component of fossil fuels and raw materials (e.g. metal ores). The highest share in global emissions from industrial processes comes from the combustion of fuels (especially hard coal and lignite), iron, steel and non-ferrous metals as well as cement production (Li et al. 2009).

Emission from the use of mercury-containing products (product use emission) result from the deliberate use of mercury to produce products and then their use, which results in the release of mercury into the atmosphere, water and soil. Mercury is released at all stages of the product's life cycle, i.e. during the extraction of raw materials, production and subsequent use of products and waste management.

The highest share of global mercury emissions to the atmosphere from industrial processes comes from the combustion of fuels, especially coal, in the energy, industrial and municipal and housing sectors, which represents about 45% of total anthropogenic mercury emissions. Cement production processes and the production of iron, steel and non-ferrous metals are another important source in global mercury emissions. The data for 2018 shows that among European countries, Poland ranks second in terms of anthropogenic mercury emissions to air (Mercury in Europe's... 2018).

The release of mercury into the environment negatively affects human health and the environment. Among the effects of mercury on the human body, the worst is the effect of methylmercury, which damages the central nervous system and brain. Human fetuses are most sensitive to methylmercury. The consumption of mercury accumulated in food, e.g. marine fish, during pregnancy by mothers may result in a lower IQ intelligence ratio and even autism of the children. The average total mercury intake was estimated at 4.74 and 3.07 µg/day in males and females, respectively (Bose-O'Reilly et al. 2010; Kim et al. 2016).

Mercury also increases the risk of heart attack, allergies and cancer. Social costs are associated with cases of hypertension in adults and children, immortal cases of heart attacks and premature death. The above social health costs have been estimated as 7 times higher than those associated with reduced intelligence (Bose-O'Reilly et al. 2010).

Changes in emission standards and thus the improvement of health and the environment are to take place through the introduction of the Industrial Emission Directive (IED). This is a combination of the IPPC Directive 96/61/EC and the directives on large combustion sources (2000/76/EC) and waste incineration (2001/80/EC). In addition, in accordance with the requirements of Directive 2008/1/EC, installations requiring integrated permits must meet the requirements of the so-called BAT conclusions (technical documents adopted by way of implementation decision of the European Commission and binding directly in the member countries) – above all the so-called emission limit values. This means much stricter requirements than currently included in the integrated permits. A comprehensive approach to the problems arising from the operation of large industrial plants, whose activities cause or may have a significant negative impact on the environment, primarily in terms of mercury emissions to the atmosphere, will be necessary. These emissions should be monitored from combustion installations with a rated thermal input greater than 50 MW. This covers the electricity and heat generation sectors and those industries in which conventional fuels are used. In addition, their combustion systems are not covered by other sector BREFs (Żmuda et al. 2017). Hard coal and lignite are considered as conventional fuels within the meaning of the LCP BREF. For countries such as Poland, over 93% of electricity comes from these fuels. Although mercury is present in coal in relatively small amounts, taking the streams of fuel burned into account, a typical coal-fired power plant emits approximately 50 kg of Hg annually to the atmosphere (Głodek and Pacyna 2016), which on a global scale gives about 1500 Mg/year.

It is therefore important to monitor mercury emissions to check compliance. In addition, it enables the installation operators to monitor the effects of their own actions and take corrective actions. The method and frequency of monitoring should be related to the scope of controlled emissions and the technology used. Therefore, choosing the right sampling points is extremely important. The points should be chosen so as to obtain a representative picture of the ventilation air flow.

It should be noted that adapting the Polish energy sector to the requirements set by BREF/BAT will be costly and will require the search for new, high-efficiency solutions. The currently used mercury removal techniques, i.e. primary, secondary and preliminary methods are not effective enough to meet them. One of the solutions, to achieve the required emission limits, may be to combine existing methods into hybrid methods in order to improve their reduction efficiency and optimize their costs. The environmental analysis i.e. Life Cycle Assessment (Life Cycle Inventory and Life Cycle Impact Assessment) solutions proposed in the article have not yet been carried out. Only LCI for the production of zeolite A (Fawer et al. 1998; Seo et al. 2019) and for the process with the application of zeolite in a warm mix asphalt (WMA) mixture (Ma et al. 2019) was investigated. In the case of technology of mitigation only for CCS, NO_x, SO_x cases LCA was carried out (Koornneef et al. 2008; Shing et al. 2016).

The purpose of the environmental analysis was to assess the potential environmental impact of the production process of zeolite materials from fly ash and to apply them to capture gaseous forms of mercury.

1. Purpose of the analysis and functional unit

The aim of the project was to develop new types of structural sorbents based on fly ash for remove gaseous forms of mercury from exhaust gases. Life cycle assessment of zeolites synthesis and their modification with silver compounds was made. Furthermore, pollution reduction was an additional factor identified for LCA and assessed in field of potential environmental impact. Due to the fact that the research proposed under the project will constitute a development of new directions in the management of aluminosilicate waste raw materials (fly ash), the designed products may be a micro/mesoporous materials alternative to commercially used. The indirect goal of the analysis was therefore also a comparative assessment of obtained sorbents with reference products available on the market. Ensuring functional equivalence is crucial in this case, because only such products can be subject to comparative LCA analyses (Kowalski et al. 2007). In order to specify the functional parameter (the so-called functional unit) it was determined that it will constitute 1 kg of produced porous material. In the case of a comparative analysis the same unit was adopted. Due to the fact that at present, the simplest and the most popular method mercury removal form exhaust gases is the injection of powdered activated carbon into the duct, this product was chosen as the reference for comparative analysis (bromine impregnated activated carbon).

2. The scope of the analysis

The next step was to outline the system boundaries, life cycle stages and unit processes, together with an indication of which areas of the product system should be considered as foreground system and which are background system (ISO ISO 14040:2009; ISO14044:2009).

A zeolite synthesis was carried out using a prototype technological line. As part of the conducted processes, zeolite type X was obtained. The basic synthesis material was fly ash, which is a by-product of coal and lignite combustion. In addition, the zeolite materials obtained were subjected to activation with silver ions, which increase the sorption properties of mercury. The following materials were selected for analysis:

- ◆ Na-X-Ag zeolite based on hard coal ash,
- ◆ Na-X-Ag zeolite based on brown coal ash.

The temporal and technological scope of the analysis applies to 2018. Data for individual synthesis and modification processes came from research carried out on the prototype installation of the Lublin University of Technology. Boundaries for the analyzed system were shown in Figure 1.

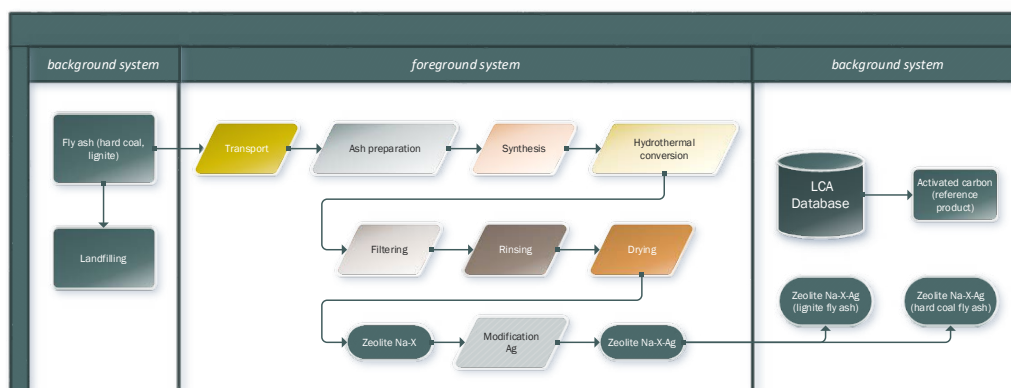


Fig. 1. The boundaries of the analyzed system

Rys. 1. Granice analizowanego systemu

Inventory data was collected in relation to the production unit constituting 50 kg of material received. The results were then converted into 1 kg of produced zeolite. The data obtained in this way is representative in terms of time, place and technology and may be the basis for conducting a potential environmental impact assessment using the LCA method. It covers, as all the inputs and outputs directly or indirectly related with environmental interventions. In order to perform a full LCA analysis, it was necessary to complete the data throughout the entire value chain. In this case, data from secondary sources (databases) was used.

3. Assumptions and exclusions

The following assumptions were adopted for LCA analysis:

- ◆ the LCA assessment of the synthesis and modification of zeolites was mainly based on data obtained directly from semi-technical scale processes implemented under the project, and the secondary data only supplemented the process data;
- ◆ due to the lack of necessary data, the infrastructure was excluded from the analysis;
- ◆ in order to determine the environmental benefits associated with the use of waste materials in the synthesis process, an approach related to system expansion and avoided burden was used. Due to the fact that at present the main technology for ash management is their land-

filling (25% is utilized and the remaining part goes to landfills) (Blisser and Rwozon 2012), using it to produce other goods avoids the necessity of depositing it in a landfill. This, in turn, contributes to avoiding emissions related to the leaching of pollutants, emissions to air in the form of dusts and energy necessary for their management. The analysis uses the profile for landfilling of average residues after hard coal combustion, available in the Ecoinvent database (v. 3.2) (<http://www.ecoinvent.org/>);

- ◆ data on the life cycle of consumables and fuels used in the supply chain are from Ecoinvent v3.2 databases and most are representative data at European level. In the case of electricity used, the data in the analysis was included in accordance with the national energy mix and also comes from the Ecoinvent v3.2 database;
- ◆ LCIA (Life Cycle Impact Assessment) was carried out using the environmental (cause-effect) model represented by the ILCD 2011 Midpoint + v.1.10 method (European 2010, 2012);
- ◆ for the weighing process, the weighting factor for each of the analyzed impact categories was 1/15.

The analyzed zeolite production process included data from semi-technical scale installations. The process of synthesis of porous materials using two reactors takes about 48 hours, where the process inputs are:

- ◆ technological water used for the synthesis and rinsing of the resulting product,
- ◆ energy in the form of heat (electric heaters), used to heat water,
- ◆ NaOH as a substrate for the synthesis of zeolite structures,
- ◆ fly ash from hard coal or lignite combustion as a base of porous materials.

We obtain 50 kg of the product in the form of zeolite from the synthesis process carried out in this way. Inputs and outputs from the process are summarized in Table 1.

TABLE 1. Inputs and outputs of the zeolite synthesis process

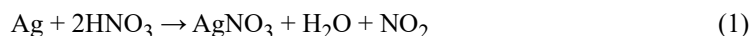
TABELA 1. Strumienie i współczynniki normalizacyjne użyte w analizie

Inputs	Amount	Unit
Process water	90	liter
Process water (first cycle)	80	liter
Process water (second cycle)	80	liter
NaOH	10,8	kg
NaOH (second cycle)	3,5	kg
Fly ash	50	kg
Electricity (reaction)	66	kWh
Electricity (drying)	35,28	kWh
Electricity (second cycle)	48	kWh
Outputs	Amount	Unit
Zeolites	50	kg
Sewage	250	liter

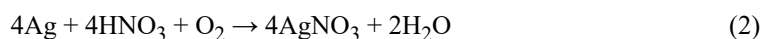
Source: own study based on data obtained in the project.

The LCI (Life Cycle Inventory) data used to build the background system of the zeolite production process came from the Ecoinvent v.3.1 database (<http://www.ecoinvent.org/>).

Due to the fact that zeolites are activated with silver nitrate, for which LCI data is not available, a simplified production process was created based on the stoichiometry of the reaction presented below.



On an industrial scale, the reaction is carried out with heat using medium concentrated acid (32%). The NO_2 formed in the reaction can be used to regenerate nitric acid. Assuming total NO_2 recovery, equation (2) describes the production process. Nitric oxide NO is oxidized in the air to NO_2 dioxide. Oxidation is also carried out by passing oxygen through the reactor.



4. Assessment of the potential environmental impact of zeolite synthesis and the silver modification process

The process of synthesis and modification of zeolites with silver compounds were assessed for their potential impact on the environment (Hauschild et al. 2012). The analysis carried out on the basis of data obtained from the semi-technical installation showed that the total cumulative indicator of the potential impact on the environment of the production of 50 kg of material (production from two reactors and 50 kg of ash feed) is 990 mPt, which is 19.8 mPt per kg (Table 7). The dominant factor is the use of silver nitrate. The share of this process is 99.5% of the overall impact indicator.

By qualitatively analyzing the obtained structure of the process impact, a significant share of four impact categories can be weighed:

- ◆ Mineral, fossil & ren resource depletion,
- ◆ Human toxicity, non-cancer effects,
- ◆ Human toxicity, cancer effects,
- ◆ Freshwater eutrophication.

Due to the dominant share of silver nitrate in the overall environmental impact indicator, the impact in the above categories is dominated by the production of silver used in the preparation of this nitrate. The majority of silver in Poland comes from the processing of anode sludges resulting from the mining and processing of copper ores. The depletion of these resources, where this metal is an accompanying element, causes a high share of the mineral and fossil raw materials category (50.17%). Other categories are the consequence of emissions in these processes, mainly

TABLE 2. Results of impact category indicators for the synthesis of 1 kg of zeolites, including their modification with silver nitrate after weighing stage [Pt,%]

TABELA 2. Wyniki wskaźników kategorii oddziaływania dla syntezy 1 kg zeolitów, w tym ich modyfikacji azotanem srebra po etapie ważenia [Pt,%]

Impact categories	Environmental impact per kg of product [mPt]	Share in the overall impact indicator [%]
Climate change	0.10	0.50
Ozone depletion	0.02	0.11
Human toxicity, non-cancer effects	4.05	20.44
Human toxicity, cancer effects	1.23	6.22
Particulate matter	0.66	3.33
Ionizing radiation HH	0.04	0.22
Photochemical ozone formation	0.23	1.18
Acidification	0.81	4.07
Terrestrial eutrophication	0.16	0.81
Freshwater eutrophication	1.78	8.98
Marine eutrophication	0.13	0.63
Freshwater ecotoxicity	0.53	2.69
Land use	0.03	0.16
Water resource depletion	0.10	0.48
Mineral, fossil & ren resource depletion	9.94	50.17
TOTAL	19.80	100

Source: own study based on the results obtained in the software SimaPro v8.5.2.

heavy metals into water and air that have an impact on human health, non-carcinogens (20.44% share) and human health, carcinogens (6 share, 22%). In the case of the freshwater eutrophication category, the emissions are responsible for sulfur compounds into the soil and groundwater resulting from the management of flotation waste generated at the stage of silver-bearing ore processing.

5. The results of the environmental impact of the zeolite synthesis process after the characterization stage

For a more detailed analysis, at the level of individual impact categories, the results are also presented after the characterization stage. At this stage, the results of the LCIA analysis represent indicators separately for each of the impact categories analyzed, taking the category characterization indicator specified in the model into account. Presenting them in this way allows them to

TABLE 3. Characterized impact category indicators for the synthesis of 1 kg of zeolites from hard coal and lignite fly ashes

TABELA 3. Scharakteryzowane wskaźniki kategorii oddziaływania dla syntezy 1 kg zeolitów z popiołów lotnych otrzymanywanych z węgla kamiennego i brunatnego

Impact category	Unit	Environmental impact 1 kg of zeolite produced	
		hard coal fly ash	lignite fly ash
Climate change	kg CO ₂ eq	3.360	3.368
Ozone depletion	kg CFC-11 eq	2.43E-07	2.46E-07
Human toxicity, non-cancer effects	CTUh	-7.39E-07	5.61E-09
Human toxicity, cancer effects	CTUh	-2.71E-08	-2.05E-07
Particulate matter	kg PM2.5 eq	0.001	0.001
Ionizing radiation HH	kBq U235 eq	0.051	0.053
Photochemical ozone formation	kg NMVOC eq	0.007	0.007
Acidification	molc H+ eq	0.024	0.024
Terrestrial eutrophication	molc N eq	0.026	0.026
Freshwater eutrophication	kg P eq	0.0005	0.0005
Marine eutrophication	kg N eq	0.002	0.002
Freshwater ecotoxicity	CTUe	-1.683	-2.401
Land use	kg C deficit	1.858	2.185
Water resource depletion	m ³ water eq	0.104	0.104
Mineral, fossil & ren resource depletion	kg Sb eq	3.63E-05	3.66E-05

Source: own study based on calculations made using SimPro 8 software.

be analyzed within a given category, without comparing and summing up the results of different categories (other characterization indicators). The results after the characterization stage are present in a more measurable way i.e. through the equivalent of a given emission, the impact of the system on a given environmental aspect. Table 3 presents them analogously to previous chapters, i.e. calculated for 1 kg of ash produced from hard and lignite.

Analyzing the results at the characterization level (Figs. 2–3) it can be seen that the consumption of electricity is the dominant impact in most of the impact categories (11 of 15), at various stages of synthesis. The presented results confirm the main hot spots identified at the weighted level. The total share of energy use in individual indicators of a given category ranges from 12 to 93% for zeolites from hard coal ashes and from 16 to 96% for lignite fly ash. Depending on a given environmental aspect, this impact will be affected by various emissions introduced into the environment at the level of energy production processes. In the case of individual impact categories, it has a dominant impact and the following are responsible for the impact associated with electricity use :

- ◆ CO₂ emissions in Climate change,
- ◆ SO₂ and PM2.5 dust emissions in Particulate matter,
- ◆ carbon emission-14 in Ionizing radiation,

- ◆ NO_x and SO₂ emissions in Photochemical ozone formation,
- ◆ NO_x and SO₂ emissions in Acidification,
- ◆ NO_x and NH₃ emissions in Terrestrial eutrophication,
- ◆ NO_x emissions in Marine eutrophication,
- ◆ phosphate emissions in Freshwater ecotoxicity.
- ◆ use of natural origin water in technological and cooling processes in Water resource depletion.

Another noticeable factor is the consumption of NaOH, the influence of which dominates in the photochemical ozone formation, category, constituting 88% of the category indicator for zeolite based on hard coal ash and 87% from lignite. This is due to CFC-10 (Methane, tetrachloro-) emissions during the production of NaOH. This relationship is also a significant share of the ionizing radiation category indicator and mineral, fossil & ren resource depletion, constituting about 51% of the impact.

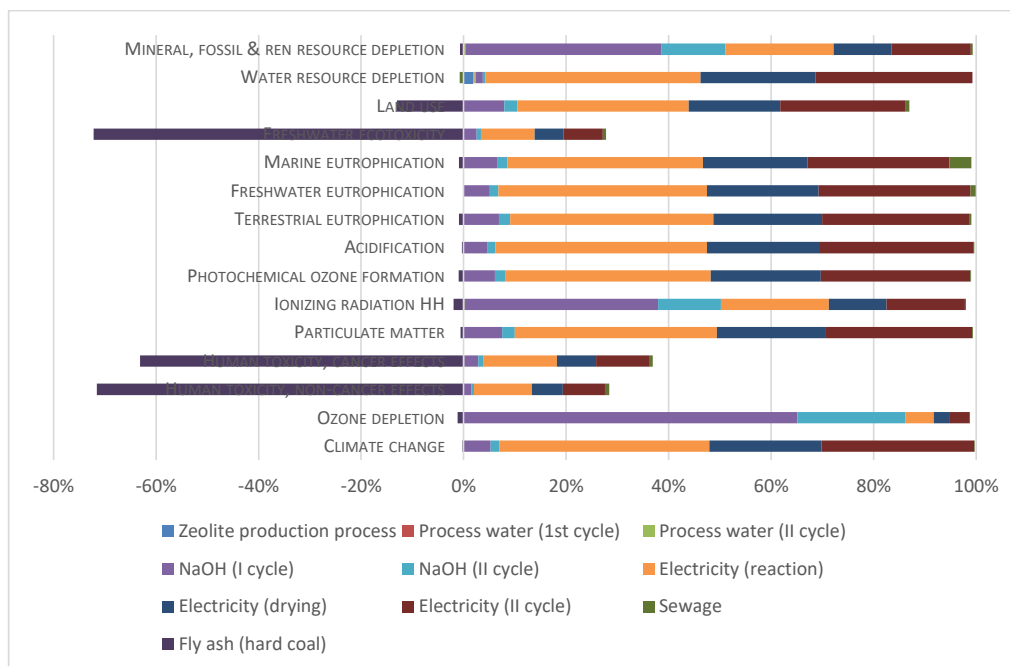


Fig. 2. Shares of individual unit processes for the synthesis of 1 kg of zeolites from hard coal ash – after the characterization stage [%]

Rys. 2. Udział poszczególnych procesów jednostkowych w syntezie 1 kg zeolitów z popiołów węgla kamiennego – po etapie charakteryzacji [%]

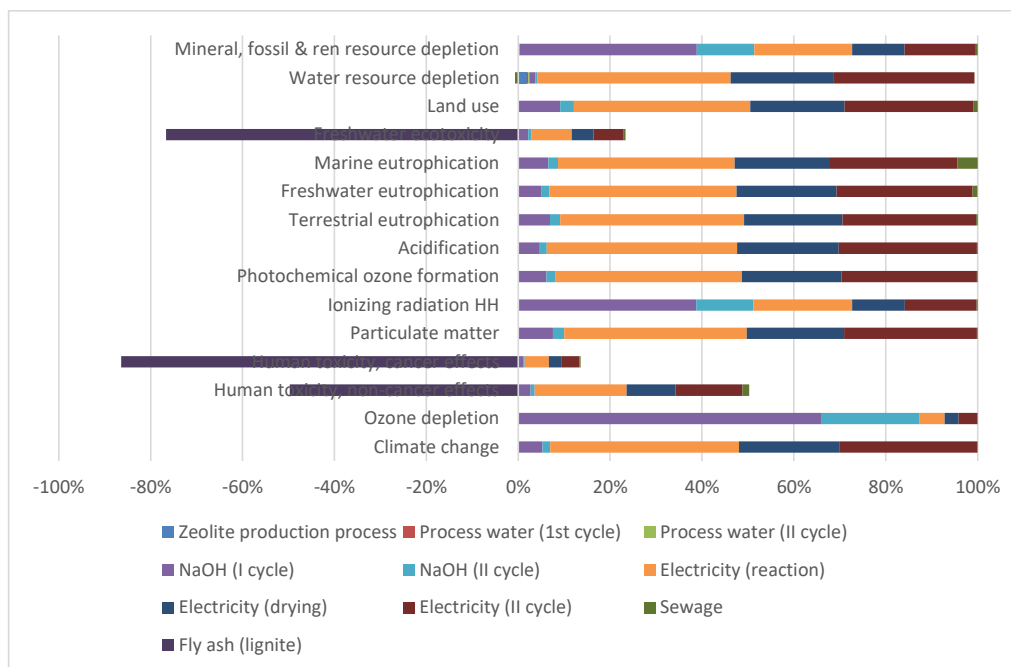


Fig. 3. Shares of individual unit processes for the synthesis of 1 kg of zeolites from brown coal ash – after the characterization stage [%]

Source: own study based on calculations made using SimPro 8 software

Rys. 3. Udział poszczególnych procesów jednostkowych w syntezie 1 kg zeolitów z popiołów węgla brunatnego – po etapie charakteryzacji [%]

Conclusions

The analysis of the environmental aspects of the zeolite sorbent synthesis and modification process allowed the following conclusions to be presented:

- ◆ The assessment of the potential environmental impact of the synthesis and modification of zeolites with silver compounds, carried out on the basis of data obtained from the semi-technical installation, showed that the total cumulative indicator of the potential impact on the environment of 50 kg material production (from two reactors and 50 kg feed) is 990 mPt, which in terms of 1 kg of product is 19.8 mPt.
- ◆ The very process of zeolite synthesis (without modification) per 1 kg of production contributes to the environmental impact of 389.70 μ Pt. Therefore, the dominant factor in the LCA analysis is the use of silver nitrate to modify the structure of the zeolites obtained. The share of this process is 99.5% of the overall impact indicator.

- ◆ The main advantage of using zeolite sorbent is the use of waste material such as fly ash, whose landfilling is still a problem. Depending on the origin of ash, various levels of environmental benefits were obtained by avoiding the landfill process. In the case of hard coal, they are at the level of $-293.20 \mu\text{Pt}$, which in relation to $389.70 \mu\text{Pt}$ of the zeolite synthesis interaction rate, is an advantage lowering it by 75.24%. In the case of ashes from lignite combustion, these benefits are even greater. At $389.70 \mu\text{Pt}$ of the impact indicator, they are $-525.04 \mu\text{Pt}$ causing its complete elimination (reduction of the index by 134.73%). This means that removing 1 kg of ash from lignite contributes to higher environmental benefits than the loads resulting from the synthesis process of 1 kg of zeolites. Thus, the net process is free from environmental impacts, contributing only to environmental benefits in the entire life cycle.
- ◆ According to the results obtained, it can be seen that the demand for electricity, at various stages of the process, plays a dominant impact. Calculated per 1 kg of material produced, this demand is at the level of 2.98 kWh and is responsible for 88.51% of the overall impact. Another factor is the use of NaOH, however, compared to electricity, its share is only responsible for 7.55% of the overall impact.

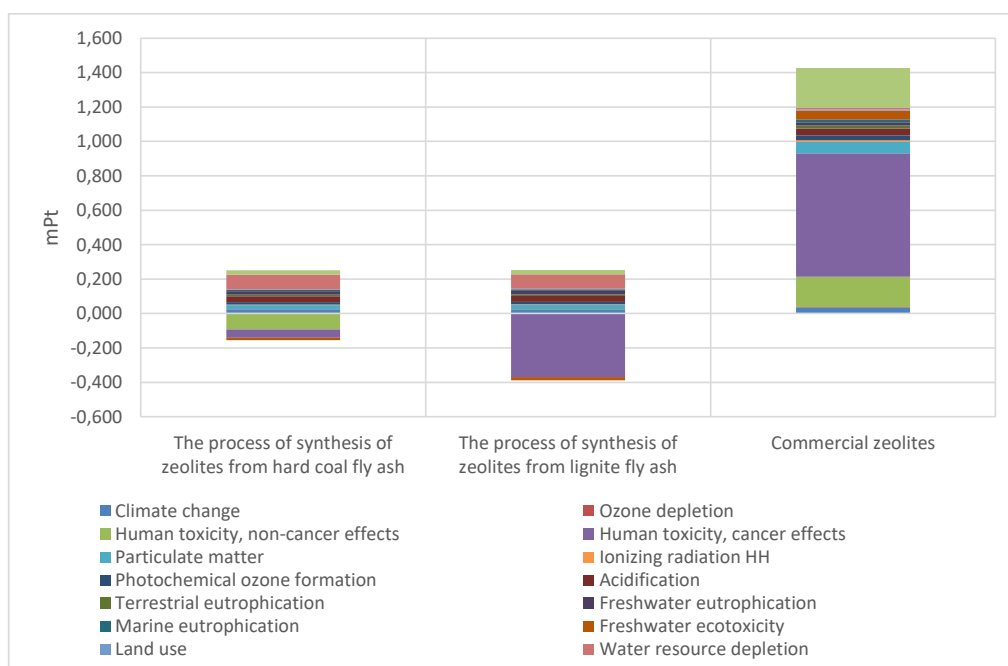


Fig. 4. Comparative analysis of the zeolite synthesis process based on hard coal and lignite fly ashes and commercially used materials

Source: own study based on calculations made using SimPro 8 software

Rys. 4. Analiza porównawcza procesu syntezy zeolitów na bazie popiołów z węgla kamiennego i brunatnego oraz materiałów komercyjnie stosowanych

- ◆ From the data presented, it can be seen that zeolites produced from ash have a significantly lower environmental impact compared to the production of 1 kg of commercial material. For zeolites based on hard coal ash it is 93.2% lower impact.
- ◆ Another advantage is its very high efficiency compared to even commercially used activated carbons (Hg⁰ sorption more than five times better than AC/Br). Comparing the production process of zeolite materials and activated carbons (Unmodified) in the production amount of 1 kg (Fig. 4), it can be seen that zeolites contribute to a lower potential impact on the environment. Zeolite materials are also suitable for regeneration, which increases their lifetime by approx. 3 times. Zeolite after sorption can be regenerated due to its good thermal stability up to a temperature of about 400°C.
- ◆ When qualitatively analyzing the structure of the process impact, a significant share of four impact categories can be weighed:
 - ◆ Mineral, fossil & ren resource depletion,
 - ◆ Human toxicity, non-cancer effects,
 - ◆ Human toxicity, cancer effects,
 - ◆ Freshwater eutrophication.

Based on the obtained results as well as the lack of efficient technology to meet BREF/BAT restrictions, the next step will be the consideration of combined methods in order to achieve EU limits. The authors take the reduction of the impact of applied new solutions onto environment into account through the application of other metals for the activation of zeolites as well as an evaluation of the practical application of the used sorbents.

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Analiza środowiskowa nowatorskich sorbentów do sorpcji rtęci

Streszczenie

Redukcja emisji rtęci w obecnie istniejących rozwiązaniach elektrowni bazujących na węglu zarówno metodami wstępnymi, jak i metodami pierwotnymi oraz metodami wtórnymi polegającymi na wprowadzeniu węgla do komory paleniskowej, a następnie usuwaniu rtęci z gazów wylotowych powstałych w procesie spalania, nie rozwiązuje problemu osiągnięcia wymaganych limitów przez elektrownie, w związku z czym istnieje potrzeba poszukiwania nowych, efektywnych rozwiązań.

Przedstawione w pracy wyniki dotyczą analizy korzyści środowiskowych dla zastosowania zeolitów otrzymywanych z ubocznych produktów spalania, jakimi są popioły lotne (z węgla kamiennego i brunatnego) w technologiach usuwania gazowych form rtęci. Badane zeolity stanowiły struktury typu X modyfikowane srebrem. Materiałem referencyjnym w rozważaniach był węgiel aktywny impregnowany bromem – komercyjnie dostępny na rynku sorbent.

W artykule rozważono korzyści środowiskowe wynikające z zastosowania badanych zeolitów uwzględniając cykl życia produktu, wydajność sorbentu oraz możliwość jego regeneracji w porównaniu do węgla aktywnego (AC/Br). Analizę LCA dokonano, uwzględniając oszacowane bilanse materiałowe i energetyczne procesów wytwarzania. Przy porównaniu procesu produkcji materiałów zeolitowych typu X na linii technologicznej oraz węgla aktywnych w ilości niezbędnej do wychwycenia z gazów odlotowych 375 g Hg, analiza LCA wykazała, iż zeolity przyczyniają się do mniejszego potencjalnego wpływu na środowisko. Zaletą jest fakt, iż do wychwycenia tej samej ilości rtęci niezbędne jest 5 razy mniej sorbentu zeolitowego niż węgla aktywnych. Ponadto materiały zeolitowe dodatkowo można regenerować, co wydłuża ich czas życia.

SŁOWA KLUCZOWE: popiół lotny, LCA, rtęć, zeolity

