The object of this study is the process of impregnation of liquid into the bulk material, in particular, into the soil. Determining the impregnation parameters is a relevant task when assessing the consequences of an emergency spill of a hazardous liquid. Infiltration of liquid into the soil leads to pollution of water resources. However, the greatest danger is the ignition of the spill of a combustible liquid.

Based on the Green-Ampt model, a mathematical description of the impregnation of liquid into bulk material was built. It is a system of two ordinary differential equations of the first order, one of which describes the reduction of the thickness of the liquid layer on the surface, and the other describes the dynamics of the impregnation of liquid into depth. The solution to the system was derived in the form of time dependence on the depth of impregnation.

An experimental study was conducted on the example of impregnation of crude oil in the sand. To this end, sand was poured into a vertical measuring glass cylinder. After that, the liquid was poured and a video recording of the impregnation process was carried out. By processing the video recording, the depth of impregnation and the corresponding time were determined. The results of the study show that the relationship between the thickness of the liquid layer on the surface of the sand and the depth of impregnation is linear in nature: the relative deviation of linear approximation from experimental data does not exceed 3.5.

By expanding the logarithmic function contained in the solution to the system of differential equations into the Taylor series, a polynomial dependence of time on the depth of impregnation was established. To determine the coefficients of the polynomial based on the experimental data, the least squares method was used. In this case, the approximation error after the first minute after spilling does not exceed 10 %.

The proposed method could be used to account for seepage in the model of liquid spreading on the ground and the burning model of a flammable liquid spill

Keywords: liquid spill, impregnation parameters, Green-Ampt model, porosity coefficient, bulk material

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# JUSTIFYING THE EXPERIMENTAL METHOD FOR DETERMINING THE PARAMETERS OF LIQUID INFILTRATION IN BULK MATERIAL

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#### 1. Introduction

A significant number of emergencies arising in the chemical, processing industry, and transportation begin with an emergency spill of combustible or other dangerous liquids [1]. Infiltration of fluid into the soil leads to pollution of water resources: both groundwater [2] and river [3]. However, the greatest danger is the ignition of the spill of a combustible liquid. This threatens not only the spread of fire to neighboring technological facilities and natural landscapes but also leads to the release of pollutants into the atmosphere [4]. Spreading over long distances, they significantly affect the state of the air and create risks for the population [5].

Despite acting regulatory documents governing the rules of fire safety during the transportation of dangerous goods, accidents with their participation still happen. This is confirmed by emergencies associated with the spill or combustion of combustible liquids that occurred on railroad transport:

- 2021 (USA, Texas) - a train with petroleum products went off the rails and collided with a truck. 3 tanks caught

fire, and the height of the flame from the fire was several tens of meters. Residents of nearby houses were evacuated;

- -2021- on the overpass during cargo work there was a fire of petroleum products in the tank;
- 2020 (USA, Arizona) tanks with flammable liquids went off the rails and ignited;
- -2020 (Republic of Kazakhstan, Zhambyl region) a tank with gasoline went off the rails, resulting in a spill and fire. The fire area was about  $600 \text{ m}^2$ ;
- $-\,2019$  (Canada, Manitoba)  $-\,a$  train with 37 oil tanks went off the rails, which led to its partial spill. As a result of the impregnation of oil into groundwater, one of the surrounding sources of drinking water turned out to be contaminated;
- $-\,2015$  (Canada, Ontario) several tanks with petroleum products went off the rails, which led to the spillage of part of their contents. Despite the cleaning of the soil, traces of petroleum products were found at the mouth of the local river system.

The development of plans for the localization of emergencies related to the spill of combustible liquids requires

determining the geometric parameters of the spill and the dynamics of their change, depending on the properties of the liquid and soil. Impregnation of liquid into the soil reduces the thickness of the layer on its surface, and hence the area of spreading. On the other hand, it leads to soil pollution and the ingress of pollutants into groundwater. Thus, a relevant issue in the spill of liquid on the surface of the soil is its impregnation in depth.

#### 2. Literature review and problem statement

Study [6] analyzes the risks arising from the transportation of dangerous goods by rail but the consequences of accidents are left out. In [7], an analysis of emergencies related to the spill of combustible liquids on railroad transport was carried out. It is proposed to use statistical data to calculate the probabilities of accidents and the volume of spilled combustible fluid. This approach makes it possible to summarize the consequences of accidents but does not make it possible to analyze a specific situation. In [8], a fire spill on a large area in a railroad tunnel is investigated. A feature of the approach is the division of the entire space into separate zones and the calculation of the temperature distribution in them. In this case, the area of the fire itself is considered a priori assigned. Spilling and burning liquid can lead to a cascading spread of fire to natural landscapes. Paper [9] considers measures to limit the spread of landscape fires. In [10], the thermal effect of fire on steel structures is considered but the dynamics of changes in the parameters of the combustion cell were left out. In [11], the environmental characteristics of fire extinguishing agents used for extinguishing oil product fires are considered. At the same time, the formation of a spill and the effect of liquid seepage into the soil on fire parameters are not considered.

In experimental work [12], the dynamics of n-butanol spreading with simultaneous flame propagation were investigated. The disadvantage of that approach is the dependence of the results obtained on the conditions of the experiment and the impossibility of their generalization. In [13], the spreading and combustion of combustible liquids on the surface of the refractory glass were investigated. In [14], an empirical model of the spreading of gasoline, isooctane, and ethanol on an aluminum surface was constructed. The use of this class of models in practice is difficult due to the fact that under real conditions the surface is not perfectly smooth, it has irregularities and inclinations.

One of the common methods for modeling the spread of liquid over a horizontal surface is the use of the principle of gravitational spreading of a cylindrical layer of liquid [15]. The analysis of models of liquid spreading on a solid surface is reported in [16]. In it, based on a comparison of calculations according to a model from [17] and experimental data, a modification of the model is proposed. The disadvantage of that approach is that the proposed correction depends on the conditions under which the experimental studies were carried out.

Our review of models of spreading of combustible liquids showed that they do not take into consideration the impregnation of liquid into the underlying surface. This, in turn, leads to errors in assessing the size of the spill, and the dynamics of its formation. That necessitates research into determining the parameters of impregnation of liquid into the soil.

#### 3. The aim and objectives of the study

The aim of this work is to devise a method for the experimental assessment of the parameters of impregnation of liquid into the bulk material. This will make it possible to take into consideration the dynamics of impregnation during the spread of hazardous liquid on the soil.

To accomplish the aim, the following tasks have been set:

- to derive a mathematical description of the process of impregnation of liquid into bulk material;
- to build a linearized model of the dynamics of impregnation of liquid into bulk material;
- to find estimates of model parameters on the example of seepage of crude oil in the sand.

#### 4. The study materials and methods

The object of this study is the process of impregnation of liquid into the bulk material. The main hypothesis assumes the presence of a clear boundary between dry and already moistened material. It is assumed that the impregnation of liquid into bulk material is described by the Green-Ampt model [18]. In order to simplify the construction of a system of equations for determining the impregnation parameters, an exact solution to the system of equations is expanded into the Taylor series. We have experimentally determined the parameters on the example of impregnation of crude oil in the sand.

# 5. Results of research on the justification of the method for determining the parameters of impregnation of liquid in bulk material

## 5. 1. Mathematical description of the impregnation of liquid into the bulk material

The impregnation of liquid into the bulk material, in particular soil, is described by the Green-Ampt model. Impregnation of the liquid deep into the soil leads to a movement down the boundary between the already moistened and still dry soil. The impregnation rate is the speed of the limit. We direct the vertical axis Z so that its direction coincides with the direction of impregnation of the liquid (Fig. 1).

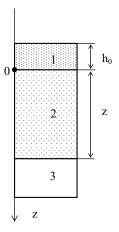


Fig. 1. Impregnation of liquid deep into the soil: 1 — liquid on the surface; 2 — moistened soil; 3 — dry soil

The impregnation speed then takes the following form

$$q = \frac{\partial z}{\partial t}. (1)$$

The rate of impregnation is described by Darcy's law

$$q = K \frac{\partial H}{\partial z},\tag{2}$$

where *K* is the hydraulic conductivity of moistened soil;  $\frac{\partial H}{\partial z}$  is the hydraulic gradient:

$$\frac{\partial H}{\partial z} = \frac{h_0 + z + h_f}{z};\tag{3}$$

 $h_0$  is the thickness of the liquid layer on the soil surface; z is the thickness of the wetted soil layer;  $h_f$  is the suction head. By combining expressions (1) to (3), we obtain the following equation

$$\frac{\partial z}{\partial t} = K \frac{h_0 + z + h_f}{z}.$$
 (4)

Impregnation of liquid deep into the soil leads to a decrease in the thickness of the layer on its surface:

$$\frac{\partial h_0}{\partial t} = -\varphi \frac{\partial z}{\partial t},\tag{5}$$

where  $\phi$  is the coefficient of soil porosity, which can be calculated from the following expression

$$\varphi = \frac{\rho - \rho_b}{\rho},\tag{6}$$

where  $\rho_b$  is the bulk density of the soil;  $\rho$  is the density of soil particles.

Thus, the dynamics of impregnation of liquid deep into the soil are described by a system of equations (4), (5) under the following initial conditions

$$h_0(0) = c_0, \tag{7}$$

$$z(0) = 0, (8)$$

where  $c_0$  is the initial thickness of the liquid layer on the soil. Solving the system (4), (5) under initial conditions (7), (8), we obtain the dependence of time on the depth of impregnation z

$$t(z) = \frac{z}{K(1-\varphi)} - \frac{c_0 + h_f}{K(1-\varphi)^2} \ln\left(1 + \frac{1-\varphi}{c_0 + h_f}z\right),$$

$$c_0 - \varphi z > 0. \tag{9}$$

The condition  $c_0$ – $\phi z>0$  means that dependence (9) is true if there is a layer of liquid on the soil surface. Complete impregnation of the liquid into the soil will occur when

$$c_0 - \varphi z = 0. \tag{10}$$

The practical use of dependence (9) requires knowledge of the hydraulic conductivity coefficient K, the coefficient of soil porosity  $\phi$ , and the suction head  $h_f$ . In general, they must be determined experimentally.

### 5. 2. Construction of a linearized model of the dynamics of impregnation of liquid into the bulk material

Note that for small values of impregnation depth  $(z < c_0 + h_f)$ , the expansion of the  $\ln(1+x)$  function into the Taylor series converts (9) to

$$t(z) = \frac{1}{2K(c_0 + h_f)} z^2 - \frac{1 - \varphi}{3K(c_0 + h_f)^2} z^3 + \frac{(1 - \varphi)^2}{4K(c_0 + h_f)^3} z^4 - \dots$$
 (11)

Limited to the first two terms of the series, we obtain the dependence of the impregnation time on the depth in the following form

$$t(z) \cong az^2 + bz^3. \tag{12}$$

The unknown coefficients a, b are to be found as values that provide a minimum of the sum of the squares of deviations calculated from the formula of time values  $t(z_n)$  depending on the experimental values of  $t_n$ :

$$L = \sum_{i=1}^{n} (t(z_i) - t_i)^2 \underset{a,b}{\longrightarrow} \min.$$
 (13)

Substitution (12) in (13) produces the minimization problem:

$$L = \sum_{i=1}^{n} \left( az_i^2 + bz_i^3 - t_i \right)^2 \underset{a,b}{\to} \min.$$
 (14)

Problem (14) has a single solution, which is determined by the necessary conditions of the extremum:

$$\frac{\partial L}{\partial a} = 2\sum_{i=1}^{n} \left( az_i^2 + bz_i^3 - t_i \right) z_i^2 = 0; \tag{15}$$

$$\frac{\partial L}{\partial b} = 2\sum_{i=1}^{n} \left( az_i^2 + bz_i^3 - t_i \right) z_i^3 = 0.$$
 (16)

Solving a system of linear equations (15), (16) relative to a,b, we obtain

$$a = \frac{c_{13}c_{22} - c_{23}c_{12}}{c_{11}c_{22} - c_{21}c_{12}}; \ b = \frac{c_{11}c_{23} - c_{13}c_{21}}{c_{12}c_{21} - c_{11}c_{22}}, \tag{17}$$

where

$$c_{11} = \sum_{i=1}^{n} z_i^4; \ c_{12} = \sum_{i=1}^{n} z_i^5; \ c_{13} = \sum_{i=1}^{n} t_i z_i^2;$$
 (18)

$$c_{21} = \sum_{i=1}^{n} z_i^5; \quad c_{22} = \sum_{i=1}^{n} z_i^6; \quad c_{23} = \sum_{i=1}^{n} t_i z_i^3.$$
 (19)

Thus, formulas (17) to (19) determine the coefficients of the approximating polynomial (12).

## 5. 3. Results of determining the parameters of impregnation of oil in the sand experimentally

For our experimental research, sand was used as a bulk material, which was poured into a cylinder with a diameter of 60 mm. Crude oil was chosen as a liquid.

The experiment is schematically shown in Fig. 2.

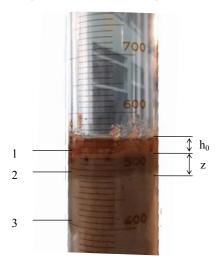


Fig. 2. Scheme of the experiment: 1 — a layer of liquid on the surface of the sand; 2 — a moistened layer of sand; 3 — a dry layer of sand

The results of measuring the depth of impregnation z, and the thickness of the liquid layer on the surface  $h_0$  at different points in time are given in Table 1.

Table 1
The dependence of the depth of impregnation on time

	·	. •
Time, t, s	Depth of impreg- nation, z, cm	The thickness of the layer on the surface, $h_0$ , cm
0	0	1.66
8	0.18	1.56
20	0.35	1.48
44	0.53	1.42
72	0.70	1.37
133	0.88	1.32
232	1.05	1.28
328	1.23	1.23
487	1.40	1.18
750	1.58	1.14
1,030	1.75	1.09
1,296	1.93	1.04
1.598	2.10	1.00

The relationship between the thickness of the oil layer on the surface of the sand and the depth of impregnation is almost linear (Fig. 3).

Equation (5) and the results given in Table 1 produce the assessment of the porosity coefficient:

$$\varphi = -\frac{\Delta h_0}{\Delta z} \approx 0.314. \tag{20}$$

Calculating the coefficients according to formulas (17) to (19) produces

$$a = 5.6 \cdot 10^5 \text{ s/m}^2$$
;  $b = 1.5 \cdot 10^8 \text{ s/m}^3$ . (21)

Fig. 4 shows the experimental dependence of time on the depth of impregnation and its approximation in the form of (11). The relative approximation error (11) is shown in Fig. 5. During the first minute, the relative error remains significant, and then it does not exceed 10 %.

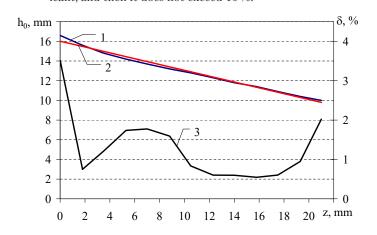


Fig. 3. The relationship between the thickness of the oil layer on the surface of the sand and the depth of impregnation:
1 - experimental data (Table 1); 2 - linear approximation;
3 - relative approximation error (right axis)

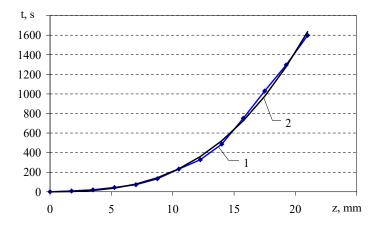


Fig. 4. The dependence of time on the depth of impregnation: 1 - experiment; 2 - approximation in the form of (11)

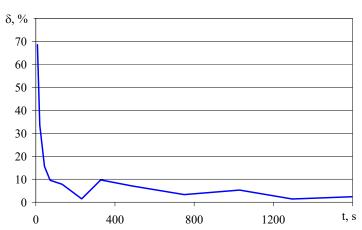


Fig. 5. Dependence of the relative approximation error (11) on the time of impregnation

## 6. Discussion of research results to substantiate the method for determining the parameters of impregnation of liquid

The mathematical description of the process of impregnation of a liquid into a dry bulk material is based on the Green-Amp model. A feature of the model is the consideration of the seepage process as a movement down the boundary between already moistened and still dry soil. Using Darcy's law, a system of ordinary first-order differential equations (4), (5) with initial conditions (7), (8) was constructed. In this case, equation (4) describes the dynamics of impregnation in depth, and equation (5) describes the decrease in the thickness of the liquid layer on the surface. The solution to the system of differential equations is derived in the form of time dependence on the depth of impregnation (9).

Dependence (9) contains parameters such as hydraulic conductivity coefficient, soil porosity coefficient, and suction head. These parameters depend on the seepage liquid, as well as on the type of bulk material and its condition (humidity, compressibility). If all these parameters are known, then their substitution in (9) makes it possible to determine the impregnation time to a given depth, as is reported in [16]. However, from the practical point of view, these parameters are not known a priori.

Direct evaluation of the parameters included in ratio (9), for example, by the least-square method, is complicated due to the nonlinear nature of the dependence on the specified parameters. The expansion of the logarithmic function into the Taylor series allows us to obtain for the impregnation time the expression (11), linear relative to the powers of the impregnation depth z. Limited to the first two terms of the series (second and third powers relative to z), a polynomial dependence of time on the depth of impregnation was established. This makes it possible to apply the least squares method to determine the coefficients of a polynomial from experimental data regarding the dependence of time on the depth of impregnation.

Our experimental study was conducted on the example of impregnation of crude oil in the sand. To this end, sand was poured into a vertical measuring glass cylinder. After that, the liquid was poured and a video recording of the impregnation process was carried out. By processing the video recording, the depth of impregnation and the corresponding time were determined (Table 1). The results of the study show that the relationship between the thickness of the liquid layer on the surface of the sand and the depth of impregnation is linear (Fig. 3): the relative deviation of linear approximation from experimental data does not exceed 3.5 %. This makes it possible to determine the porosity coefficient from equation (5):  $\phi$ =0.314.

Our analysis of dependences in Figs. 4, 5 reveals that after the first minute after the liquid spill, the time dependence on the depth of impregnation is satisfactorily approximated by polynomial (12). The error of such fitting does not exceed 10 % and tends to decrease over time.

Thus, the proposed method for determining the parameters of impregnation of liquid into bulk material experimentally involves the following:

- replacement of the exact solution (11) to the problem of impregnation of a liquid with an approximate solution in the form of polynomial (12);
- calculation of the coefficients of the approximating polynomial for (18), (19), obtained by using the least squares method;

- calculation of the porosity coefficient according to formula (20).

Owing to the proposed approach, it is possible to solve the task of determining the parameters of impregnation of liquid into the soil.

From a practical point of view, polynomial (12) with coefficients determined from formulas (17) to (19) makes it possible to determine the volume of liquid that will have time to seep deep into the underlying surface before the spill is eliminated. This, in turn, makes it possible to assess the thickness of the contaminated soil layer and the volume of liquid that could get into the groundwater.

The limitations of the proposed method are that the results obtained are fair for a given state of the soil (moisture and compressibility) and cannot be transferred to other states.

The disadvantages of the proposed method include the impossibility of determining such parameters as the coefficient of hydraulic conductivity and suction head. Therefore, the prospects for further research are related to determining them by applying the least squares method directly to dependence (11). It should also be noted that for certain bulk material and a certain liquid, the coefficient of hydraulic conductivity and the suction head are associated with the porosity coefficient.

The proposed method could be used to take into consideration the impregnation in the model of liquid spreading on the ground [15] and the combustion model of the spill of combustible liquid [19]. Taking into consideration the impregnation of liquid into the soil during its spreading and combustion makes it possible to refine the thermal effect of fire on steel and concrete structures [20].

#### 7. Conclusions

- 1. Based on the Green-Ampt model, a mathematical description of the impregnation of liquid into bulk material has been built. The impregnation process is considered as the movement of the boundary between the already moistened and still dry material. Using Darcy's law, a system of ordinary first-order differential equations was constructed. One of the equations describes a decrease in the thickness of a layer of liquid on the surface, and the second describes the dynamics of impregnation in depth. The solution to the system was derived in the form of time dependence on the depth of impregnation.
- 2. A method for determining the parameters is proposed, which involves replacing the resulting irrational time dependence on the depth of impregnation with an approximating polynomial. To estimate the coefficients of a polynomial, the least squares method is used, which, in this case, leads to a minimization problem that has a single solution. The approximating polynomial contains terms of the second and third powers relative to the depth of impregnation. The relative error of such approximation after the first minute of spill does not exceed 10 %. The values of the coefficients at the second and third powers of the impregnation depth were  $5.6 \cdot 10^5$  s/m<sup>2</sup>; 1.5·10<sup>8</sup> s/m<sup>3</sup>, respectively. The use of an approximating polynomial makes it possible to determine the volume of liquid that seeps deep into the underlying surface before the spill is eliminated. This, in turn, makes it possible to assess the thickness of the contaminated soil layer and the volume of liquid that could get into the groundwater.
- 3. Our analysis of the impregnation of crude oil in the sand reveals that the depth of impregnation and the thick-

ness of the liquid layer on the surface of the sand are linearly related. The relative error in the linear approximation of experimental data does not exceed  $3.5\,\%$ . This makes it possible to estimate the value of the static impregnation parameter – the porosity coefficient. Its value in the experiment was 0.314.

#### **Conflict of interest**

The authors declare that they have no conflict of interest in relation to this research, whether financial, personal, authorship or otherwise, that could affect the research and its results presented in this paper.

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