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Radiation stability of lindane and active substance of hexachlorane dust preparation under electron irradiation

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Abstract. Electron beam impact on the radiation resistance of organochlorine hydrocarbon is considered for two forms of pesticide – a pure chemical compound and a component of active substance of a pesticide product to be disposed of. State standard reference samples (SRS) of hexachlorocyclohexane (HCH) – lindane and hexachlorane dust – are used. The fullness of decomposition are determined for lindane and organochlorine compounds as part of the active substance, depending on the aggregate state of the preparation and the absorbed dose of electronic radiation. In all samples, a reduction in the radiation resistance of HCH and its isomers is detected within the range of the radiation dose (D) from 10 to 200 kGy. The maximum decomposition is observed in the suspension of the preparation. The dependence of a degree of decomposition on the amount of radiation dose is not linear.

1. Introduction

The disposal of acutely toxic pesticides from the list of persistent organic pollutants (POP) is a priority in the field of hazardous waste management both in the Russian Federation and abroad. Full destruction of accumulated stock of organochlorine pesticides is an important task. Tens of thousands tons of pesticides were found following the inventory audit conducted in Russia in accordance with the international treaty on the disposal of POP [1].

Possibilities of using different types of radiation in the destructive processes for bulk hydrocarbon raw stock are discussed in some studies [2–3], that can include natural gas, gas condensates, oil and oil products etc. The examples of successful implementation of such scientific developments in different economy sectors are provided. Thus, the development of radiation technologies (RT) in the field of large-tonnage POP wastes management can be considered as promising technique [2–3].

A possibility of the POP disposal by means of radiation exposure is primarily connected with the selection of a radiation source. As it follows from the literature data [4] as well as from our primary studies [5], the equal absorbed doses of radiation cause an equivalent destruction effect on organochlorine hydrocarbon in case of both gamma and electron beam impact. From practical point of view, a difference in the degree of decomposition of, e.g., organochlorine pesticides (OCP), which belongs to the POP group, is not so important as compared to the disadvantages and advantages of the process technology. It is noted [4] that in case of gamma-radiation, it is possible to use containers for

simultaneous exposure of the substance in large volumes. However, the exposure time for a gamma installation is not less than several hours, while using a high-capacity electron beam accelerator ensures a higher mobility.

In addition to the serch for the best technological solution for the POP radiation disposal, it is important to design the optimal conditions for the radiation exposure of the subject. This study deals with some methodological aspects of electronic impact on the samples including SRS of lindane [6], hexachlorane dust wastes produced in accordance with the technical specifications [7] and the preparation's alkaline water suspension prepared by the method described in [8].

In this work, the dose dependence is studied for the radiation-induced destruction of γ -HCH isomer as a component of an active substance of the pesticide preparation exposed to radiation in different aggregate states.

2. Materials and methods

The 'Hexachloran dust' pesticide formulation, intended for utilization, was substituted for radiation exposure. Electronic irradiation was carried out in doses of 10, 44, 100, 120, 150 and 200 kGy given by the radiation-technological unit (RTU) with an UELR-10-10-40 accelerator. At each radiation dose, several samples (3-7) were irradiated; each sample was arranged as a container (test-tube or weighing bottle) with pesticide formulation in a given form. The test-tubes are made of glass, in the form of a cylinder of 17 cm in length and 1.5 cm in diameter (with a stopper), intended for chemical, biological and microbiological laboratory procedures. Irradiation was delivered via multiple transmission of a transport container (box) with samples, placed on the conveyor belt, through the electron-beam; at this, the necessary dose value was supplemented step-by-step. As soon as the required dose was reached in a certain cycle by a set of samples, this set was removed from the transport container. During irradiation process, dose measurements were carried out.

The initial and irradiated samples of the 'Hexachloran dust' pesticide formulation were analyzed for the content of three stereoisomers of the active substance $-\alpha$ -, β -, γ -HCH. The active substance was extracted with hexane. Methods of sample preparation for irradiation are presented earlier [8, 9]. The experiment on sample irradiation and subsequent sample preparation for chemical analysis was carried out in triplicate. Samples analysis was carried out on GC Crystal-5000.1 chromatographic installation (Russia) in the column temperature programming mode. Statistical processing of data was done with MS Excel 2016 and Origin 2015.

More detailed information on radiation conditions, characteristics of the radiation device, the methods for analysing and processing the experimental data obtained are described earlier [8, 9]. In this publication, some results of the study are discussed that are important for the variation of technical parameters if electronic radiation sources are used in the RT developed for the above purposes.

3. Results and discussion

The radiation destruction of γ -HCH is calculated through the change in the degradation degree of substance (P, %) as part of the preparation's dry powder [9]. The destruction turned out to be instable following the electronic radiation within the range of increasing doses. The substance decomposition efficiency was various for different absorbed doses ranges (0–10, 10–100, 100–200 kGy).

Table 1 presents the results from the degradation degree determination (P, %) for γ -HCH in the samples of lindane and the preparation's suspension analysed, as well as the data for dust radiation degradation [9] in the broad range of doses from electron beam exposure.

According to the results obtained (table 1), common mechanisms can be utilized as a basis for the electronic decomposition of the γ -HCH pure substance and γ -HCH as a component of the pesticide preparation's active substance. However, as it was noted earlier in our studies, an integrated study is necessary to understand the mechanism of radiation-chemical transformation of substances in a multicomponent system, such as the preparation's suspension. This involves, for example, studying the interaction between active particles formed not only with OCP molecules but also with the inorganic component of both water medium and the filler including natural talc and magnesite.

D, kGy	P, %		
	Lindane	Preparation (dust)	Preparation (suspension)
10	24.9	41.9	42.5
44	46.0	-	60.0
100	71.0	66.3	63.5
120	-	-	70.0
150	-	-	85.0
200	71.9	68.5	67.7

Table 1. Degradation degree of γ -HCCH in samples of the drug dust hexachlorane after electron irradiation.

During our previous studies [8], it was found that γ -HCH as a pure substance and a component of the pesticide preparation is very much the same in terms of radiation resistance under the exposure of gamma-radiation. The difference determined is of no practical significance. Similar results were obtained for electronic radiation of the substance. That is why, the design of optimal conditions for the destruction of pesticides by using the RT for different radiation sources is possible by the example of complex heterogenous systems, such as the commodity preparation form, which is accumulated in large amounts and is the subject to disposal.

The determination of a differential dependence between the values of P and D helped discover a trend in the destruction activity of γ -HCH in the electronic radiation dose range studied (figure 1).

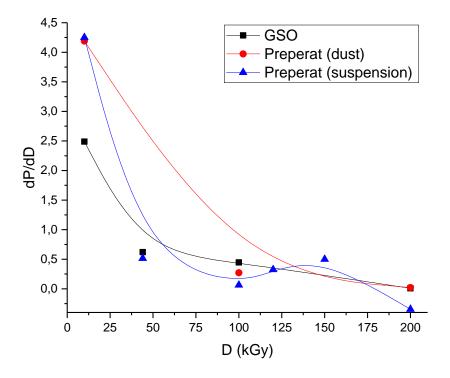


Figure 1. Differential dependence curve $(\Delta P/\Delta D(D))$ for γ -HCH in the composition of the lindane SRS, the hexachlorane dust (dry powder) and the hexachlorane dust (suspension) irradiated with accelerated electrons

According to the data shown in figure 1, at the impact of accelerated electrons on the samples under study with the doses of 10–200 kGy (electron beam energy of 10 MeV), a degree of decomposition of lindane and γ -HCH as part of the preparation's active substance is the highest at the first stage of radiation exposure until the dose of 10 kGy. When the dose increases to 100 kGy, the activity of the destruction process in the substance slows down and remains unchanged in the range from 100 to 200 kGy. The similar form of the dose dependences for the samples obtained (at different aggregate states) confirms the presence of a common mechanism in a consumption and transformation of the electron beam energy that results in the destruction of chemical bonds in the organochlorine hydrocarbon structure.

We also observed a reduction in the destruction activity of ionising radiation with an increase in the radiation dose when studying the radiation resistance of OCP under the impact of gamma-rays. However, due to the variation of dose levels in the experiments conducted, we did not manage to determine reliably a change in the P values to plot the dose dependence curve.

4. Conclusions

It is known that the radical mechanism of radiation-induced destruction of substance is determined by the ratio of rates between two contrary processes: forming active particles under the impact of ionising radiation and developing reactions of their recombination. It can be assumed that in our experiment, we observe a reduction in the rate of the first process. In this connection, the following problems should be solved to intensify the process (for practical purposes):

- the possibility of a rapid increase in the electronic radiation dose for a full mineralization of organochlorine pesticide;
- an application of additional factors enhancing the availability of ionising radiation energy for a decomposed substance on the molecular level, even at small doses of radiation exposure. Such factors can include, for example, reducing the duration of the preparation presence in the active zone, maintaining the medium's turbulence and increasing its temperature and degree of dispersion;
- an expansion of the preparation's aggregate states in the radiation medium; e.g., transferring the preparation's active substance to the vapor phase.

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