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USEFULNESS OF FLY ASHES FROM POWER STATIONS IN THE TECHNOLOGY OF FINAL WASTEWATER TREATMENT

Results of investigations aiming at the examination of the usefulness of fly ashes in the technology of final wastewater treatment are presented. The investigations were performed for combined wastewater pretreated by the activated sludge method and consisting of domestic sewage and synthetic dyeing wastewaters. Fly ashes were found to be highly effective for the removal of residual colour, phosphates and detergents. High effects of COD removal were also obtained. The possibility of using fly ashes as a sorptive agent was found. Parameters of the operation of adsorption columns with an expanded bed were determined. Basing on results obtained, it was concluded that the process of colour removal using fly ash as a sorbent might be due to physical and chemical sorption. The tests conducted under dynamic and static conditions allowed to determine the degree of the desorption of impurities arrested on fly ashes.

1. INTRODUCTION

Solid wastes resulting from the coal combustion process in power stations consists of fly ashes (called also power station dusts), bottom ashes and slag.

Physicochemical characteristics of coal fly ashes depend on the properties of coal and the technological conditions of the coal combustion process.

The mean percentages of basic chemical components of three kinds of fly ashes are given in table 1 [3], [11], [12].

Besides the main component listed in table 1 the ashes may contain small amounts of TiO_3 (0.4–1.3%), barium (0.1–0.6%) and phosphorus (0.01–0.3%), as well as traces of copper, manganese, nickel, zinc, barium, mercury, vanadium, arsenic and zirconium. Fly ashes also contain radioactive elements, but the content of natural radioactive pollutants varies considerably even when the coals come from the same basin. It is known

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that the concentration of radioactive elements depends on the kind of coal rocks [2], [12].

The components of fly ashes can be generally classified among crystalline matter, non-combusted coal, silty ingredients and glassy.

The observations of the chemical composition of fly ashes have shown that the fraction of glass components and the kind of graining depend on the temperature in the furnace and on the technique of fuel preparation. The fly ashes whose mean grain size equals 2–3 μ change the colour from light-grey to black, depending on the coal content. The pH value of fly ashes ranges from 6.5 to 12.0 with an average pH of 11.

Table 1

Component	Kind of fly ashes (%)		
	silicate	silicon aluminium	silicon calcium
SiO ₂	47.9	50.9	47.2
Al ₂ O ₃	23.2	34.1	5.2
Fe ₂ O ₃	10.7	5.8	6.6
CaO	5.2	2.3	27.2
MgO	2.8	1.9	3.4
K ₂ O	2.0	2.0	2.0
SO ₂	1.3	1.2	7.2
Mass losses while roasting at a tem- perature of 900 °C	7.0	1.5	1.5

Considering the amount of furnace wastes from power stations (about 10 mln tons annually), Poland is one of main producers and the estimated area covered by fly ash dump is about 1200 ha, showing an increasing tendency [12]. In 1990 the amount of power station wastes is expected to be as high as 40–45 mln tons a year. So far according to the technologies developed in this country [11] about 20% of fly ash has been utilized, namely:

a) in the industry for production of
cellular concrete — ca. 860,000 tons (1975),
cement — ca. 675,000 tons (1975),
aggregate concrete — ca. 320,000 tons (1975),
buildings and special ceramics — ca. 80,000 tons (1975),
ash-pyrite aggregate — ca. 80,000 tons (1975),
special materials, i.e. Al₂O₃ backfills, insulating materials, fertilizers for agriculture, magnetite powder,

b) in highway engineering, i.e.
road and soil stabilization — ca. 50–70,000 tons/year,

c) in embankment engineering — ca. 630,000 tons/year.

Physicochemical composition of ashes and their physical properties suggest the possibility of their application in wastewater treatment by physicochemical methods.

Methods of final wastewater treatment are used to remove primary and secondary wastewater treatment residuals as well as secondary products of biologically treated wastewater. They include not only refractory suspended solids and organic matter expressed by BOD₅ and COD, but also non-biodegradable detergents, colour, phosphates and nitrogen compounds, as well as substances typical of each group of wastewaters.

The methods used in the technology of final wastewater treatment are based mainly on the expensive home-made or imported agents (as e.g. ion-exchange resins, synthetic, organic and inorganic sorbents, polyelectrolytes), therefore their application on large scale is still limited. Literature data, however, concerning the applicability of waste products in the technology of wastewater treatment indicate the high usefulness of various kinds of soils, peat and waste products from energy coal processing [1], [3], [5], [7], [9], [10].

2. PURPOSE OF INVESTIGATION

The main purpose of the reported research was to determine the possibility of the utilization of fly ashes in the technology of final treatment of combined wastewater containing a mixture of dyeing wastewaters and domestic sewage [13]. Other investigations on the technology of textile wastewater treatment [8] have shown that new economic methods for final treatment of this kind of wastewater should be developed. Textile wastewaters alone or combined with domestic sewage treated by biological or physicochemical methods in a conventional two-stage plant are still characterized by high contents of colour and refractory substances (detergents, dyes, assistants employed in the dyeing and finishing of textile). In this case, textile wastewater should be treated by multistage technology.

3. SCOPE OF INVESTIGATION

Investigations performed were to determine the usefulness of fly ashes in the sorption of residuals present in secondary effluent. The efficiency of colour, COD, phosphates and detergents removal was assumed to be the criterion of evaluation of the test sorbent. The range of the assumed pollution parameters allows utilization of the data obtained in treating various groups of wastewater.

The investigations conducted under laboratory conditions were comprised of laboratory tests and investigations on adsorption and desorption processes. In laboratory tests, the adsorption equilibrium capacity of the fly ashes was determined under static conditions. The tests were carried out with combined wastewater containing the mixture of 0.1% solution of Lanazol rot 5B reactive dye bath in domestic sewage, pretreated by the activated sludge method and filtered through a sand bed and with the textile wastewater discharged into the Group Wastewater Treatment Plant at Komorowice.

The process of impurities adsorption and desorption of fly ash under dynamic conditions were examined in adsorption columns with an expanded bed. These investigations were also conducted on the combined wastewater of the same composition as in the former case and pretreated by the activated sludge method.

The composition of the 100% bath of the investigated dye was as follows:

- 30 g/dm³ — reactive dye Lanazol Rot 5 B,
- 26.4 g/dm³ — Na₂SO₄,
- 20.0 g/dm³ — Na₂CO₃,
- 2 g/dm³ — Rokaphenol Wg — non-ionic surfactant.

The data concerning the composition of the dye bath and the degree of bath dyeing during the textile coloration process, obtained at Associated Wool Industries "Południe" and textile plants, allowed determination of the concentration of the investigated synthetic wastes [13].

The investigated fly ashes were taken from power station "Łaziska", their physico-chemical composition was as follows: SiO₂ — 51%, Al₂O₃ — 34%, Fe₂O₃ — 6%, CaO — 2.5%, MgO — 2% and mass losses while roasting at a temperature of 900 °C amounted to 2%. The percent of each fraction in the total mass of the ashes is presented below:

- 0.5 mm — 2–3%,
- 0.25 mm — 5–7%,
- 0.20 mm — 5–7%,
- 0.15 mm — 7–9%,
- 0.12 mm — 15–18%,
- ≤0.10 mm — 56–66%.

Laboratory tests were performed for ashes in the natural form and for those of the average grain size ≤0.1 mm, 0.12 mm, 0.15 mm and ≥0.2 mm.

The obtained values for equilibrium adsorption capacities of the fly ashes examined were evaluated with respect to the corresponding values determined for domestic granular activated carbon Z₄ and powdered Carbopol WS.

4. METHOD OF INVESTIGATION

During laboratory tests on adsorption, the optimal time for wastewater contact with sorbent and the optimal sorbent dose were determined at the constant temperature (20 °C). The values obtained allowed to determine adsorption isotherm; the latter is expressed as a ratio of the impurities removed to the unit of sorbent weight and of the concentration of impurities remaining in the solution in the equilibrium state. ECKENFELDER [4] and FORD [6] suggest the application of the Freundlich empirical equation to the interpretation of adsorption conducted on the solutions of pure substances and some diluted sewages. Therefore, the following equation was also applied to the reported studies:

$$\frac{C_0 - C}{M} = K \cdot C^{1/n},$$

where: C_0 — initial concentration of a pollutant,
 C — pollutant concentration after adsorption,
 M — sorbent quantity,
 K, n — constants;

hence we get:

$$\log \frac{C_0 - C}{M} = \log K - \frac{1}{n} \log C.$$

The relationship determined is represented by a straight line of the slope $1/n$; its intersection with the y -axis allows to record the value of constant K .

Glass pipes, 30 mm in diameter and 1500 mm high, with conical bottom, equipped with a supply tube were used as adsorption columns in the investigations conducted under dynamic conditions. The adsorption bed filled with fly ashes of the average grain size ≥ 0.12 mm was used. A metal net was used to support the fly ash bed. The height of fly ash bed was 700 mm, but after expansion it was about 900 mm. A technological continuous system consisted of 2 columns working in series. When the adsorption beds

Table 2

Effects and parameters of impurities adsorption in adsorption columns with the aerated bed of fly ashes of the grain-size 0.12 mm

Zestawienie efektów i parametrów procesu adsorpcji zanieczyszczeń w kolumnach adsorpcyjnych ze złożem spulchnionym pyłów elektrownianych o granulacji 0.12 mm

Parameter	Unit	Surface loading of beds		
		Series I		
		2.0 m ³ /m ² · h	5.7 m ³ /m ² · h	6.0 m ³ /m ² · h
Time of wastewater contact with sorbent	h	0.75	0.30	0.25
Volume of wastewater flow	dm ³ /h	1.4	4	4.2
COD of wastewater before adsorption	mg O ₂ /dm ³	53	70	80
COD of wastewater after adsorption	mg O ₂ /dm ³	24	25	30
Effect of COD removal	%	55	64	63
Effect of colour removal	%	100	100	100
Effect of detergents removal	%	88	—	—
Effect of phosphates removal	%	99	40	40
Working time of the set till the point of breakthrough	h	53	36	26
Adsorption capacity in relation to COD	mg O ₂ /g of sorbent	2.9	7.2	6.8
Adsorption capacity in relation to colour	% of colour/g	9.3	18	13.6

Set of 2 columns working in series

The height of I column packing 0.70 m

The height of II column packing 0.70 m

in the system were exhausted the first column was replaced with a second one and a column with fresh filling was used as the second column.

The adsorption columns were operated with an expanded bed, i.e. with an upward feeding of wastewater. The analytical control of the adsorption under dynamic conditions included determination of the following parameters: pH, COD, colour, detergents, phosphates, and total hardness. For that purpose the adsorption column effluents were sampled every 30–60 min. during the whole period of the system's operation. Basing on the results obtained, the concentrations of the test pollutants in effluent were plotted as a function of the total volume of wastewater that had passed through the column in a given time interval. Those relationships allowed to determine the "breakthrough" point of the II column bed and the moment of the exhaustion of the I column bed adsorption capacity. The adsorption capacity of the sorbent under dynamic conditions was then calculated.

Technological examinations were carried out for the following values of hydraulic loading of beds: 2, 5.7 and 6.0 m³/m² · h.

5. RESULTS AND DISCUSSION

The values of equilibrium adsorption capacities obtained for the examined fly ashes are given in fig. 1. A comparison of the effects of colour and COD removal during adsorption on fly ashes and activated carbon is given in figs. 2 and 3. Parameters and effects of adsorption in columns with fly ash expanded bed are presented in figs. 4 and 5.

The values of adsorption capacity of ashes varied according to their grain-size. These are as follows:

- for raw, i.e., unsieved fly ashes — 18 mg COD/g and 14% of colour/g,
- for ashes of 0.10 mm grain-size — 18 mg COD/g and 13% of colour/g,
- for ashes of 0.12 mm grain-size — 18 mg COD/g and 10.6 of colour/g,
- for ashes of 0.15 mm grain-size — 16 mg COD/g and 8.3 of colour/g,
- for ashes of 0.20 mm grain-size — mg COD/g and 10.7 of colour/g.

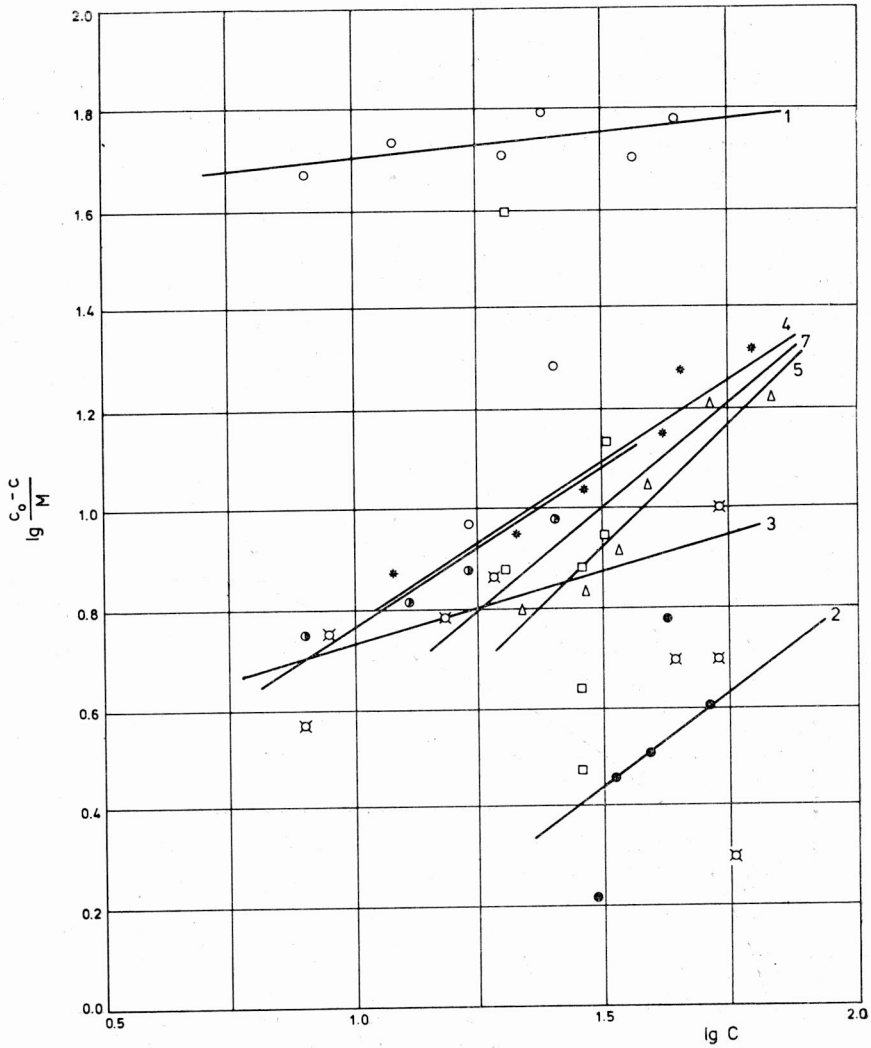
The corresponding values obtained for granular activated carbon Z₄ were equal to 6.6 mg COD/g and 6.4% of colour/g.

Adsorption capacities of fly ashes were also estimated with respect to impurities present in the textile wastewater combined with domestic sewage taken from the Group Wastewater Treatment Plant at Komornice. The pretreatment of the wastewater comprised the equalization and filtration through a sand bed. The adsorption capacity of the test ashes determined with respect to this wastewater was equal to 27 mg COD/g, 9% of colour/g and 8 mg of detergents/g.

The results obtained do not differ substantially from those of synthetic wastewater. The differences are due to different physicochemical composition of the sewages investigated.

The relationships presented in the figures allow to draw the following conclusions:

- the adsorption of residual impurities on fly ashes results in a high removal of COD and colour,



- * Powdered activated carbon WS
- Granular activated carbon Z-4
- Unsieved fly ash
- Δ Fly ash of the grain size 0.10 mm
- ⊠ Fly ash of the grain size 0.12 mm
- Fly ash of the grain size 0.15 mm
- Fly ash of the grain size ≥ 0.20 mm

Fig. 1. Adsorption isotherm estimated for COD removal
Rys. 1. Izoterma adsorpcji oszacowana dla usuwania ChZT

— the results obtained ranged from 50 to 90% of COD removal and from 65 to 100% of colour removal depending on the ash grain-size.

The best results were obtained for raw ashes and those of the average grain-size of 0.10 mm and 0.12 mm; about 100% colour removal was stated for a 7 g/dm^3 dose and about 80–90% COD removal for doses ranging between 7 and 10 g/dm^3 .

The corresponding effects of COD and colour removal obtained during adsorption on granular activated carbon Z_4 with the same sorbent dose i.e. 7 g/dm^3 were 40% and 60%, respectively.

Analysis of the relationships of the effects of COD and colour removal due to adsorption on activated carbon and fly ashes with various sorbent doses (figs. 2, 3) suggests that both physical and chemical adsorptions of a reactive dye may take place on fly ashes. This is confirmed by additional spectrophotometrical analysis performed within the UV and visible spectra ranges.

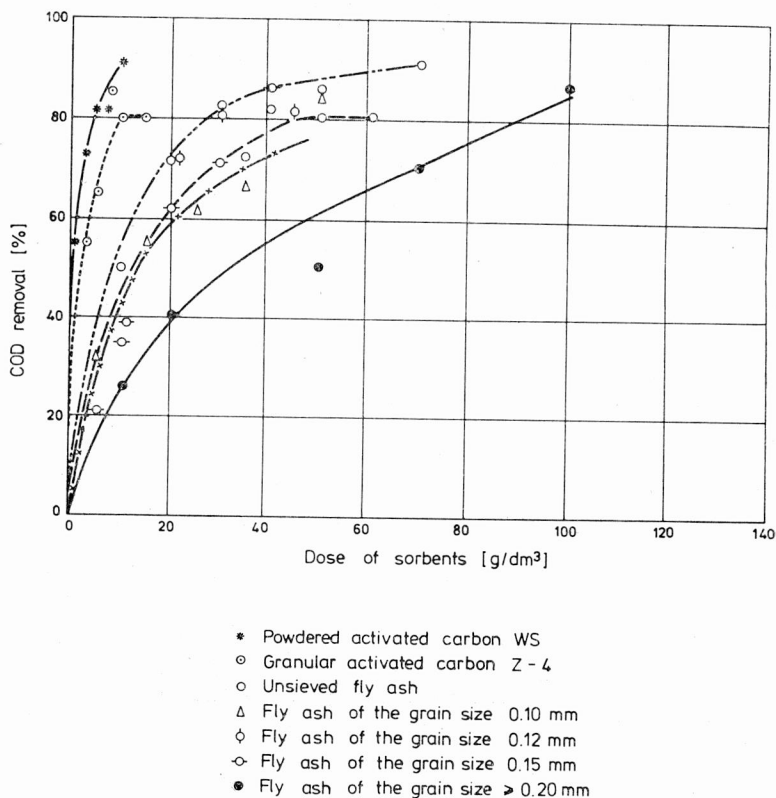
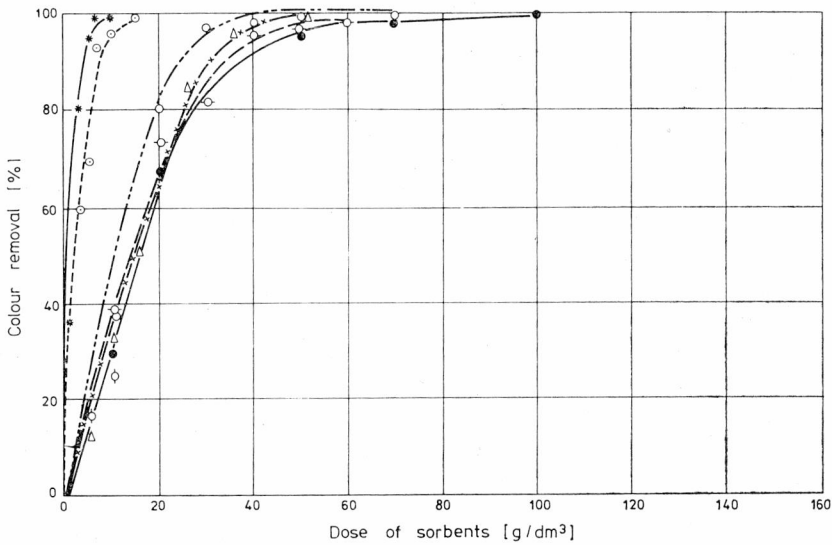


Fig. 2. Correlation between COD removal and dose of test sorbents in adsorption process of synthetic dyeing wastes: 0.01% of water solution of reactive dye — Lanazol Rot 5 B

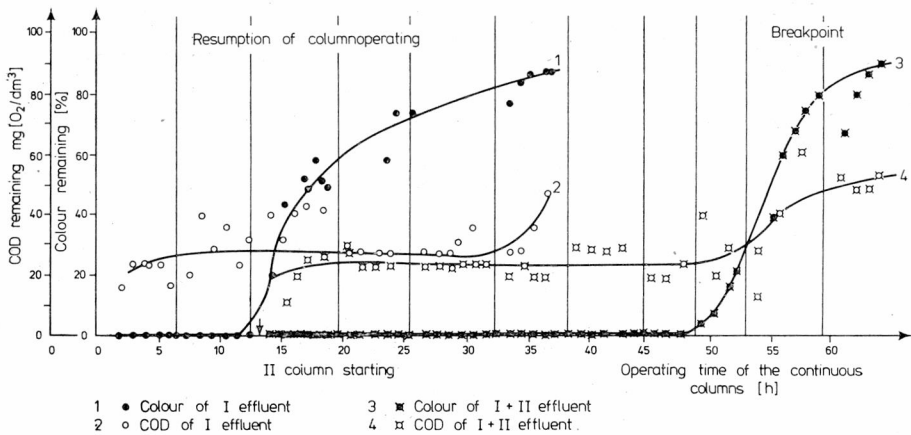
Rys. 2. Korelacje pomiędzy usuwaniem ChZT i dawką sorbentów testowych w procesie adsorpcji syntetycznych ścieków farbiarskich; 0,01% roztwór wodny reaktywnego barwnika Lanazol Rot 5 B



- Powdered activated carbon WS
- Granular activated carbon Z-4
- Unsieved fly ash
- △ Fly ash of the grain size 0.10 mm
- ◇ Fly ash of the grain size 0.12 mm
- ▽ Fly ash of the grain size 0.15 mm
- Fly ash of the grain size > 0.20 mm

Fig. 3. Correlation between colour removal and dose of test sorbents in adsorption process of synthetic dyeing wastes: 0.01% of water solution of reactive dye — Lanazol Rot 5 B

Rys. 3. Korelacje pomiędzy usuwaniem barwy i dawką sorbentów testowych w procesie adsorpcji syntetycznych ścieków farbiarskich; 0,01% roztwór wodny reaktywnego barwnika Lanazol Rot 5 B



- 1 ● Colour of I effluent
- 2 ○ COD of I effluent
- 3 ■ Colour of I + II effluent
- 4 □ COD of I + II effluent

Fig. 4. Effect of adsorption process on fly ash continuous columns (combined wastes treated by activated sludge method): hydraulic loading $2.0 \text{ m}^3/\text{m}^2 \cdot \text{h}$

Rys. 4. Efekt procesu adsorpcji na kolumnach ciągłych popiołów lotnych (ścieki połączone oczyszczane metodą osadu czynnego); obciążenie hydrauliczne $2,0 \text{ m}^3/\text{m}^2 \cdot \text{h}$

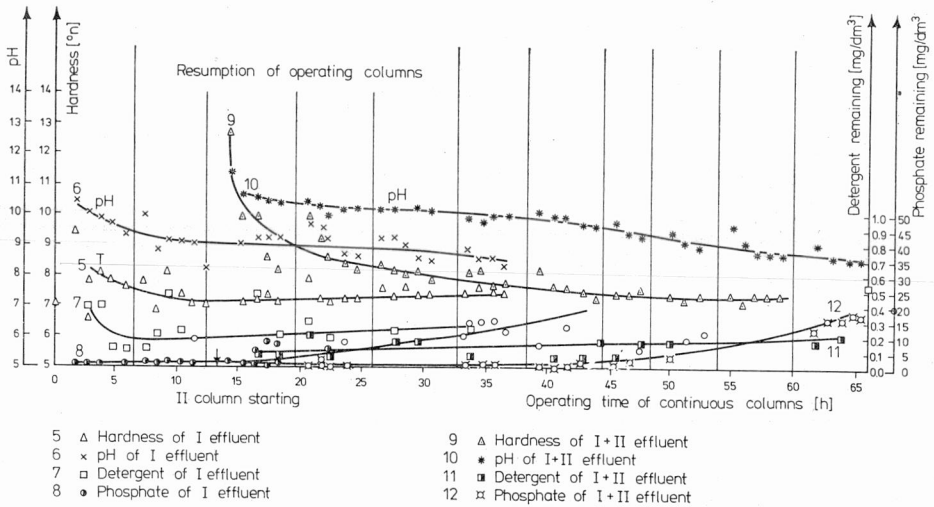


Fig. 5. Effect of adsorption process on fly ash continuous columns (combined wastes treated by activated sludge method) surface loading $2.0 \text{ m}^3/\text{m}^2 \cdot \text{h}$

Rys. 5. Efekt procesu adsorpcji na kolumnach ciągłych popiołów lotnych (ścieki połączone, oczyszczone metodą osadu czynnego); obciążenie hydrauliczne $2,0 \text{ m}^3/\text{m}^2 \cdot \text{h}$

The above statement could explain the relatively low adsorption capacities of fly ashes estimated according to the Freundlich adsorption isotherm, when compared with high effects of colour removal.

From the results of the continuous adsorption column process, it can be concluded that the process may be carried out in columns with expanded fly ash bed. High removal efficiency expressed in terms of colour and COD was found for residual impurities and three test hydraulic loading rates of adsorption bed. The effects of phosphate removal varied depending on the loading rate, the best results, i.e. 99%, being obtained at the bed loading rate of $2.0 \text{ m}^3/\text{m}^2 \cdot \text{h}$ and the contact time of about 40 min.

Considerably lower effects, i.e. 40%, were obtained at the bed loading rate of $6.0 \text{ m}^3/\text{m}^2 \cdot \text{h}$ and the contact time of 15 min. The effects of detergent removal, examined only at the loading rate of $2.0 \text{ m}^3/\text{m}^2 \cdot \text{h}$, were of the order of 98%.

During the operation of the column system the values of sewage quality parameter (pH, alkalinity, total hardness) decrease; the pH value, after the initial increase to 10, decreased to a range of 7–8, and total hardness decreased from 10° to a range of 8–9°. The test fly ashes were found then to be quite useful for removal of residual colour, phosphates and detergents.

To determine the degree of potential water pollution with the impurities arrested in the exhausted fly ashes, the desorption of impurities expressed by colour and COD was investigated both under static and dynamic conditions. It was found that the process is very slow and that only a part of the impurities is desorbed. For desorption under static

conditions the values of the impurities desorbed, calculated for 1 g of ashes, amount to 1–1.5% of colour/g and 0.5–1.0 mg of COD/g, while for desorption under dynamic conditions they amount to 0.25% of colour/g and to 0.5 mg COD/g [23]. This means that only 8% of COD load and only 1.5% of colour load were desorbed.

6. CONCLUSIONS

Technological investigations and tests conducted to determine the efficiency of fly ash adsorption used in the technology of final treatment of combined wastewater containing dyeing wastewater have allowed to conclude:

— a high usefulness of the test ashes for the removal of residual colour, phosphates and detergents as well as for lowering the level of residual COD;

— the feasibility of using continuous adsorption columns with expanded bed of fly ashes. Fly ashes of the average grain-size ≥ 0.12 mm were used as a sorbent bed. At the hydraulic loading rate of bed equal to 2 and 6 $\text{m}^3/\text{m}^2 \cdot \text{h}$ a complete removal of residual colour, 50%–80% (depending on loading rate) removal of COD, 98% removal of detergents, as well as phosphate removal ranging from 10% to 99% were obtained;

— colour removal by fly ash adsorption results probably from physical and chemical sorptions (ion exchange);

— about 10% of impurities adsorbed on fly ashes are desorbed under conditions of the periodic contacts of ashes with water environment. When the flow of water through the ash bed is continuous, the desorption of impurities is very slow and their concentration is equal to about 10% of their concentration before adsorption, i.e. to about 0.5 mg of COD/g and about 0.25% of colour/g are desorbed;

— dumping of exhausted fly ashes seems to be possible suggested; however the stabilization of the adsorbed impurities by introducing an agent able to react with the adsorbed substances should be also considered. The applicability of the fly ashes either to the chemical conditioning of sewage sludge or to the stabilization of areas dewatered by the industry seems worth examining;

— the necessity for further investigation on the kinetics of colour removal by fly ashes and the methods for the disposed fly ashes utilization has been justified.

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PRZYDATNOŚĆ PYŁÓW ELEKTROWNIANYCH W TECHNOLOGII KOŃCOWEGO OCZYSZCZANIA ŚCIEKÓW

Praca zawiera wyniki badań, których celem było określenie możliwości wykorzystania pyłów elektrownianych w technologii końcowego oczyszczania ścieków. Badania przeprowadzono na oczyszczonych w procesie osadu czynnego ściekach mieszanych stanowiących mieszaninę ścieków bytowo-gospodarczych i syntetycznych ścieków farbiarskich.

Badania wykazały istotną przydatność pyłów elektrownianych w procesie usuwania resztkowej barwy, fosforanów i detergentów. Uzyskano również znaczne efekty usunięcia ChZT. Stwierdzono przydatność pyłów elektrownianych jako środka sorpcyjnego. Określono parametry pracy kolumn adsorpcyjnych ze złożem spulchnionym.

Uzyskane wyniki sugerują, że proces usuwania barwy na pyłach elektrownianych jest prawdopodobnie wynikiem procesu sorpcji fizycznej i chemisorpcji, czyli wymiany jonowej. Na podstawie badań testowych określono stopień desorpcji zanieczyszczeń zatrzymanych na pyłach w warunkach statycznych i dynamicznych.

DIE NUTZUNG VON FLUGSTAUB AUS ELEKTRIZITÄTWERKEN IN DER WEITERGEHENDEN ABWASSERREINIGUNG

Die vorliegende Arbeit hatte zum Ziel, die Anwendung von Flugstaub aus Elektrizitätswerken zur weitergehenden Abwasserreinigung zu überprüfen. Untersucht wurde ein Mischabwasser welches sich aus kommunalem Abwasser nach dem Belebungsverfahren und aus synthetischem Färbereiabwasser zusammensetzte.

Die hier dargestellten Untersuchungen haben die grundsätzliche Anwendbarkeit von Flugstaub zur Eliminierung von Restfarbstoffen, Phosphaten und Detergentien erwiesen. Im selben Verfahren wurde auch der CSB wesentlich gesenkt. Flugstaub kann somit als ein Sorptionsmittel angesehen werden. Bestimmt wurden Arbeitsparameter von Adsorptionskolonnen mit loser Schüttung.

Die bisher bekannten Ergebnisse lassen vermuten, daß die Entfärbung mittels Flugstaub sowohl auf physikalische wie auch auf chemische Sorption zurückzuführen ist. Die Desorption wurde sowohl im statischen wie im dynamischen Prozeßverlauf getestet.

ПРИГОДНОСТЬ ПЫЛЕЙ С ЭЛЕКТРОСТАНЦИЙ В ТЕХНОЛОГИИ ОКОНЧАТЕЛЬНОЙ ОЧИСТКИ СТОЧНЫХ ВОД

Работа содержит результаты исследований, целью которых было определение возможностей использования пылей с электростанций в технологии окончательной очистки сточных вод. Исследования произведены для очищенных в процессе активного ила смешанных сточных вод, представляющих собой смесь хозяйственно-бытовых сточных вод и синтетических красильных сточных вод.

Исследования показали существенную пригодность пылей с электростанций для процесса удаления остаточной окраски, фосфатов и детергентов. Были достигнуты также значительные эффекты удаления ХПК. Отмечена пригодность пылей с электростанций как сорбционного средства. Определены параметры работы адсорбционных колонн с разрыхленным слоем.

Достигнутые результаты позволяют предполагать, что процесс удаления окраски на пылях с электростанций является, вероятно, результатом процесса физической сорбции и хемисорбции, т. е. ионного обмена. Определена степень десорбции загрязнений, задержанных на пылях, на основе экспериментальных испытаний в статических и динамических условиях.