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New Approaches to Chemical Technologies of Plant Materials for Aromatherapy

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Abstract

A new approach to the production of commercial products used in aromatherapy and household aromatizing agents based on induction heating of plant raw materials and the use of hydrophilic polymer hydrogels is proposed. It is shown that obtaining highly purified essential oils is neither technologically nor economically justified from the point of view of their use in aromatherapy. The proposed approach makes it possible to obtain products for aromatherapy with minimal processing of raw materials and low production costs. The main end product is a polymer hydrogel saturated with a liquid phase formed during induction heating of a mixture of a plant component with metal inclusions. Such a product, among other things, allows the implementation of electronic aromatherapy systems and household aromatizing agents, in which the generation of aroma oils is also provided by induction heating. In the operation of such systems, the basic property of thermosensitive hydrogels is used – a shift in the hydrophobic-hydrophilic balance with temperature variations, which makes it possible to exclude parasitic evaporation of volatile components. Specific technical solutions that implement this approach are proposed.

1. Introduction

It has now been proven that aromatherapy products can have a more than the noticeable effect on a person's health [1, 2], including his/her psycho-emotional state [3]. Such methods are widely used for the rehabilitation of patients, including maintenance therapy [4], etc. Natural aroma oils, in particular, can also have a beneficial effect on people who are under constant stress [5, 6].

This factor in modern conditions becomes very significant. Indeed, as it was shown, in particular, in works [7, 8], a sharp change in the usual way of life of people, caused by strict quarantine taken due to the Covid-19 pandemic, had a negative impact on the psychophysiological state of a very high number of people. In the context of significant geopolitical turbulence, in particular, the associated threat of a global food crisis, it is obvious that the need to improve methods of psychological correction will only increase. Factors such as burnout syndrome (recognized by the World Health Organization as a disease [9]) cannot but increase the negative consequences of crisis phenomena even in those countries where the economic situation remains relatively prosperous.

This issue is directly related to extremely specific aspects of health protection. Indeed, it is now recognized that the occurrence of many diseases is associated with psychosomatic factors, i.e. those that, among other things, are directly dependent on various stress factors due to the impact of the environment on the psychological state of the individual, etc. [10, 11]. For example, some arguments force even obesity to be included in this category [12].

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It is also important to emphasize that it is aromatherapy that is a very convenient area for practicing the mass use of telemedicine, which is also becoming more and more relevant at present [13, 14]. Namely, aroma oil generators can be made very compact, since the approach proposed in [15, 16] applies to them, according to which the control functions of an electronic device are carried out through a program installed on the user's cell phone. This approach also provides the possibility of pairing aromatherapy products with the Internet, receiving recommendations in a remote/automatic mode, etc.

We emphasize that the possibility of mass use of aromatherapy products allows raising the question not only of improving the life quality of individual consumers but also of improving society as systemic integrity. The consideration of society as an analog of a neural network [17] acquires a new impetus in the case of the publication [18], in which a similar idea was substantiated by means of physics. To date, it can already be considered as established that complex systems of any nature, including those related to different levels of organization of matter, can be considered based on analogies with neural networks. For systems based on hydrophilic polymers, this conclusion was substantiated in [19] using the results of [20].

The mass use of aromatherapy systems makes it possible to use the neural network theory of society in practice, but for this, among other things, it is necessary to significantly reduce the cost of the aromatic components themselves, expand their range, and ensure accessibility for a wide range of consumers. These tasks are also of interest from the point of view of standard methods of using aromatherapy.

To solve these problems, this paper proposes a new approach to the technology of processing plant raw materials to obtain products used in household aromatizing agents and aromatherapy products.

2. Formulation of the problem

The production of aromatic (essential) oils traditionally used in aromatherapy requires the implementation of high-tech production [21]. Not all essential oils, the usefulness of which has been proven by clinical practice, are currently produced industrially, and, strictly speaking, there is no need to obtain highly purified essential oils.

Indeed, any aromatizing agents, including those involving remote control, involve the generation of very small amounts of vapors of essential oils. Technically, it is more convenient to use various types of compositions, and the proportion of the aroma component is relatively small. It is these compositions (for example, polymer networks saturated with aromatic components) that are now on the market. It is significant that the use of compositions also makes it possible to block the spontaneous evaporation of essential oils, which is very important for the implementation of controlled generators of their vapors.

Therefore, it seems appropriate to raise the question of the direct production of compositions containing essential oils from plant raw materials, which is a much simpler task from the point of view of chemical technology.

The easiest way to prove this thesis is as follows. There is a possibility to realize household aromatizing agents that directly use plant raw materials. For example, it is acceptable to use technology similar to that used in tobacco heating systems Glo™ [22]. In this case, the herbal product containing nicotine is placed inside an electrically conductive shell, which is heated by a high-frequency electromagnetic field. Simplifying, the dry distillation of crushed tobacco leaves is carried out.

The direct use of plant raw materials in household aromatizing agents, however, is also not entirely convenient. It is much more convenient to isolate the liquid component containing essential oils and natural surfactants from them and saturate the polymer network (polymer hydrogel) with it, thereby obtaining a product suitable for further use in household aromatizing agents.

It should be emphasized that at present many types of hydrogels have been synthesized, which simultaneously include both hydrophilic and hydrophobic functional groups. An example of such hydrogels are cross-linked polymer networks based on poly- N-isopropylacrylamide, which were emitted, in particular, in [23]. A significant part of thermosensitive hydrogels also belongs to the same class, the ability of which to change the degree of swelling with temperature variations is determined precisely by the shift in the hydrophobic–hydrophilic balance [24]. The presence of hydrophobic groups in the composition of networks allows them to effectively interact with the hydrophobic component, which has been repeatedly demonstrated, including in experiments between gels of this type and surfactants [25]. This makes it possible to ensure the saturation of highly swelling hydrogels with essential oils, despite the pronounced hydrophobic nature of the latter. In addition, the saturation of hydrogels with a hydrophobic component can be ensured by using surfactants as mediators.

Thus, the purpose of this work is to develop the design of a device that makes it possible to obtain products intended for use in aromatizing agents most simply and cheaply. As will be clear from the following, the proposed approach allows to simplify the design of the device for the specified purpose so that it can be operated directly in the field, including by small agro-industrial businesses.

3. The design of the device for the production of products for aromatherapy based on polymer hydrogels

The functional diagram of the device for the production of products for the aromatherapy based on polymer hydrogels is shown in Fig. 1.

The entire structure is mounted in an outer cylindrical body (1). Crushed plant raw materials (2), mixed with metal inclusions, are loaded into a cylindrical cartridge (3), on top of which an induction heater (4) is located. The module has a mesh bottom (5). The heater (4) is a rigid coil fixed to a seal (6) in the upper part and a seal (7) in the bottom.

Through the seal (7) and outlet (8), the cartridge (3) is connected to the tank (9), which is filled with powdered hydrogel (10).

The heating of plant raw material is carried out due to electromagnetic oscillations that occur in the space inside the cartridge (3) due to the alternating current flowing through the coil (4). An alternating current is generated by a generator (11). Control over the change in the state of the hydrogel is carried out using a capacitive sensor (12). The generator (11) and the sensor (12) are connected to a control unit based on a microcontroller (13), which is connected via a radio channel to the user's smartphone (14), on which the control program is installed. The diagram also shows the power supply unit (15).

The advantage of the proposed construction is the possibility of direct production of aromatic products using the cheapest possible design and minimal energy consumption. In accordance with the above, all products formed during the dry distillation of plant materials are accumulated by the hydrogel, i.e. the technological stage of separation of the distillation products is excluded as technologically and economically unjustified (in any case, unjustified from the point of view of products intended for use in household aromatizing agents).

Optimization of energy costs is ensured due to the fact that during induction heating, only the raw material located inside the cartridge (3) is heated, i.e. heat is spent targeted.

We emphasize that optimization in terms of energy consumption is of interest, including from the point of view of the application of the developed device. Providing that the energy consumption is low enough, it becomes possible to use power from solar photovoltaic panels (PVC), and therefore, to make the equipment that ensures the processing of raw materials as autonomous as possible, up to the possibility of its operation in the field. It should be noted that to provide a power supply from PVC, there are currently all the necessary prerequisites

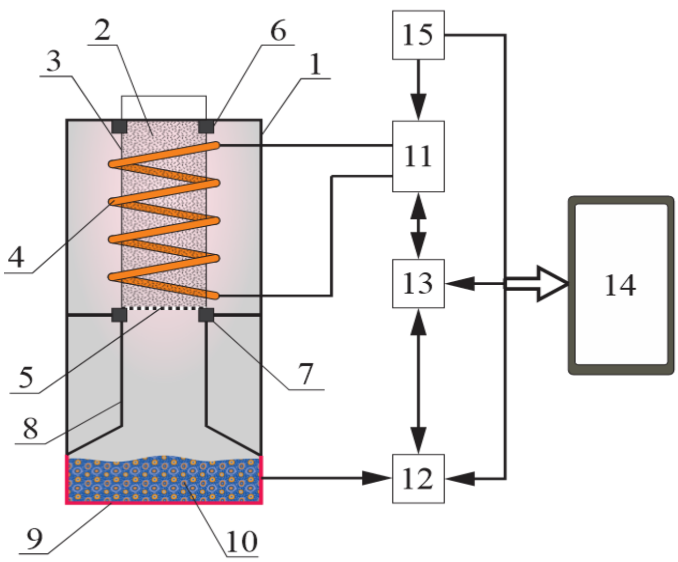

Fig. 1. Functional diagram of a device for the production of aroma products based on polymer hydrogels.

[26, 27], up to the use of optical neural networks [28], which can provide regulation of light fluxes. Moreover, the issue of energy independence of small and medium-sized businesses, as well as rural households, is becoming increasingly acute [29].

The advantage of the proposed approach is also the fact that the production of aroma products, depending on the formation of logistics chains, can be made practically waste-free (in terms of obtaining a commercial product oriented to the end consumer). Namely, the so-called sachets ("dry perfumes") are widely represented on the market, which is a bags of dried plant components (lavender, rose petals, etc.). Losses of the aromatic component during the processing of plant raw materials using an electromagnetic field according to the proposed technology are comparable to those that occur during conventional drying. Consequently, both resulting products – dried plant raw material remaining in the cartridge (3), Fig. 1, and hydrogel saturated with the aroma component are commercial products.

Further, the effectiveness of the so-called business ecosystems [30, 31], formed on the principle of focusing on the interests of consumers, has now been proven. To the technology considered, this approach corresponds to the formation of the entire consumer chain – from the cultivation and processing of raw materials to sales to end consumers. Based on this, the production of devices, the scheme of which is shown in Fig. 1, should be supplemented with the production of household aromatizing agents that use the resulting commercial product.

It should be noted that there is an opportunity for further improvement of the proposed approach. It is associated with the effect of redistribution of concentrations, which occurs, among other things, during contact between gels with unequal network charge density [32]. This effect makes it possible to ensure the separation of the liquid-phase component from plant raw materials using the proposed method through the use of several types of hydrogels, each of which provides the predominant accumulation of components of a certain type.

4. Experimental verification of the extractor performance

In the experiments, the design described above was used, in which a thermosensitive hydrogel based on N-isopropylacrylamide was used as a sorbent, containing hydrophilic and hydrophobic groups in its structure, which makes it possible to absorb all released substances assuredly.

We emphasize that, by the main idea of the proposed method, the role of a hydrogel or other sorbent is to completely accumulate all substances released during the heating of the initial plant material. We are not talking about pure essential oil, there is no need for it. Moreover, the use of purified essential oil creates additional difficulties in the design of household aromatizing agents due to its increased volatility; it is much more convenient to use a gel that contains a wide range of natural components in terms of controlling the separation of the aroma component.

Most of the evaporation is the water contained in the plants, which provides interaction with the hydrogel or other sorbent. The location of the gel in the cell de facto does not matter, since a closed volume is used, which is acceptable since the task of vapor separation is not set. It also allows the use of gels or sorbents of a wide variety. This approach is acceptable since the gel or another sorbent is then used as a filler in the flavoring cartridge, in which reheating takes place.

Below are the results related to the study of the mixture absorbed by the sorbent and then separated from it due to additional heating with the help of a household aromatizing agent.

The objects of study are fresh leaves of peppermint (*Mentha piperita L.*) and fragrant basil (*Ocimum basilicum L.*) of the *Lamiaceae* family, collected in the flowering phase. Fresh leaves of plant raw materials were purchased in retail trade in Almaty, Kazakhstan. The initial moisture content of each plant was respectively 92% for peppermint, 90% for basil.

The processes of extracting valuable components from natural raw materials by the method described above were studied, and the influence of the main parameters on the extraction of valuable components from the above-mentioned plant raw materials was studied. With the optimization of extraction, the parameters that affect the yield of valuable components from plant raw materials, such as the degree of crushing raw materials, the extraction time, and the power invested in the working chamber of the device described above, were studied (Table 1).

From the data in Table 1 it follows that the yield of essential oils of peppermint and fragrant basil increases as the size of the material decreases. When changing the size of plant leaves from whole

Table 1
The parameters of the extraction process of the proposed type from plants *Mentha piperita L.* and *Ocimum basilicum L.*

Parameters under study	Yield, %	
	Peppermint (*Mentha piperita L.*)	Fragrant basil (*Ocimum basilicum L.*)
Selection of the degree of crushing of raw materials		
1–2 mm	0.18	0.11
3–5 mm	0.30	0.32
6–8 mm	0.49	0.45
9–10 mm	0.23	0.24
20–30 mm	0.21	0.22
The effect of time on the extraction process		
30 min	0.13	0.13
45 min	0.14	0.15
60 min	0.49	0.50
75 min	0.62	0.59
90 min	0.70	0.66
105 min	0.41	0.38
Power input		
15 W	0.39	0.37
30 W	0.50	0.52
45 W	0.65	0.61
60 W	0.63	0.60

to crushed, the yield of essential oil increases from 0.11 to 0.49%. The highest yield of valuable components is provided with a raw material particle size of 6–8 mm. The use of finer raw materials (1–4 mm) leads to the occurrence of parasitic processes associated, among other things, with the separation of high-molecular compounds during heating.

With an increase in the extraction time from 30 to 90 min, the essential oil yield gradually increases from 0.13 to 0.70% for 90 min, but decreases to 0.38% for 105 min due to the denaturation of some substances in the oil caused by prolonged exposure to high temperature.

If the temperature is raised too high, some temperature-sensitive substances in the essential oil may decompose, negatively affecting the extraction yield, oil quality and production cost due to increased energy consumption. In general, it has been established that it is expedient to carry out the extraction of essential oils from plant raw materials by the proposed method at an input power of 45 W, an extraction time of 90 min, and a degree of raw material crushing of 6–8 mm.

The obtained oils were transferred for the study of the component composition by gas-liquid chromatography using mass spectroscopy (GC-MS). The results of the analysis are presented in Table 2.

The component composition of essential oils was studied on a TRACE 1310 gas chromatograph (Thermo Fisher Scientific, USA) with a triple quadrupole mass spectrometric detector TSQ 8000 (Thermo Fisher Scientific, USA). Chromatographic separation was performed using a TG-5SILMS capillary column (30 m × 0.25 mm × 0.25 μm). The carrier gas is helium with a constant flow of 1 ml/min. The initial temperature of the column oven is 70 °C with a delay of 2 min. Then the thermostat was heated to 200 °C at a rate of 5 °C/min with a delay of 1 min, heated to 285 °C at a rate of 10°C/min with a delay of 10 min, followed by a decrease in temperature to the initial state of 70 °C. The temperature of the injector was 250 °C and of the mass spectrometric detector was 285 °C. The extract was injected in a volume of 1 μl in the split mode flow (split ratio 1:10). Ionization mode EI was 70 eV, ion source temperature was 230 °C, Full scan mode within m/z range was 50–550. The chromatography process was controlled using the XCalibur program. The biologically active compounds recovered from the two extractions were identified based on the retention time of on the GC column using a library of reference mass spectra with data from the National Institute of Standardization & Technology (NIST) database for GC-MS systems.

As a result of the research, the chemical composition of the essential oils of the leaves of *Mentha piperita L.* (*Lamiaceae*) was studied after extraction by the proposed method (EPM) in comparison with hydrodistillation (HD).

5. The construction of the household aromatizing agent

The functional diagram of a household aromatizing agent oriented to the use of a hydrogel saturated with an aroma component, according to the above technology, is shown in Figs. 2 and 3 show a photograph of a prototype obtained using a 3D printer.

The aromatizing agent is oriented towards the use of disposable cartridges, which are commercial products manufactured using the technology described above.

Table 2
Chemical composition of essential oils of peppermint (*Mentha piperita L.*) leaves obtained by the proposed method (EPM) and hydrodistillation (HD)

No	Component	Retention time, min	Content of components, % wt.	Retention time, min	Content of components, % wt.
		EPM		HD	
1	2,4-Dimethylpentane	1.82	0.65	1.82	0.57
2	*n*-Hexane	1.87	0.60	1.87	0.50
3	3,4-Dimethylhexane	-	-	1.93	0.99
4	Methylcyclopentane	1.96	1.88	1.96	1.33
5	Cyclohexane	2.09	0.68	2.10	0.98
6	*α*-Pinene	5.10	0.49	5.12	1.24
7	Sabinene	5.92	0.73	5.92	1.69
8	*β*-Pinene	6.06	1.13	6.09	2.18
9	*β*-Myrcene	6.22	2.42	6.25	2.99
10	2,6-Dimethyl-1,3,5,7-octatetraene, *E,E*-	6.64	0.39	6.67	1.25
11	Limonene	7.25	9.49	7.39	10.79
12	*trans*-*β*-Ocimene	7.39	1.85	7.67	0.98
13	1,8-Cineole	7.46	2.83		
14	3-Oxatricyclo[4.1.1.0(2,4)] octane, 2,7,7-trimethyl-	-	-	8.37	0.37
15	Terpinolene	-	-	8.71	0.39
16	Linalool	9.08	1.40	9.10	1.51
17	2H-Pyran-3(4H)-one, 6-ethenyldihydro-2,2,6-trimethyl-	-	-	9.24	0.26
18	Carvone	13.36	26.61	13.41	17.70
19	Piperitenone	15.70	0.40	15.79	0.74
20	*trans*-Carveyl acetate	16.13	1.83	16.20	1.83
21	Copaene	16.59	0.49	16.65	0.78
22	*β*-Bourbonene	16.85	2.28	16.92	2.79
23	*β*-Elemene	16.93	1.38	17.00	1.81
24	*cis*-Jasmone	17.05	0.40	17.11	0.68
25	*α*-Gurjunene	17.42	1.12	17.48	1.33
26	Caryophyllene	17.81	4.54	17.88	4.92
27	Bicyclosesquiphellandrene	18.39	2.57	18.46	3.24
28	*β*-Farnesene	18.49	2.01	18.56	2.46
29	Humulene	18.65	1.30	18.71	1.30
30	*cis*-Muurola-4(15),5-diene	18.84	3.49	18.90	3.68
31	Germacrene D	18.97	9.59	19.49	9.99
32	Longifolene	-	-	19.58	0.42
33	Bicyclogermacrene	19.67	2.70	19.75	2.95
34	*γ*-Cadinene	20.03	0.71	20.08	0.92
35	Cadina-1(10),4-diene	20.15	1.00	20.20	1.38
36	*cis*-Calamenene	20.21	1.13	20.26	1.32
37	*α*-Muurolene	20.58	0.59	20.63	0.90
38	Germacrene D-4-ol	21.58	0.83	21.61	0.99
39	Viridiflorol	-	-	22.05	0.30
40	Cedrol	22.38	0.65	22.41	1.00
41	Cubenol	22.49	1.60	22.52	1.91
42	*τ*-Cadinol	23.07	0.56	23.10	0.91
43	*α*-Cadinol	23.41	1.58	23.45	1.92
44	(1*R*,7*S*,E)-7-Isopropyl-4,10-dimethylenecyclodec-5-enol	-	-	24.27	0.33
45	Shyobunol	24.58	1.08	24.58	1.34
46	Phytol	32.55	0.82	32.53	1.26
47	Cyclodecasiloxane, eicosamethyl-	34.81	4.22	37.25	0.24
48	Octacosane	-	-	38.08	0.38
49	Tetratriacontane	-	-	40.04	0.26
	Total		100.00		100.00

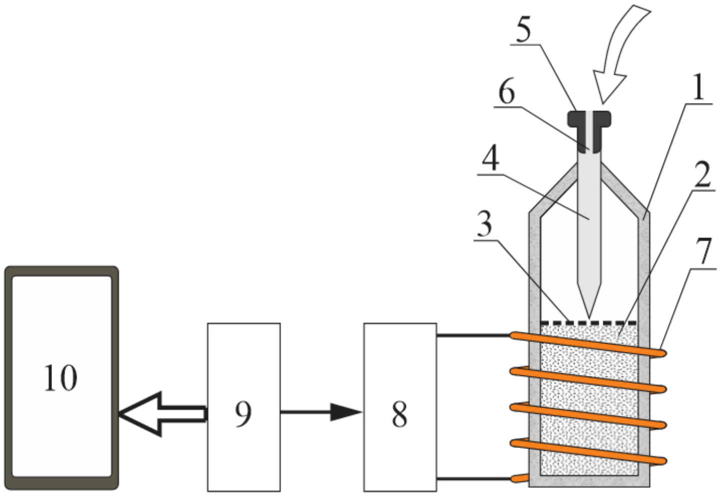

Fig. 2. Functional diagram of a household aromatizing agent with induction heating.

Fig. 3. Photo of a household aromatizing agent's prototype.

The cartridge consists of an outer shell (1), an area (2) filled with a hydrogel saturated with an aroma component, which is protected by an easily tearable partition (3). A punch (4) is located in the upper part of the device, the lower tip of which in the initial state adjoins the partition (3). The upper part (6) of the piercer (4) is permeable to the gas phase medium. The piercer is supplied with a head-stopper (5).

The cartridge is inserted into the socket of the aromatizing agent, which includes an induction heating coil (7), a high-frequency signal generator (8), as well as a control radio-electronic system (9).

The last one includes a Bluetooth module that provides communication with the user's smartphone via a radio channel.

The system is controlled by transmitting data from a smartphone (10), which almost eliminates the need to use any moving parts: buttons, toggle switches, etc., in the construction itself. Due to this, it can be manufactured in a cheap monolithic case.

The advantages of this scheme are that it is not only easy to manufacture, but also focused on the use of the most accessible and cheap radio-electronic components. This is achieved by the fact that the use of specific radio-electronic components is minimized by transferring control functions to the program installed on the user's smartphone.

Another advantage of the scheme presented in Fig. 2 is the possibility of using different types of hydrogels, including thermosensitive ones [33]. These gels, as shown, including in the cited work, are distinguished by a wide variety of characteristics, however, they all contain both hydrophobic and hydrophilic functional groups. The presence of both hydrophobic and hydrophilic functional groups in the hydrogels of the type considered largely determines the additional advantages of the proposed technique. Indeed, the liquid phase formed during the processing of plant raw materials containing essential oils also obviously contains both hydrophobic and hydrophilic components. Accordingly, it becomes possible to provide a controlled separation of essential oils from the polymer matrix due to temperature variations that cause a shift in the hydrophobic-hydrophilic balance. This factor is quite significant: "chemical blocking" of the parasitic evaporation of essential oils is much more effective than any other. The problem of parasitic evaporation of highly volatile essential oils will inevitably face any attempt to provide controlled vapor generation of such oils in order to create methods of electronic aromatherapy.

From an economic point of view, the advantage of the proposed approach is that, in accordance with the principles underlying business ecosystems, it provides the implementation of the entire range of operations that ensure not only the manufacture, but also the sale of a commercial product.

6. Conclusion

Thus, it is possible to implement a simple technology for the processing of plant raw materials, focused on obtaining products used in aromatherapy, as well as for household room aromatization.

The technology is based on the use of induction heating and the accumulation by polymer hydrogel of liquid released from plant raw materials during heating. This technology is based on the fact that in the manufacture of products obtained from the processing of plant raw materials for subsequent use in aromatherapy, there is no need to achieve a high degree of purification of essential oils.

Rather, on the contrary, to create means of electronic aromatherapy, as shown in this work, the use of highly purified essential oils creates quite certain inconveniences.

The advantage of the proposed technology is also the possibility of controlled separation of essential oils from the polymer matrix during heating, which is achieved, for example, by using thermosensitive hydrogels, the behavior of which is determined by the hydrophobic-hydrophilic balance.

Products obtained using the developed technology are intended for use in household aromatizing agents and electronic aromatherapy systems, also proposed in this paper. This closes the logistics chain, which provides for the entire cycle of operations from growing raw materials to completing consumer-oriented household devices, which removes one of the main problems faced by modern developments in the field of chemistry – the problem of promoting new developments to the market.

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