INTRODUCTION

Rapid industrial development of humanity is accompanied by occurrence of the whole range of serious environmental problems related to surface and ground water pollution, accumulation of municipal solid and industrial wastes and the atmosphere and soil contamination. Therefore, studies in the area of prospective methods of biological (Blyashyna et al., 2018; Malovanyy et al., 2018; Malovanyy et al., 2019; Tymchuk et al., 2020; Vankovyñh et al., 2021), reagent and membrane (Tulaydan et al., 2017; Shmandiy et al., 2017) treatment of municipal and industrial waste water (Malovanyy et al., 2016a), collection stabilisation and utilisation of cyanobacteria (Nykyforov et al., 2016; Malovanyy et al., 2016b), and modern approach to utilization of municipal solid wastes (Vambol, 2016; Vambol et al., 2016) have become considerably relevant. Poultry farming which has recently been characterised by rapid production development tendency (Tertychna et al., 2015; Tereshchenko et al., 2011) is also accompanied by a range of negative consequences for the environment. These consequences may include the atmosphere pollution with emissions of dust and toxic gases, generation of huge volumes of waste water containing hazardous contaminants (xenobiotics and ammonia ions), accumulation of solid wastes (manure and other poultry metabolic by-products), microbiological contamination of the environment and deterioration of the epizootic situation as a consequence, withdrawing considerable amounts of agricultural lands for poultry farms and their infrastructure, and biodiversity decline (Warner et al., 2017; Behera et al., 2013).

Deterioration of the environmental safety in the area of intensive industrial poultry farming requires development of a complex measures system for reducing this environmental danger that would simultaneously provide sanitary and hygienic requirements for poultry maintenance. Such environmental danger reduction strategy from poultry farming complies with the provisions of the EU
which guarantees food safety in accordance with «From Farm to Folk» strategy (adopted in 2002 at the Food Safety Conference in Geneva). This strategy comprises environmental analysis environmental assessment in the field of poultry farms activity: quantitative and qualitative analysis of the environmental pollution and its impact on all components of the ecosystem. The final and most important part of the strategy is development and implementation of a system of technical and organisational measures that would enable achieving environmental safety of poultry farming. From this perspective, solution to the task of environmentally safe disposal of solid wastes from a poultry farm is considered from position of developing innovation technology of maintaining and using chicken manure, implementation of which would also enable to reduce the level of environmental danger in the area of poultry farming impact and provide additional competitive advantages for agribusiness, meeting the international environmental agricultural requirements.

One of the most dangerous contamination factors of the atmosphere and hydrosphere in the field of poultry farms impact is emission of ammonia (Philippe et al., 2011; Ghaly et al., 2013). Since ammonia emission occurs at each step of poultry growth, measures on reduction of emissions must be complex as well – throughout the entire nitrogen life cycle from bedding in poultry yard to applying manure in soil. A prospective way would be to use natural adsorbents throughout the entire natural adsorbents nitrogen life cycle (Kanda et al., 2016; Kanda et al., 2017) with the help of which it is possible to fix free ammonia (gaseous phase) and ammonia ions (liquid medium) in them. Advantages of natural sorbents are their availability, low cost, high adsorption properties for ammonium and ammonia ions, experience of their use in field-crop cultivation for applying micro- and macroelements as well as improvement of the soil structure. Studies of the researchers (Rekha et al., 2000; Poznyak 2016) have revealed that under conditions of NPK doses meeting the environmental safety requirements during the application of manure in soils, soil productivity increases with its agrochemical properties improvement, including the rise of microbiological activity and enhancement of water absorption capacity, buffering and other indicators defining soil class. Application of organic-mineral fertilizers containing sorbents is of special interest as one of the prospective methods to ensure environmentally safe and resource-saving technology of crops cultivation.

The purpose of this study is to propose a strategy for minimizing environmental danger from pollution by poultry farms with the help of adding a mixture of natural dispersed sorbents to the mixture and to use the resulting material as a fertilizer of prolonged action in the future.

MATERIALS AND METHODS

The following natural sorbents were used for the studies: natural zeolite (the main component ensuring adsorption properties is mineral clinoptilolite) and palygorskite.

As natural zeolite was used clinoptilolite that was mined from Sokyrnytsia Deposit (southeastern part of the Transcarpathian Inner Trough) with balanced deposits of 126,1 million tons. It is considered high-silicone zeolite with the silica to alumina ratio from 3,5 to 10,5 and contains about 60% of silicone dioxide. Variability of Si/Al ratio and the composition of exchangeable cations is observed depending on a deposit.

The mineral composition of zeolite is represented in the following way: clinoptilolite – 60–90%, quartz and feldspar – 6–7%, clay minerals – 2–6%, plagioclase – up to 2%.

The clinoptilolite composition meets the following formula:

\[
0,2\text{Na}_2\text{O} \cdot 0,26\text{K}_2\text{O} \cdot 0,43\text{Ca} \cdot 0,2\text{Mg} \\
\cdot 9,57\text{Si}_2\text{O}_5 \cdot \text{Al}_2\text{O}_3 \cdot 0,09\text{Fe}_2\text{O}_3
\]

The chemical composition of zeolite (mass fraction, %) is: SiO\text{2} – 70.21; Al\text{2}O\text{3} – 12.27; Fe\text{2}O\text{3} – 1.2; FeO – 0.55; Ti\text{2}O\text{3} – 0.14; MnO – 0.073; P\text{2}O\text{5} – 0.033; K\text{2}O – 3.05; Na\text{2}O – 1.77; SO\text{3} – 0.10; CaO + MgO – 10.604.

For 0.5÷1 mm fraction, true zeolite density equals 2,38·10\text{3} kg/m\text{3}, bulk volume – 1,04 cm\text{3}, bulk density or volumetric density – 0,93·10\text{3} kg/m\text{3}; specific surface – 653 m\text{2}/g; pore volume by benzol – 0,181·10\text{3} m\text{3}/kg, by water – 0,33 · 10\text{3} m\text{3}/kg; pore diameter by benzol – 11 nm, by water – 20 nm.

Clinoptilolite has grinding 2,4%, abrasion capability – 0.32%, layer porosity – 53–60%, granule porosity – 25%. It is very resistant to dehydration and heat-resistant to 700°C air temperature.

Palygorskite from Dashukivske Deposit of the Cherkasy Region was also used in the study.
deposit is one of the largest not only in Ukraine but in the world. Its uniqueness is due to not only enormous deposits (18 million tons), but also to the concentration of five layers located by the nature layer-by-layer horizontally or under slight slope of minerals or their mixes with adsorption properties. The third layer contains predominantly palygorskite – (95–87%). Palygorskite refers to low-swelling natural adsorbents. Due to the substitution of silicon part on aluminium stoichiometrically, the exchange complex of large cations of potassium, calcium and magnesium, which are part of the gross chemical composition, is manifested. Total exchange capacity for all palygorskites is 20–30 mg-eq./100 g of adsorbent.

The mineral composition of palygorskite is represented by 85–97% palygorskite and admixtures of calcite, quartz and manganese hydroxides. By the mechanical composition, the rock is composed of the dispersed phase: fraction less than 0.01 mm – 98.8 – 99.2%; 0.01 ÷ 0.1 mm – 0.1 – 3.1%; 0.1 ÷ 0.25 mm – 0.4–4.2%; more than 0.25 mm – 0.2 – 4.0%. Large fractions contain comparatively limited complex of minerals (light part: quartz, opal, feldspar, mica; heavy part: ilmenite – around 80%).

Palygorskite is natural mineral of the banded-porous structure with the general formula: (Mg\(_{8.5}\)\(\text{Fe}_{0.8}\)\(\text{Al}_{1.4}\))\(\text{(Si}_{2.7}\)\(\text{Fe}_{0.3}\)\(\text{Al}_{0.8}\))\(\text{O}_{20}\)\((\text{OH})_{2}\)(\text{OH})\(3,15\times4.3\text{H}_{2}0\times\text{K}_{0.22}\text{Ca}_{0.02}\text{Mg}_{0.17}\).

The average chemical composition of palygorskite (mass fraction, %) is the following: \(\text{SiO}_{2}\) – 50.65; \(\text{Al}_{2}\text{O}_{3}\) – 11.97; \(\text{Fe}_{2}\text{O}_{3}\) – 7.45; \(\text{TiO}_{2}\) – 0.2; \(\text{MgO}\) – 7.75; \(\text{MnO}\) – 7.75; \(\text{CaO}\) – 0.14; \(\text{H}_{2}\text{O}\) – 10.56; \(\text{H}_{2}\text{O}\) – 9.72; \(\text{Na}_{2}\text{O} + \text{K}_{2}\text{O}\) = 0.56.

Palygorskite density (by water) is 2700 kg/m\(^3\), and density (by benzol) is 2600 kg/m\(^3\). Specific (general) pore volume is 0.6×10\(^{-3}\) m\(^3\)/kg, primary pore (micropore) volume is 0.015×10\(^{-3}\) m\(^3\)/kg, specific surface is 300 m\(^2\)/g and bulk density is 520 kg/m\(^3\). Mechanical stability is not less than 90%. Natural moisture content varies within 60–75%.

Palygorskite belongs to minerals composing of pyroxene chains paired in bands. Adjacent bands are joined along the bases of tetrahedrons making their peaks pointed in the opposite directions in each band. This results in formation of a space or a channel repeating after each next band in the same direction, located strictly parallel to the band.

Palygorskite is characterised by high adsorption capacity, provided by zeolite channels 0.37×0.64 and 0.56×1.1 nm in size (primary pores), which are located in crystals and form small part of the band. Sometimes the bands are strongly combined with each other, forming corn-shock structure. By packing, the bands create pores of a different size up to 200 ÷ 300 nm long with mean cross section of 0.27 nm (secondary pores).

A scheme of experimental unit used for conducting the study aimed at determination of the optimum conditions and an appropriate ratio of the components in a mixture of natural mineral sorbets and chicken manure is shown in Fig. 1.

The study was performed using unit that consists of a reaction flask with Atinan At-A850 compressor connected and Drechsel bottles. 10 ml of sulphuric acid (0.5 mol/dm\(^3\)), 5 drops of methyl red and 100 ml of distilled water were put into the Drechsel bottles. After certain time intervals (each 5, 15, 30, 60 minutes), one Drechsel bottle was replaced with another, and the amount of sulphuric acid taken for neutralisation of distilled ammonia was analysed by backward titration using sodium hydroxide solution with the concentration of 1 mol/dm\(^3\) (1 n).

Sample mixtures were equilibrated for several hours. This time period was sufficient to determine the steady-state concentration of ammonia in sorbent and air medium of the flask. Based on the study results, the weight of ammonia absorbed by sorbent or the studied composition was calculated.

Kinetics of the ammonia adsorption process was studied with the help of the following technique: 250 cm\(^3\) flasks were filled with 10 g batches of air-dry sorbent and 5 cm\(^3\) of ammonium solution.
solution (25% wt.). The purpose of the experiments was to determine and compare the capacity of natural sorbents for absorbing ammonia from 5 cm³ of ammonium solution (25%) in different time range. 250 cm³ flasks containing sample mixtures were preliminarily equilibrated for 0.5 hour (flasks 1–3) and 7 days (4–6 flasks).

Since the primary objective of the study was to determine the optimum conditions in order to minimise ammonia emission into the environment, we analysed not time dependence of ammonia adsorbed by natural sorbents (applied to the bedding composition) but change in time of so called «ammonia slip» – ammonia amount released from bedding into the environment polluting it.

The ammonia content in the fluid mixture was determined by a weight titrimetric analysis, and the weight fraction of moisture was determined by drying to the constant weight in a drying oven.

The experimental studies were conducted in order to determine the optional ratio of a mixture of natural mineral sorbents for efficient absorption of ammonia. For this purpose, we prepared batches in the following weighing ratios: 0 g of palygorskite : 10 g of clinoptilolite; 0 g of clinoptilolite : 10 g of palygorskite; 1.5:8.5; 3:7; 5:5; 7:3; 8.5:1.5 g of the sorbents, respectively. The studies were conducted with the help of aforementioned method.

Further studies were conducted in order to determine the optimum ratio of the sorbent mixture to chicken manure. For this purpose, batches were prepared in the weighing ratios 10 g of the mixture: 50 g of manure (1:5); 10 : 60 (1:6); 1:5.5; 1:4; 1:3.5; 1:3; 20 : 50 (1:2.5), respectively. The studies were performed with the help of the aforementioned method. The study results are presented in Table 1.

In order to study an impact of the drying temperature on mechanical resistance of granules of the proposed composition of organic fertiliser, this composition (a mixture of sorbents in a certain proportion mixed with raw chicken manure at the studied ratio) in form of bricks was placed into a special mould with 15×15×15 mm cells. The obtained samples were kept for 24 hours to achieve a fixed form (Fig. 2).

The formed cubes were dried in two ways: in a drying oven to the constant weight during 6 hours at the temperature corresponding to the study temperature, and under a vent hood during 24 hours at 20°C.

Mechanical resistance of the granule samples to compression was determined using a universal press UMM-5 with maximum load of 50 kN, which is designed for tension, compression and bending tests.

RESULTS

The prospects from use of natural sorbents for adsorption of free ammonia from bedding in poultry houses were studied for the purpose of their further application in production of long-acting organic-mineral fertilisers. Preliminary studies (Kanda et al., 2016; Kanda et al., 2017) have shown that palygorskite and clinoptilolite are the most prospective natural sorbents to achieve this goal. This is why these two sorbents were used in the research. It was necessary to determine the optimum ratio by an exploratory way to derive beneficial properties of both sorbents. Their application to chicken manure will contribute to ammonia absorption from manure as well as reduction of moisture in bedding prior to the beginning of the granulation stage. This will help to fix nitrogen in the exchangeable form and reduce its loss.

The results of experimental studies on determining the optimum proportion of a mixture of natural mineral sorbents for effective ammonia absorption are presented in Figure 3. To obtain reliable results, the experiments were repeated 3 times, and the average value of the parameter was chosen to plot. The relative error of the constructed dependencies did not exceed 5%, which confirms the correctness of the obtained results.

Fig. 2. General view of the organic fertiliser composition in 15×15×15 mm bricks form.
According to the results of the study, the highest capacity (and the lowest amount of ammonia slip, respectively) was shown by the composition at the ratio of 1:1 (5 g of clinoptilolite: 5 g of palygorskite).

The results of studies to determine the optimum ratio between the mixture of sorbents and chicken manure are shown in Table 1.

To study the optimum conditions for granulation of litter containing sorbents at the pre-defined optimum ratio, bricks were formed using the aforementioned method. The results of the study on the bricks of the proposed mixture for the weight moisture fraction and ammonium nitrogen content are presented in Figure 4. The initial value of relative moisture before drying was 60%, and the initial ammonium nitrogen content was 3.2%. The process of drying is accompanied by loss of ammonium nitrogen and free moisture.

To obtain reliable results, the experiments were repeated 3 times, and the average value of the parameter was chosen to plot. The relative error of the constructed dependencies did not exceed 5%, which confirms the correctness of the obtained results.

Significant reduction was observed of the moisture content in the studied composition – the moisture weight fraction in the final product was 51% to 28%.

### DISCUSSION

During drying process of chicken manure granules at $T=105\,^\circ C$ harsh, strong foul odour was observed enhancing throughout the test. Immediately after adding the sorbents the foul smell disappeared and did not occur with further drying of the granules with the sorbents at different temperatures (80–140°C). This can be explained by following: moisture from poultry manure was

**Table 1.** Determination of the optimum ratio of mixture of the natural dispersive sorbents to bedding. The highest capacity (and the lowest amount of ammonia slip, respectively) was shown by the composition «sorbents: chicken manure» at the ratio of 1:5. It makes about 1,56 mg-eq/g of sorbents.

| Ratio of palygorskite + clinoptilolite (1:1) mixture to bedding | Weight of absorbed ammonia, mg-eq/g of sorbents |
|---------------------------------------------------------------|-----------------------------------------------|
| 1:6                                                           | 0.92                                          |
| 1:5.5                                                         | 1.3                                           |
| 1:5                                                           | 1.56                                          |
| 1:4                                                           | 1.5                                           |
| 1:3.5                                                         | 1.1                                           |
| 1:3                                                           | 0.84                                          |
| 1:2.5                                                         | 0.34                                          |

![Fig. 3. Dependence of specific amount of ammonia slip on a type of mineral sorbent.](image3.png)

![Fig. 4. Changing dynamics of the moisture and ammonium nitrogen content in granules during drying process in the oven.](image4.png)
actively absorbed by the sorbents; therefore, microorganisms could not spread and release new portions of ammonium. At the same time, absorbed ammonia was also adsorbed strongly enough at the active centres of minerals, and its desorption did not occur in the specified temperature range.

The results of the conducted experiments analysed demonstrate that drying must be performed at minimum drying temperatures 20–25 °C in order to prevent ammonia loss. Drying at this temperature results in the ammonium nitrogen content 0.83–0.86%. In order to achieve the required moisture content of a granule (∼30%), drying must be performed in a filtering layer using dehumidified air as heat medium.

In order to prevent the negative impact of poultry houses on the environment, research is being conducted in every country of the world: from effective management to produce energy or value-added products (Dróżdż et al., 2020) to finding optimum technologies for processing chicken manure. Researchers pay considerable attention to the factors influencing the processes of adsorption and emission of ammonia by mineral sorbents. In particular, (Li et al., 2010) determined the rate of adsorption by clinoptilolite depending on the moisture content (in percent from 20 to 60%) in the manure of pigs on a livestock farm. The development of manure processing technologies by granulating poultry manure to reduce odor and slow release of the target component (nitrogen) has received attention (Purnomo et al., 2017). According to the report (Li et al., 2020) low-temperature drying of manure by poultry heat helps to reduce ammonia emissions from manure, and the higher the drying temperature, the greater the total nitrogen loss.

This coincides with the results of the research presented in this article.

The development of the basic technological scheme of production of granular organic–mineral fertilizer on the basis of chicken manure is based on the task of determining the optimum parameters for acceptable sizes, density and humidity of granules, ecological safety of production. This solves several problems—processing of the accumulated solid waste of the poultry farm and prevention of soil contamination, adsorption of ammonia and prevention of air pollution, prevention of hydrosphere pollution by wastewater highly concentrated by ammonia from the storage of litter from is valuable for agricultural development.

Based on the obtained research results, the basic technology of making a composition of organic–mineral granular fertilizer of prolonged action from the material of the poultry farm litter, which contains a mixture of clinoptilolite and paligorskite in a ratio of 1:1 and chicken manure. The proportion is maintained in the composition (mixture of sorbents: chicken manure) = 1:5. The introduction of these natural dispersed sorbents will contribute to the effective reduction of moisture to the granulation stage, as well as the adsorption of ammonia from the manure. The structural diagram of the obtaining fertilizers of prolonged action based on chicken manure is shown in Figure 5.

The basic technological scheme of production of organo-mineral fertilizers of prolonged action will consist of the following stages:
1. Equalizing bedding material in an equalizing mixer to achieve uniformity.
2. Granulating in a closed screw granulator to granules 4–6 mm in diameter and 15–20 mm long.
3. Drying of granules to humidity ≈ 30% in the unit of filtration drying at a temperature T = 25°C;
4. Packing of commercial fraction.

The basic technological scheme of the unit on bedding material utilization, which includes chicken manure and a mixture of sorbents, to obtain an effective organic-mineral fertilizer of prolonged action is shown in Figure 6.

Bedding material is delivered to a feed hopper 1 with a tractor trailer and loaded to a receiving section. Feather, shell, small rocks and lime over 1 mm in size impede the process of granule formation and drying. This can cause induced and
lasting delays in the process because of a necessity to clean the consistence from these solid impurities. Therefore, the process flow is provided with an equalising mixer 3 intended for grinding impurities and equalising the bedding material composition. Bedding material is supplied to the equalising mixer 3 by a screw feeder 2.

Mixture from the equalising mixer 3 is directed to the closed screw pelletizer 5. Granules are formed of a commercial size 4–6 mm in diameter and 15–20 mm long, allowing for their dispersion with a seeder. The moisture content of the proposed composition after granulation is ≈ 50%. Chicken droppings are naturally sticky. So, in order to provide powdering and prevent the produced granules from caking, we suggest adding milled natural zeolite from a zeolite hopper 6 at the stage of drying. At the same time, excessive zeolite adsorbs free ammonia released in proceed of filtering drying, ensuring ecological cleanness of the production and preventing from pollution of the environment with this ammonia.

The drying stage takes place in a dryer in filtering drying mode with the air heated to 25°C in an electric air heater equipped with an air drying unit 4. Condensation resulted from drying is taken away from the electric air heater 4. Granules of organic-mineral fertilizer are supplied to a filtering dryer 7, where they are dried to the moisture content ≈ 30% in filtering mode. Exhaust air is supplied to a cyclone 8, where it is purified from dust. Dried air is released into the atmosphere, and entrapped dust is returned to the drying stage, where it is added to milled zeolite used for powdering granules of fertilizer.

The obtained dry granules are sieved from the crushed particles on a vibrating screen 9 and sent to the packing apparatus 10, where they are packed in plastic bags. The bags are delivered by truck to the warehouse of finished products. From the bags are formed batches of fertilizers, which are sent to consumers.

The proposed strategy of minimizing environmental safety from environmental pollution by poultry farms has a clear economic advantage. On the one hand, reducing the level of ammonia emissions into the atmosphere reduces the level of environmental pollution by the enterprise, and thus reduces the level of penalties for environmental pollution. On the other hand, the obtained fertilizers of prolonged action are a valuable innovative product, the use of which in agriculture will increase the efficiency of crop production and at the same time reduce environmental pollution through better absorption of nitrogen by plants.

**CONCLUSIONS**

To determine the optimum conditions for the implementation of the ammonia adsorption process from chicken manure, the kinetics of the ammonia adsorption process was studied, the influence of air temperature on the ammonia adsorption process was determined, the dependence of sorption on the type of mineral sorbent was determined.

![Fig. 6. Principal technological scheme of the unit on chicken manure utilization with obtaining effective organic-mineral fertilizer of prolonged action](image-url)
According to the experimental data, the process of adsorption runs most efficiently in the medium at $T = 20$ °C. Nevertheless, ambient temperature within $10\pm 25$ °C apparently has no meaningful impact on the adsorption process. The weight of adsorbed ammonia varies within $8.17\pm 13.26 \times 10^{-2}$ g of NH$_3$/10g of sorbent.

An analysis of the results of the experimental studies showed that the highest capacity (and the lowest amount of ammonia slip, respectively) was demonstrated by the composition of clinoptilolite and palygorskite at the ratio of 1:1. The highest capacity for ammonia adsorption was shown by the composition «sorbents: chicken manure» at the ratio of 1:5. It makes $1.56 \times 10^{-2} \text{mg-eq/g}$ of sorbents.

The mechanical compressive strength of granules of organic–mineral fertilizer composition was investigated. The results showed that the static compressive strength of the granules increases with decreasing moisture content of the granules. Therefore, in the technological mode it is necessary to maintain the lowest possible final humidity of the granules, at which there is no loss of adsorbed ammonia.

The results of the conducted experiments analysed show that drying must be performed at minimum drying temperature values $20–25$ °C in order to prevent from ammonia loss. With drying at this temperature under filtration conditions, relative moisture of granules of organic-mineral fertilizer will be $\approx 30\%$, and the ammonium nitrogen content will be $0.86\%$.

Based on the analysis of these studies, a basic technological scheme for the production of organic–mineral fertilizers based on chicken manure and a mixture of natural adsorbents is proposed. The developed technology of poultry farm waste utilization is low-waste, resource- and energy-saving, and environmentally safe. The granular organic–mineral fertilizers of the prolonged action received on this technology are effective for use in agriculture.

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