

## **PERSPECTIVES OF APPLYING OF NEW BIOSORBENTS AT LIQUIDATION OF OIL POLLUTION OF THE SEA.**

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### **ABSTRACT**

The increase activities to exploit the hydrocarbon resources in the sea areas around the world, including the Arctic, have posed an increased threat of pollution to the sea environment. In the Arctic and some remote areas, present knowledge of pollution protection and oil spill combating has turned out to be inadequate. The applying of conventional methods of liquidation of oil pollution frequently does not prevent ecological disasters. Therefore, the Institute of Limnology of Russia (ILR) has established a research project on the use of biosorbents for oil combating purpose. The biosorbents are yields received by a biotechnological way, and capable to activate processes of a self-cleaning from oil of seawater, shore-line and sediments too. This paper provides some results of biosorbents designed specifically for marine oil spill response and some other water objects.

## **INTRODUCTION**

The cleanup of marine oil spills by bioremediation has been mostly limited to the cleaning of polluted beaches, as was done to some of the beaches affected by the Exxon Valdez spill (Pritchard P.H. 1999) and in some other situations (Swanell R.P.G., et al 1999). But given the natural ability of many microorganisms to transform petroleum hydrocarbons to innocuous products, the full potential of bioremediation is not being used. Bioremediation, in conjunction with physical and chemical technologies, has a major role in eliminating or reducing environmental damage before the spill reaches sensitivity areas (Worthington, T. F, 1993). We have taken for a basis of development methods (biosorbents) intensifying natural process of a self-cleaning of water from oil, i.e. capacity of a natural ecosystem to manage impurity. In our work we have aggregated also some of stages of purification of natural ecosystems from petroleum (Levtchenko A., V. Roumiantcev, 1995). Besides this we have constructed new technology on usage, in basic, natural components, that after applying a methodology in natural system did not remain of stranger ingredients of an anthropogeneous genesis. In studies the special attention on both forms of usage of physical-geographic and ecological differences of different aqueous ecosystems in the practical purposes - oil combating in waters in northern latitudes is reversed (V. Roumiantcev, A. Levtchenko, 1997). The preparations are made only on the basis only of the natural components, including specially wrought aluminum silicate (Vermiculite, Perlite) and natural microorganisms enable to destroy petroleum and to absorb it. The operating of biosorbent on crude oil has two main directions: fragmentation of an oil slick on the water, and biodestruction of oil.

For the first time basic stands on a case in point were by us formulated in middle of 80th (Levtchenko A. 1984). The preparation created on the base of specified principles we have called as biosorbents. Now the term "biosorbent" is successfully applied in the domestic

and foreign literature. The performance of experimental types of biosorbents for cleaning of sea water and shore line area was demonstrated during joint workings of Murmansk Marine Biological Institute of Russia (MMBI RAS) and the Technical Research Center of Finland (VTT) (J. Rytönen et al 1997; Matishov G., et al, 2001). Now we elaborated some new type of the preparations for oil combating purpose:

1. Purification from the crude oil and petroleum products in the surface, in water column and shoreline – " Biosorb- A";
2. Purification from the crude oil and petroleum products in the sediments and shore-line – «Biosorb-D». In this paper we review some results of practical and experimental use of biosorbents on miscellaneous objects.

## **BIOSORBENTS FOR PURIFICATION OF WATER**

Biosorb-A is a combination of aluminosilicates, natural microorganisms, and nutrients (see table 1). The aluminosilicates substrate is heat treated to heighten the flotation and oil absorbent properties (see table 2). Some natural surfactant of bacterial genesis is used as a header to control the spreading ability of the oil to maintain a 'lens' of on the surface. A second effect is the generation of interfacial area in the nature of finely dispersed oil droplets. Aerobic and anaerobic microorganisms are selected to degrade oil in various temperatures and salinity conditions. The aerobic organisms remediate the surface oil and the anaerobic degrade the oil that is dispersed in the water column and sediments. In composition of biosorbents the oxidizing oil hydrocarbons of a bacteria of groups *Bacillus*, *Micrococcus*, *Rhodococcus*, *Pseudomonas* and some other are customarily used. The bacteria will be used in various combinations that depend on tasks of cleaning (type of petroleum, type of water, temperature etc.). The relevant condition of ecological safety and

performance of technology is usage of bacteria from region of potential applying of biosorbent. We fulfil the express methods of insulation of indispensable bacteria from water and bottom sediments (Levtchenko et al 1986). In biosorbent bacteria are not simply mixed with fragments of absorbent, but immobilized with these particles. From the industrial immobilization of microorganisms (Webb K. 1990) it is well known, that in this case the biodestructive activity of oil oxidizing bacteria essentially grows. The organisms are dried on the absorbent for ease of storage and distribution. The rate-limiting nutrients of nitrogen, phosphorus, and carbon are included in the product. As it was mentioned above, the operating of biosorbent in crude oil on the surface of water has two main directions (table 3):

a) Fragmentation of an oil slick and preventing of it' flowing on the surface of water. This effect is created optimal conditions for mechanical deleting of oil from water;

b) By integral activity in destruction of oil. The activity, in destruction of oil, biosorbent is piled from three main components:

1. Biological destruction under operating of bacteria's inclusive in the particles of biosorbent (40-60 % of activity, depending on conditions: temperatures, aeration, such as oil and others);
2. Destruction of oil at the expense of the physicochemical factors (10-18 %). This effect stimulated by biosorbent too;
3. The destruction of oil by means of a natural self-cleaning of natural pools, which one is stimulated by biosorbent (20-30 % of activity);

As it was shown in more than 20 accidents of oil pollution, biosorbents "Biosorb-A" have a cascade-like effect in the oil layer on the water (see Table 3). It can be spread by helicopter (quickest way) and from the vessels (Fig. 1). In conditions of strong gale, at a wind up to 25 m/s, the helicopter is the alone way of evaporation of biosorbent on a spot of

oil (Fig. 1). In this case, the biosorbent works in an autonomous mode. So in storm conditions (when special vessels with skimmers can not be used) practically there is no alternative of usage of biosorbent (or so types of preparations) with help of aircraft for oil combating purposes in the sea. The self-sufficiency of usage of biosorbents does not require of their gathering from water. It is linked to a natural genesis of the majority of components of a biosorbent. After destruction of 75-85 % of oils in water remain predominantly ecologically inert components. It was shown even in early studies of experimental biosorbents in Institute of Marine Biology (Murmansk) (Matishov G., et al, 1994). If the gathering of biosorbent with oil from a surface of water is possible, there are simple methods of salvaging of the collected material (Fig. 1). The biosorbent with oil can be housed on special site. There the process of degradation of oil can be lead up to the end. Probably and incineration of the collected material (biosorbent with oil). In this case aluminosilicates fragments of biosorbent can be repeatedly used for producing of the new biosorbent.

During liquidation more 20 of oil pollution's of other water objects with help of biosorbents it was shown that, for destruction of 1 ton of crude oil (in average on 70-80%) is spent from 150 up to 180 kg of a biosorbent at temperatures above + 7 + 8<sup>0</sup> C. Proportion biosorbent : the oil is equal approximately 1: 5-7). In can be used from 200 up to 300 kg at more low temperatures (proportion biosorbent: the oil is equal approximately 1: 3-4). At higher temperatures (>+ 10 + 15<sup>0</sup> C) this proportion is a compound 1: 7-10. At applying biosorbents on boiler oils, their quantity is augmented on 20-30% (V. Roumiantcev, A. Levchenko, 2001).

In practice it is customary between emergency and acceptance of measures of cleaning of its consequences transits considerable time. In conditions of storm for this period there will be an emulsification of oil, and penetrated this emulsion in strata of water and in

bottom's sediments. These processes essentially complicate tasks of liquidation of emergency spill petroleum, as require cleaning of impurity not only surface of water, but aqueous strata and bottom. For cleaning of strata of water the shallow sedimentation particles of aluminosilicates (15-20 %) are introduced into composition of biosorbent. They interface with drops of oil in water (or water in oil). It allows to activate the process of disintegrating of oil in thickness of water. In case of pollution of bottom, especially in a coast band, in composition of biosorbent the high-gravity fragments are added. The presence in composition of a yield of the anaerobic strains of bacteria promotes process of refining of bottom sediments. Thus, depending on entity of cleaning, the various modifications of biosorbent (table 4) are applied. In composition of biosorbents, (table 4), the strains of bacteria of a natural genesis are used. The bacteria were taken from regions of potential applying of product. The modification of biosorbents can differ by composition and also by floatation characteristic of mineral absorbing fragments. For example, some forms of zeolites are introduced into composition of biosorbents for oil combating in bottom area. There are differences in composition of the nutritious additives in biosorbents. The detailed analysis of properties of miscellaneous types of biosorbents and ways of their operational use leaves for frameworks of a subject of sectional paper. For us it is important to stress attention to broad possibilities of such technological approach to cleaning (or addition purification) different entities from oil. It is especially important, because sometimes used methods of oil combating enter inconsistency with the international legislation which must regulate the methodology of oil combating. For example, the rules of HELCOM do not recommend the usage of absorption materials, which are able to result in to encroaching of a part of oil. It seems that such rules were preparing before wide usage of bacterial components in composition of absorbents and today have become outdated. The biosorbents created on principles, depicted above, are

capable to be used not only for cleaning of a surface of water from oil, but also bottom sediments. Therefore we will discuss more detail about last types of biosorbents.

## **BIOSORBENTS FOR PURIFICATION OF BOTTOM SEDIMENTS AND SHORE-LINE.**

The impurity of near-shore area and bottom sediments after oil spill is a significant problem of the wreckers of all countries. The special place in these problems is taken drowned, or - with sunken ships - constant sources of oil impurities. Usage of means of clearing is linked as a matter of fact to retraction of the contaminated earth. The dredgers of miscellaneous constructions are applied to cleaning of bottom, if hydrological and the meteorological conditions it allows. As have shown recent incidents near to beaches of Denmark in 2001 and Spain in 2002-2003 years in cleaning of a seacoast the playing key role belongs to hand-operated retraction (Fig. 2) (Pesceli Maria, et al 2002). If the depth of water in a crash scene is great, laden down on bottom the petroleum are not put away at all. It results in non-reversible breaking downs of base ecosystems. Last it is apart dangerously in regions of a spawning of fishes, places of net making of birds and in other similar cases. As usage of biosorbents, or similar method of applications of cleaning, substantial path of activating and keeping up of a self-cleaning of bottom sediments in region of oil spill seems, to us. Already in the mentioned before Russian-Finnish studies in the Kola bay the performance of biosorbents in cleaning of the soil, contaminated with oil, in a band of a marine afflux was rotined. Now we design the new experimental type of biosorbent for cleaning of naturally bottom sediments. Its performances are given in table 6 The recent experiment on testing of biosorbent was conducted on the sandy bottom taken with

contaminated boiler oil of the small river, contaminated with boiler oil, from depth about 1,5 meters. Testing conducted in 1-m bulbs close to applied earlier (Ryttonen J. et al. 1994-b).

The methods of analysis of oil are similar to that were applied in experiments on investigation of biodegradation of oil in an intertidal zone of the Kola bay, including amount of biosorbent (Liukkonen, S., Ryttonen, J., 1994-a; Matishov G., et al 1999), in view of the recent advisories (Wang Z., et al 1999). For aeration of water and precipitation have used valve drain of air through customary aquarium's pump. Some received data are given in table 6.

As follows from the reduced data, the rather effective biological breaking down of oil product, including is watched at low temperatures. Thus the basic process went per the maiden 3-4 weeks. Probably, the complete cycle of cleaning of bottom requires 2-3 series treating of the contaminated object. It is possible to guess, that in conditions of fissile hydrodynamic processes in natural basins the breaking down of oil products will be even more effective. Thus, as we marked above, the performance of biosorbent in full-scale conditions will be on 20-30 % more effective, than during the laboratory tests. It is linked to activating of processes of a self-cleaning, as it had place in case of usage of biosorbents for cleaning of water.

Some preliminary results let us to suppose, that the biosorbent is suitable for cleaning of bottom and a near-shore area too. In table 7 the results of testing several are introduced for model of soil contaminated with different types of petroleum. The method of application of depositing of biosorbents was similar to experiments on a coast of the Kola Bay (Matishov G., Petrov V. et al 1999). At depositing biosorbent the proportions of 1 kg of a product on 1 m<sup>2</sup> purged soil started with. The particles of biosorbent were uniformly were distributed to depth 5-7 cm. During this experiment we also select technological conditions of using

of biosorbents. It was shown that the soil, treated by biosorbent, was not rich by a nutrient's material, which follows from the data of table 7. It is characteristic for mineral soils (sand, alum earth and others). Though such soils have some spares of natural nitrogen (see, table 7). There is a major nutrient material promoting effective biological destruction of petroleum, - includes indispensable - ammonium nitrogen. The level of an admixture of nitrogen compound matters needs for high activity of process. Basically, there are some way's to make adequate the tasks of biodegradation of nutrient materials in purged soil of a coast band. Instead of synthetic fertilizers together with biosorbent it is possible also to use for example slurry of sewage. However, as follows from the literary data, mixed fertilizers and some of peat (Puustinen J., et al 1995) reach the best effect. The discussion of details for practical using of such additives is out of the scope of this paper. For us it is important to state that the optimal result of cleaning of beach will be reached in case of making in soil of an indispensable balance of mineral and biogenic reductants.

## **CONCLUSION REMARKS.**

In geologic retrospective the nature was interfere witched by scale oil impurities. A natural self-cleaning process reached the recovery of an environment. This process includes a broad complex of biological and physicochemical affecting on oil hydrocarbons. Biosorbents it as a matter of fact attempts for modeling of such process. However, modeling is adequate to quantitative and quality composition of contaminating ingredients. Many aspects of operating of biosorbents on oil, reviewed above, were learnt not only in laboratory, but also at liquidation of a series of oil spill. We review some data in sectional paper. First of all it concerned biosorbents for cleaning of water. However, in several cases the high-gravity biosorbents suitable for clearing of bottom and a near-shore area were

applied also. Some examples of successful applying of biosorbents are naturally depicted below. In most cases of oil combating process's, the refining measures conducted specialized rescue services. Some results of practice application of biosorbents with technical equipment and in autonomy regime in 1993 – 2003 are introduced below:

- Liquidation of flood (0,7 t.) of black oil, Bourgas harbor, Bulgaria - July 1993.
- Liquidation of flood (0,4 t.) of crude oil, Kola bay, Murmansk (Barents Sea) - May 1994
- Liquidation of flood of 1,5 t. Black oil, Gulf of Finland, Primorsk (w. - ice) - March 1996
- Clearing of water near lakes, Luga- city (black oil, 1,2 t, water - ground) - April 1996.
- Liquidation of flood 10 tons of diesel fuel in Gulf of Finland, Vyborg, - July 1996.
- Clearing of water from petroleum (up to 6 ton) harbor St. - Petersburg - November 1996.
- Liquidation of oil spill of 21 tons of fuel on ground, Tichvin - April 1997.
- Using together with skimmers in liquidation on 75 % of pollution of Ladoga Lake by petroleum (35 - 40 tons), June 1997.
- Liquidation on 85 % of pollution, Valdai lake by oil (18 tons) - August 1997
- Liquidation of flood at rise of a vessel (3-4 t of black oil) Kamchatka - December 1997
- Liquidation of spill of fuel (18-20 tons; water) Kronshtadt - January 1998
- Liquidation of flood of black oil 14- 16 t, (soil-snow), near S. -Petersburg - March 1998
- Liquidation of spill of fuel (1,5 t), S. -Petersburg, Harbor - August 1998
- Liquidation of spill (black oil, 0,8 t) ports Novorossiysk, Black Sea - December 1998
- Liquidation of spill of fuel at rise of a vessel (1,2 t.) Novorossiysk, Black Sea - March 1999
- Liquidation of spill of fuel (1,5 t), Harbor, Eisk, Asov Sea - July 1999
- Using in all - up liquidation of flood (black oil 60 - 70 t.), r. Neva, S – Pb. - October 1999
- Liquidation of emergency flood of fuel (1,5 t), St. Petersburg, harbor - April 2000
- Using in liquidation of flood (fuel, 8-12 t), Kronshtadt, Nevsky inlet - November 2000
- Liquidation of overflow of 12 tons of oil on the river (republic of Komi, city Uchta) - July 2001
- Using in liquidation of flood ( fuel, 12 t), Kronshtadt , Nevsky inlet - November 2001

- Applying at a loading of oil ships by petroleum; Harbor of Primorsk City - February 2002
- Liquidation of oil film (up to 2000 m<sup>2</sup>, 1-3 mm) harbor, Eisk, Asov Sea - March. 2002
- Using in liquidation of flood (d. fuel, 2 t) near river, Uchta, Komi - July 2003
- Using in liquidation of oil on water (3-4 t), Neva river - August 2003
- Liquidation of flood of fuel at rise of a vessel (7-8 t.) S.-Petersburg - September 2003

Usually biosorbent was applied in combination to posing of booms and usage of skimmer's technique. Thus the performance of usage of conventional engineering of gathering of oil increased. However in a series of cases the biosorbent appeared by the alone remedy of oil spill. For example, in Komi republic marshed coasts of the rivers and the shallow depths eliminated usage of high-gravity engineering. In autonomy mode biosorbents were used at oil spill in the Neva labium. 3-4 ball storms confined applying ship facilities. Rather effective there was a biosorbent and at liquidation of oil films on water near to oil terminals. If in cleaning of water and near-shore area from oil products of know-how such as ours biosorption is active are developed (including usage of fertilizers), in cleaning of bottom of such attempts it is not enough. According to our reckoning usage of biosorbents is perspective in this direction too. Such technologies allow conduct-refining measures minimally encompassing an ecosystem of applying. It specially is important at impurities reserved and apart of guarded territories. In this connection we work not only above new types of such biosorbents, but also above adequate means on their applying. Such facilities should provide not only drawing of biosorbent on a place of impurity, but also to realize depositing fragments of biosorbent on depth not less than 5-7 cm in soil in shore or base of the contaminated regions. Certainly biosorbents are only a part of a big arsenal of facilities for oil combating in cases of oil spills at open sea and freshwater areas. In too time such technological methods in some cases are alone alternative in combating with progressing oil impurities of the sea and other's water

objects.

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Tables.

Table 1 Material of the preparation "Biosorb"

<b>Si O<sub>2</sub></b>	<b>50-60%</b>	<b>- Bio components - 3-4% including:</b>
<b>Mg O</b>	<b>13-14%</b>	<b>- phosphorus</b> 2-3%
<b>Al<sub>2</sub>O<sub>3</sub></b>	<b>13-14%</b>	<b>- nitrogen</b> 7-8%
<b>K<sub>2</sub>O+Na<sub>2</sub>O</b>	<b>2-5%</b>	<b>- carbon</b> 45-65%
<b>Ca O</b>	<b>1-2%</b>	<b>- other components</b> 24-46%
<b>H<sub>2</sub>O</b>	<b>2-4%</b>	<b>-Natural water bacteria</b> 4-5% (dry weight)

Table 2. Properties of the preparation of the group "Biosorb-A"

1. Volume weight (kg/m <sup>3</sup> )	<b>140 -180</b>
3. Decrease of interphase tension water/oil (erg/cm <sup>2</sup> .)	Up to <b>2,5-1.8.</b>
4. Absorption of oil from water surface kg oil / kg Biosorbent	<b>5-8</b>
5. Relative hydrophobicness, (%)	<b>85-90</b>
6. Flotation property, (%)	<b>80-85</b>
7. Biodegradation activity in the water of preparation during 10-18 day (%)	
Aerobic conditions, <b>Temperature: 0<sup>0</sup> - +10<sup>0</sup> C</b>	<b>16-45</b>
<b>Temperature: &gt;+10<sup>0</sup> C</b>	<b>40-75</b>

Table 3 The cascade likes effect of the preparation "Biosorb-A" usage.

<b>Type of effect :</b>	<b>Time of effect :</b>
1. Destruction of oil film or localization of oil spot. («physical-chemical booms»)	0,5- 1 h
2. Sorption of oil.	2-4 h
3. Start of the Biodegradation	2-8 h
4. Activation of natural self- -purification of the water from oil (bioremediation).	10-18 h
5. Purification of water surface and column from oil (> 75%) Temperature : 0 <sup>0</sup> C -> +20 <sup>0</sup> C ( aerobic)	7-25 days
6. Destruction of oil (50-60%), in bottom deposits (aerobic process) Temperature: > +3+4 <sup>0</sup> C	60->90 days
7. Destruction of oil on the preparation in shore line (65-75%), Temperature: 0 <sup>0</sup> C -> + 20 <sup>0</sup> C	30- >60 days
8. Destruction of oil (30-40%), in bottom deposits under the conditions of semi anaerobes Temperature: > +3+4 <sup>0</sup> C	>150 days

Table 4 Some modifications of the preparation "Biosorb"

Type of the Preparation	Indication to Usage	Dominating Activity (in priority order)
?	Autonomous applying for clearing of a surface of water on opened water areas	Flotation Surface activity Absorption Biological
?	Applying together with mechanical gathering of oil on opened water areas	Surface activity Flotation Absorption Biological
?	Biosorbent for clearing of swamps.	Biological Absorption Physics-chemical Flotation
D	Is applied to clearing of bottom sediments and coast zone.	Biological Physics-chemical Absorption Flotation
?	Biosorbent powder for clearing of a coast zone of the rocky nature	Absorption Biological Physics-chemical
F	Biosorbent for clearing of pebbly sites of beach	Biological Physics-chemical Absorption

Table 5 Properties of the preparation of the group "Biosorb-D"

1. Volume weight (kg/m <sup>3</sup> )	<b>600-700</b>
2. Bioabsorption of oil from water surface kg <sub>oil</sub> / kg Biosorbent	<b>3-5</b>
3. Relative hydrophobicness, (%)	<b>75-80</b>
5. Biodegradation activity in the sediment (0-5 cm deep) of preparation during 60-90 day (*)	
Aerobic conditions, <b>Temperature: 0<sup>0</sup> - +10<sup>0</sup> C</b>	<b>12-24</b>
<b>Temperature: &gt;+10<sup>0</sup> C</b>	<b>20-65</b>

**The notices to table 6:** (\*) - If necessary, after 30-40 day of applying of biosorbent, is conducted after-treatment of entity. Thus, the complete cycle of clearing of bottom requires 2-3 series treating of the contaminated object.

Table 6. Performance of biosorbent "Biosorb-D", compared with check samples, in sand bottom sediments. (depth - 1 meter)

Test	Method of testing (1).	Source amount of oil in send (mg)	Soluble fractions (%)	Biological destruction (%)	Aggregate quantity of decrease of oil (%)
Boiler oil Check Biosorb	1	456* 423	12 8	0 43	12 51
Boiler oil Check Biosorb	2	460 428	10 7	0 37	10 44
Boiler oil Check Biosorb	3	494 451	5 3	0 27	5 30

The notices to the table 5: (1). Methods of the Tests:  
 Incubation time - 6 weeks

- \* - Quantity of boiler oil in sand the ambassador 1 hour of incubation (0 - mark) mg \ 100 g of sand
- 1 - + 20<sup>0</sup> C; static; brackish water (Finnish bay);
- 2 - + 10<sup>0</sup> C; static; brackish water (Finnish bay)
- 3 - + 2+4<sup>0</sup> C; static; brackish water (Finnish bay);

Table 7. Comparative performance of biosorbent in clearing of soil of different petroleum in vivo experiment for 3 summer months (T<sup>0</sup>C: + 14 + 20<sup>0</sup>).

Type of a soil pollution	Total oil products mg / kg <sup>-1</sup> of soil	N Total mg/ kg <sup>-1</sup>	NH <sub>4</sub> <sup>+</sup> -N Total mg/ kg <sup>-1</sup>	P Total mg/ kg <sup>-1</sup>	pH	Dr. W. %	Organic matter %	% destruction of oil product	
								Biosorb <D>	Check
1. Crude oil	2400	780	23	310	6,6	87	3.6	65	28
2. Diesel fuel	1700	1490	54	284	6.0	76	11.4	44	19
3. Gasoline	700	300	22	220	6.2	91	3.5	95	43

Captions to figures

Figure 1. The tactical scheme of applying of biosorbent in distresses.

Figure 2. Coal-face works on coast of Spain in November 2002 (photo BBC 19.11.2002).

Figure 1

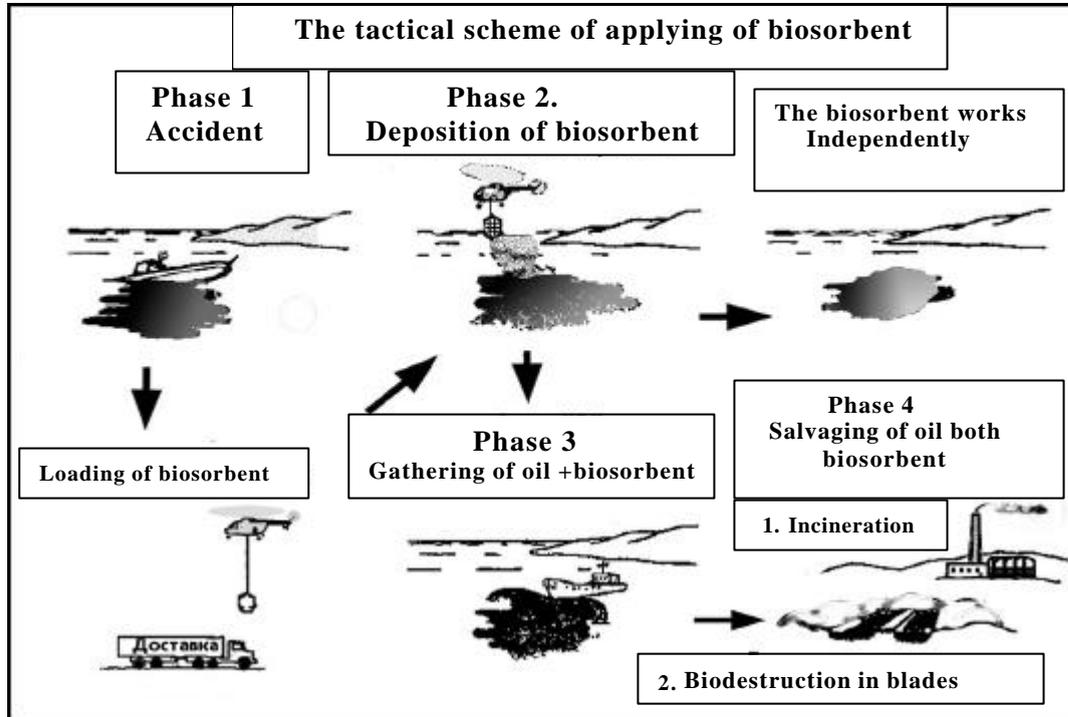


Figure 2

