Properties of Nanostuctured Motherwort Extract and Its Application in Fruit Jelly Candy Production

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Abstract—**The research paper includes the information on the application of nanostructured motherwort extract in the production of fruit jelly candy, which can be used as a therapeutic functional food product. The size of nanostructured motherwort extract was determined by a nanoparticle tracking analysis (NTA). It depended on a shell nature significantly.**

Keyword—fruit jelly candy, nanostructured motherwort extract, NTA method, functional products, self-organization

I. INTRODUCTION

Motherwort represents the perennial herb species of *Lamiaceae* family.

According to these authors [1, 2] the results of the clinical studies show that motherwort and its preparations have hypotensive, cardiotonic and sedative activity, as well as antimicrobial, antiflammatory and antioxidant properties. The motherwort is also used for the treatment of gastrointestinal disturbances, thrombosis, epilepsy and Graves's disease.

A pharmacological activity of motherwort raw materials and preparations is caused by the presence of a complex of biologically active substances including iridoids, dihydroxy-cinnamic acid derivatives, flavonoids and others. As noted by the authors of the research [3], the main flavonoids of motherwort herb are rutin, hyperoside and isoquercetin.

This research paper is the continuation of our research of nanostructured bioactive compounds [4-14].

It is well-known that nanometer objects have superior bioavailability. This quality is used both in medicine and in pharmacology. The size of capsules containing bioactive compounds is significant for their physiological activity in an organism [15]. An example of many drug substances shows that the reduction in the capsule size results in change of their bioavailability and efficiency [16].

No works on the study of nanostructured motherwort extract were found in literature.

In 1987 J.M. Lehn, one of the founders of supramolecular chemistry, used the terms of “self-organization” and “self-assembly” in order to describe the ordering of high molecular weight compound systems at equilibrium conditions, for example, DNA formation. Lehn himself defined supramolecular chemistry as chemistry of molecular assemblies and intermolecular bonds, i.e. as chemistry beyond the bounds of molecules. This definition is figurative and not quite precise. The formation of intermolecular bonds cannot depend on the structure of molecules contained in an assembly. There exist many reactions of self-organization and self-assembly because of different types of interaction during which big molecules and molecular assemblies are forming. The common examples can be DNA and various complex compounds of “guest-host” type. If the process takes place in a solution, they can be clathrates or, more generally, inclusion compounds. They can also exist in more solid state, for example, gas hydrates. The components can be crown ethers, cryptoids, iodatids, spherolites, cyclodextrins and so on [17].

It should be particularly emphasized that the processes of supramolecular chemistry are carrying at a nanoscale level and supramolecules themselves are nanoscale. In order to form a supramolecule its components shall have the binding sites with proper electrical specification (for example, a presence of a donor and an acceptor, polarity, possibility to form a hydrogen bond, structure rigidity or softness and so on). Besides, there shall not be any steric hindrances for the processes of supramolecular self-assembly.

Hydrogen bonds play a significant part in supramolecular structures. One of the most interesting classes of supramolecular structures are dendrimers (cascade molecules) – monodispersed macromolecules having highly branched 3D structure. Dendrimer can be considered as a multicomponent compound growing from the central core like a tree. The capabilities of self-assembly of molecules are practically unlimited, as well as the structures of self-generated molecules are also unlimited: capsules, spirals, supramolecular squares, cubes, boxes, dendrimer structures, coordination nanoscale structures, rosette-like structures and others. Various electronic devices are designed on basis of different supramolecular structures: switches, wires, rectifiers, as well as various molecular machines, materials for nonlinear optics and so on.

Biological systems play a significant part in the development of supramolecular chemistry. Many synthetic supramolecular systems were obtained within a biomimetic approach, i.e. by means of imitation of a structure or a function of more complicated biological objects. It is believed that supramolecular chemistry originates from

Fischer`s model “key in lock” used in enzymatic catalysis, which was known long before the appearance of complicated systems of cryptoid types and self-assembling devices and systems. This is a model of consistency between a substrate form (guest) and a receptor (host). “Substrate-receptor” binding is often very selective and plays a very important part in biochemistry. It is reversible. To a great extent the substrate binding is often required for induction of conformational change in the receptor, which includes a biochemical process.

The study of creation of a complex nanostructure and its evolution during a crystallization process also required a description of these phenomena as self-organization. However, as opposed to a synergetic approach these phenomena take place in conditions near to thermodynamic equilibrium [18, 19].

The nature of self-organization in supermolecules and supramolecular crystals remained practically unstudied in spite of a large number of publications on supramolecular chemistry. This uncertainty even relates to the terminology used in supramolecular chemistry. First of all the issue is about the so-called self-assembly and self-organization processes, which are sometimes (but not often) distinguished. “Self-assembly” term has broader meaning. It includes any types of spontaneous binding of the components with the use of both of covalent and noncovalent interactions. Self-organization includes the interaction of systems capable of spontaneous ordering in space and/or time, as well as space (structural) order and temporal (dynamic) order both in even dissipative structures and in uneven ones. It only relates to a noncovalent supramolecular level and brings to the formation of polymolecular assemblies due to specifically interacting events of recognition of molecular components of each other.

The higher the level and dimension of 3D organization of the fragments is the more reasons there are for considering them as organizational structures (molecular layers, membranes, micellas, colloids, liquid crystals, molecular crystals). That is to say, the self-organization includes coordinated interaction between the parts and integration of these interactions resulting in collective behavior of a system (which can be observed, for example, during transitions or during the occurrence of spatial or temporal waves).

We were the first to carry out a comprehensive study of the influence of the shell nature on a nanocapsule size through the example of motherwort extract.

II. EXPERIMENTAL

The subject of the research was nanostructured motherwort extract in different carbohydrate shells. Sodium alginate, sodium carboxymethylcellulose, konjac gum and carrageenan were used as shells.

The powder of nanostructured motherwort extract was dissolved in water in order to examine the nanocapsule self-organization. The concentration of solutions was 0.125% and 0.25%. A drop of the solution was applied on a cover-glass and was evaporated. The dried-up surface was scanned by a confocal microscopy method on OmegaScope microspectrometer (produced by AIST-NT Zelenograd) combined with a confocal microscope.

The size of nanoparticles was examined by NTA (nanoparticle tracking analysis). Capsule supramolecular properties were also studied be means of self-organization.

III. RESULTS AND DISCUSSION

Supramolecular chemistry uses the laws of organic synthetic chemistry for obtaining supramolecular assemblies, the laws of coordination chemistry of complexes and physical chemistry – for study of component interaction and biochemistry laws – for examination of functioning of supramolecular assemblies. Supramolecular properties includes self-assembly and self-organization [17, 18]. Noncovalent interactions [19, 20] are applied in supramolecular chemistry for obtaining of controllable assembly of molecular segments and spontaneous organization of molecules in a stable structure. Self-organization structures can be imitated as aspects of biological systems: artificial cells of membranes, enzymes or channels [21].

The obtained results are demonstrated in Fig. 1.

The fractal compositions are capable of self-organization as they were found in aqueous solution of nanocapsules at their rather low concentration. Nanocapsules appear spontaneously because of a noncovalent interaction, which means that the self-assembly is typical of them. Therefore, the nanostructured motherwort extract has supramolecular properties.

The size of particles of nanostructured motherwort extract in different shells is shown in Fig. 2-5 and Table I.

Therefore, taking into account the chemical composition of motherwort extract nanocapcules and the size of nanocapcules in different shells it can be assumed that it is reasonable to use the nanostructured motherwort extract when making functional foodstuff for the purpose of better digestion of bioactive substances.

Taking into consideration the positive influence of nanostructured motherwort extract on the organism it is recommended to include it as an ingredient in well-liked delights – fruit jelly candy – in an amount of 0.025%. In this case both organoleptic and physical-and-chemical parameters of fruit jelly candy remain unchanged (Table II), which is very important for a consumer. Its bioavailability will increase due to bioactive substances of motherwort extract. It is important in conditions when a human organism does not have sufficient amount of bioactive substances.

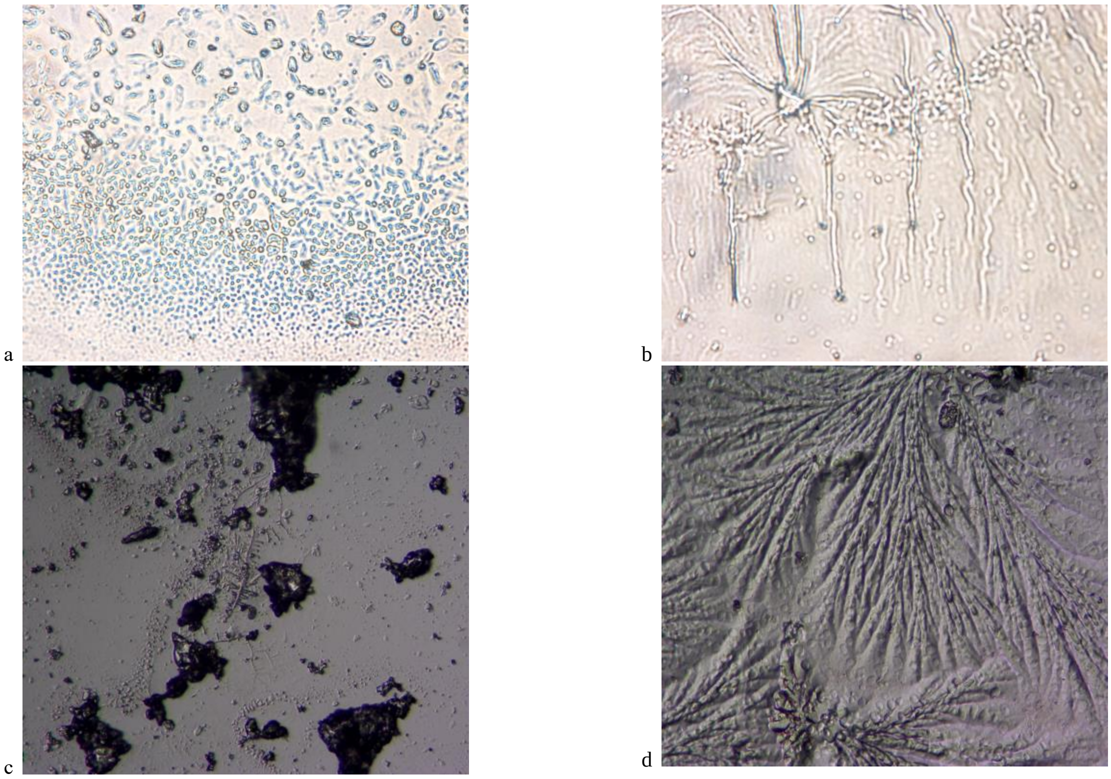

Fig. 1. Confocal image of nanostructured motherwort extract: a) in sodium alginate, magnification – 920 times, concentration - 0,125%, core : shell proportion is 1 : 3; b) in konjac gum, magnification – 920 times, concentration - 0,125%, core : shell proportion is 1 : 3; c) in carrageenan, magnification - 1200 times, concentration – 0,125%, core : shell proportion is 1 : 3; d) in sodium carboxymethylcellulose, magnification - 400 times, concentration – 0,25%, core : shell proportion is 1 : 3.

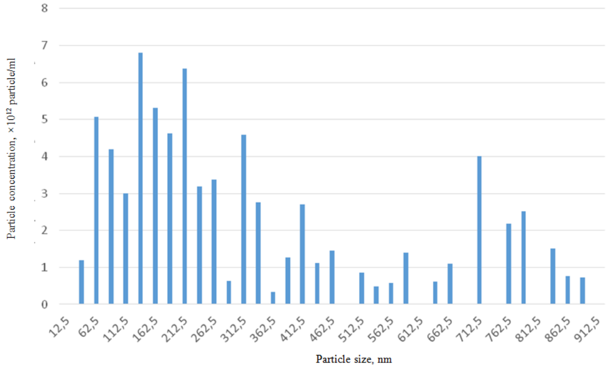

Fig. 2. Particle-size distribution in a nanocapsule sample of nanostructured motherwort extract in sodium carboxymethylcellulose (core : shell proportion is 1 : 3).

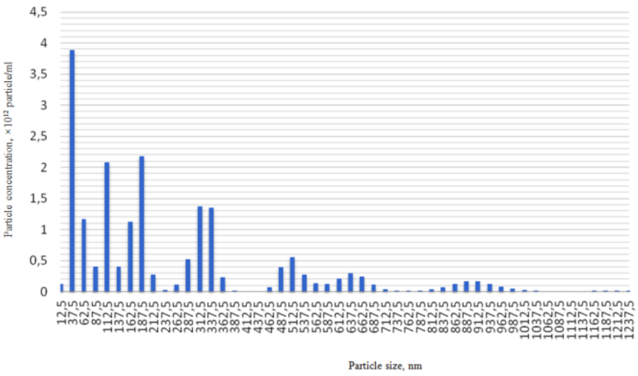

Fig. 4. Particle-size distribution in a nanocapsule sample of nanostructured motherwort extract in sodium alginate (core : shell proportion is 1 : 3).

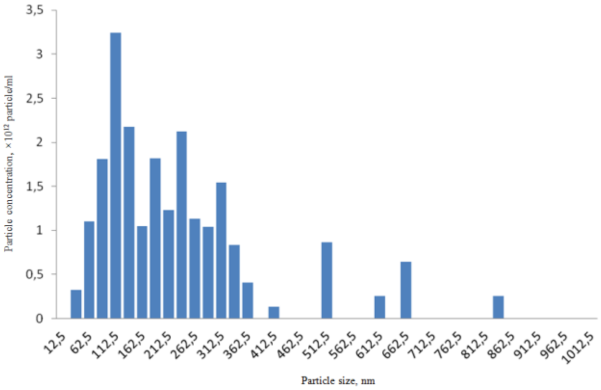

Fig. 3. Particle-size distribution in a nanocapsule sample of nanostructured motherwort extract in carrageenan (core : shell proportion is 1 : 3).

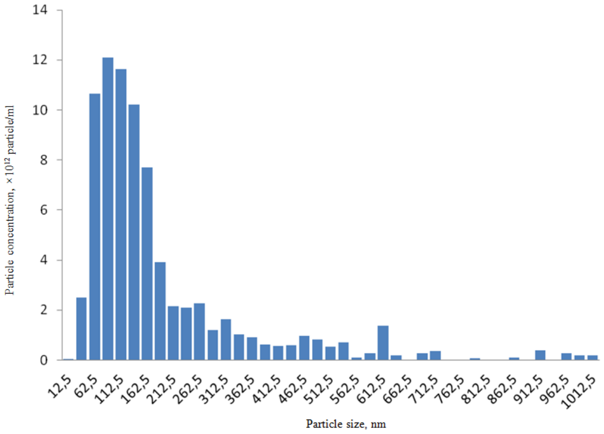

Fig. 5. Particle-size distribution in a nanocapsule sample of nanostructured motherwort extract in konjac jum (core : shell proportion is 1 : 3)

TABLE I. DISTRIBUTION STATISTICS

Characteristic	**Shell**			
	Sodium carboxymethyl-cellulose	***Carrageenan***	***Sodium alginate***	***Konjac jum***
Medium size, nm	322.00	227.00	249.80	192.00
D10, nm	81.00	88.00	25.00	63.00
D50, nm	228.00	190.00	152.10	129.00
D90, nm	741.00	381.00	579.20	421.00
Polydispersity index, (D90-D10)/D50	2.89	1.54	3.64	2.78
Total particle concentration, ×1012 particle/ml	0.75	0.22	18.80	0.79

TABLE II. ORGANOLEPTIC AND PHYSICAL AND CHEMICAL PARAMETERS OF FRUIT JELLY CANDY

Parameter	**Characteristic**
Taste	Typical of the given type of fruit jelly candy
Color	Light-yellow, typical of apple sauce
Flavor	Typical of the given type of fruit jelly candy, no foreign flavor
Surface	Shiny, even
Consistency	Jelly-like, tender
Acidity, degree	5.5-5.6

IV. CONCLUSION

Therefore, the obtained results show that the size of motherwort extract nanocapsule depends on the nature of carbohydrate shell significantly: the size of 10% of nanocapsules is from 63 to 88 nm; the particles only have smaller size of 25 nm in sodium alginate. That said, the smallest medium size of nanocapsules is 192 nm in konjac gum, medium size of 227.00-249.80 nm is in carrageenan and sodium alginate, and correspondingly, the biggest medium size (322 nm) is in sodium carboxymethylcellulose.

Maximum polydispersity index of 3.64 is observed in the particles covered by a sodium alginate shell, the particles in a konjac gum shell (2.78) and in a sodium carboxymethylcellulose shell (2.89) have mean index and nearly similar indices; the particles in a carrageenan shell (1.54) have minimum index, which makes it possible to conclude that in such case the motherwort extract nanocapsules approach to a globe-shaped form.

Introduction of nanostructured motherwort extract to fruit jelly candy ingredients makes it possible to intensify the finished product features and to enlarge the range of special-purpose confectionery products.

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