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# EXTRACTION FROM SOLID MATERIALS IN AN ELECTROMAGNETIC FIELD OF SUPERHIGH FREQUENCIES

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*On the basis of the kinetic theory of fluids and the theory of SHF-heating of dielectrics, a theory of extraction is developed and kinetic equations are obtained. The adequacy of the equations is confirmed by experiments. Traditional extraction (without SHF-heating) is a particular case of the general solution. SHF-heating allows one to increase the rate of extraction considerably with a low consumption of energy.*

Heating in an electromagnetic field of superhigh frequencies (SHF-heating) is widely used for treatment of various materials [1, 2]. Its basic advantages over traditional methods of heating (contact, convective, etc.) are the short duration and the high efficiency of energy consumption. The technology of SHF-heating is also being developed [3]. However, the influence of an SHF-field on the processes of mass transfer (extraction, adsorption, etc.) is not yet clear. Rare publications [4, 5] pointed to a noticeable acceleration of extraction of vegetable oils on exposure to an SHF-field, which was found only empirically.

It is known [6] that the internal structure of particles and their size have a decisive influence on the rate of extraction. The changes in these parameters in different technologies of preparing materials for extraction has not noticeably accelerated the process in recent decades, since the mechanism of internal mass transfer remains unchanged, namely, molecular, that is, this process is known to be slow. At a temperature of the liquid inside the particles close to or equal to its boiling point the molecular mechanism of internal mass transfer is replaced by a convective one and, as already shown long ago [6], the rate of the process increases sharply. But a method for attaining this state has not been developed as yet. This can be done by means of SHF-heating.

The work is based on the following methodological principles:

1. For a solid–liquid system the laws of liquid established by its kinetic theory are used [7]. This means that for a liquid contained inside a porous solid, in particular, the elementary translation of a molecule and the period of its oscillations about the equilibrium state remain the same as for the free liquid. This statement is based on the fact that the size of the pores of the material prepared for extraction is considerably larger than the elementary translation of a molecule ($10^{-10}$ m) [7]. Thus, in [8] it was shown that the diameter of the pores in a raw soybean petal varied within the limits $1.4 \cdot 10^{-8}$–$8.4 \cdot 10^{-5}$ m, with the main amount (77%) belonging to macropores (with a diameter of $2 \cdot 10^{-7}$–$8.4 \cdot 10^{-5}$ m), while in the particles of a prepressed oil cake of sunflower seeds all the oil was contained in pores with a diameter of $2 \cdot 10^{-7}$–$7 \cdot 10^{-6}$ m. The size of the particles subjected to extraction varies from tenths of a millimeter (a petal) to several millimeters (a grit). A generalization of the parameters of the porous structure of sorbents [9] shows that the diameter of the prevailing pores varies within the limits $1.6 \cdot 10^{-9}$–$10^{-8}$ m. Sorbents consist of finely divided material and are usually characterized by a specific surface that varies within the limits $(3.0–7.5) \cdot 10^5$ m$^2$/kg. Sometimes sorbents are subjected to extraction to extract an absorbed oil. The inhibiting effect of the porous structure can be taken into account by increasing the activation energy of diffusion or by introducing a corresponding coefficient, if the former is impossible.
2. Use is made of the theory of SHF-heating of dielectrics [10], according to which the SHF-energy in a particle decreases following a parabola law, and of the corresponding solution for the dependence of the body temperature on the coordinate and the time.

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3. Traditional extraction (without SHF-energy) is regarded as a particular case of extraction in an SHF electromagnetic field.

4. The theory is compared with experiment, and upon establishing their adequacy, situations that cannot be implemented so far because of technical problems are considered.

As a basis, we take the expression for the coefficient of diffusion in a liquid [7]:

$$D = \frac{\delta^2}{6\tau_0} \exp\left(-\frac{W}{kT}\right) . \tag{1}$$

To save room, we do not present in complete form the dependence of the temperature on the coordinate and the time [10], but at rather large times (Fourier numbers) and at $X < 1$ ($X^4 << X^2$), it is possible to neglect small quantities without sacrifice of accuracy and, after elementary algebraic transformations, to obtain

$$T(x, \tau) = T_0 + (T_0 - 273)(AX^2 - B) , \tag{2}$$

$$A = 0.5\text{Po}_0 - 0.1bR^2\text{Po}_0 , \tag{3}$$

$$B = -2\text{FoPo}_0 + 0.3\text{Po}_0 + 0.6bR^2\text{Po}_0\text{Fo} + 0.39bR^2\text{Po}_0 . \tag{4}$$

The limiting depth of penetration of the SHF-energy $\lambda_{\text{lim}}$ is equal to the wavelength of the electromagnetic oscillations in the dielectric and can be calculated from the formula [10]

$$\lambda_{\text{lim}} = \frac{c}{f\sqrt{\varepsilon'}\,(1 \mp \tan\delta_{\text{d}})\exp(\pm\,\delta_{\text{d}})} . \tag{5}$$

Calculations carried out for media that have the smallest (sunflower oil, $\varepsilon' = 2.7$, $\varepsilon'' = 0.222$, $\tan\delta_{\text{d}} = \varepsilon''/\varepsilon' = 0.0882$) and largest (water, $\varepsilon' = 49.0$, $\varepsilon'' = 11.0$, $\tan\delta_{\text{d}} = \varepsilon''/\varepsilon' = 0.224$) [11] values of the dielectric characteristics at $f =$ 2450 MHz, $t_0 = 55°\text{C}$, $R_1 = 0.5\cdot10^{-3}$ m and $R_2 = 3\cdot10^{-3}$ m showed that for sunflower oil $bR_1^2 = 1.23\cdot10^{-4}$, $bR_2^2$ $1.48\cdot10^{-3}$ and for water $bR_1^2 = 0.8\cdot10^{-3}$, $bR_2^2 = 0.9\cdot10^{-2}$, respectively. The parameter $b$ decreases in direct proportion to $f$, so that at other resolved frequencies (915 and 433 MHz) the product $bR^2$ will be considerably smaller than at 2450 MHz. This allows one to neglect terms that contain $bR^2$ in formulas (3) and (4) and to obtain

$$A = 0.5\text{Po}_0 , \tag{6}$$

$$B = \text{Po}_0(0.3 - 2\text{Fo}) . \tag{7}$$

Thus, substituting Eq. (2) into Eq. (1), we have

$$D = h\exp\left(-\frac{W}{k\,[T_0 + (T_0 - 273)(AX^2 - B)]}\right) , \tag{8}$$

where $h = \delta^2/6\tau_0$.

The intrinsic time of penetration of the SHF-energy $R/v$, heat conduction $R^2/a_{\text{h}}$, and diffusion $R^2/D_{\text{sol}}$ increases in this series by several orders of magnitude in passage from one phenomenon to the next. For the SHF-energy this is obvious in view of the superhigh frequency of the oscillations at wavelengths of the order of $10^{-1}-10^{-2}$ m. Furthermore, for virtually all products of vegetable and animal origin $a_{\text{h}} = 10^{-7}$ m$^2$/sec in order of magnitude [12], whereas for solid bodies of vegetable origin $D_{\text{sol}} = 10^{-11}$ m$^2$/sec in order of magnitude [6]. In other words, the temperature field is formed much more rapidly than the concentration field, and then in a first approximation the problem of mass conduction can be considered at a certain fixed time of SHF-heating, i.e., in

Eq. (7) Fo = const and in Eq. (8) the dependence of $D$ only on $X$ remains (the other quantities $T_0$ and $W$ are considered to be prescribed).

The equation of diffusion with a coefficient $D$ depending on the coordinate has the form

$$\frac{\partial C(x,\tau)}{\partial \tau} = \frac{\partial}{\partial x}\left(D(x)\frac{\partial C(x,\tau)}{\partial x}\right). \tag{9}$$

In conformity with [13], we introduce the new variable

$$z = \int_0^X \frac{x\,dx}{D(x)}. \tag{10}$$

Then Eq. (9) takes the form

$$\frac{\partial C(z,\tau)}{\partial \tau} = \frac{1}{q(z)}\frac{\partial^2 C(z,\tau)}{\partial z^2}, \tag{11}$$

$$q(z) = z^s. \tag{12}$$

Integration of Eq. (10) within the limits from 0 to $r$ yields

$$z = \frac{r^2}{2h} + \frac{WR^2}{2hkT_0A}\ln\frac{a^2+r^2}{a^2} - \frac{W^{n+1}R^{2(n+1)}}{2hnk^{n+1}T_0^{n+1}A^{n+1}}\left(\frac{1}{(a^2+r^2)^n} - \frac{1}{a^{2n}}\right), \tag{13}$$

where

$$a = R\sqrt{\left(\frac{1-B}{A}\right)}, \quad n = 2, 3, 4, \ldots$$

Since, according to Eq. (10) or Eq. (13), $z$ is the time, then, in order to satisfy Eq. (11), it is required that $S = -1$. Physically, $z$ is the intrinsic time of diffusion inside a particle up to the point $r$. Actually, when $r \to 0$, $z \to 0$ (the center of the particle).

In Eq. (13) $W$ is the activation energy of a molecule in the free liquid. We will determine the activation energy of a molecule of the diffusing substance for the solid–liquid system ($W_{\text{sol}}$); naturally, $W_{\text{sol}} > W$. If we substitute $D_{\text{sol}}$ into Eq. (1) in place of $D$ and the quantity $W_{\text{sol}}$ in place of $W$, we can easily obtain

$$W_{\text{sol}} = -kT\ln\frac{D_{\text{sol}}}{h}. \tag{14}$$

Then instead of Eq. (13) we have

$$z = \frac{r^2}{2h} - \frac{TR^2\ln D_{\text{sol}}/h}{2hT_0A}\ln\frac{a^2+r^2}{a^2} - \frac{(-1)^{n+1}T^{n+1}R^{2(n+1)}\ln^{n+1}\frac{D_{\text{sol}}}{h}}{2hnT_0^{n+1}A^{n+1}}\left(\frac{1}{(a^2+r^2)^n} - \frac{1}{a^{2n}}\right). \tag{15}$$

In traditional extraction (without exposure to SHF-energy) $A \to 0$, and after resolving the indeterminacies we obtain

$$z = \frac{r^2}{2hd}. \tag{16}$$

In expression (16) the coefficient $d$ is introduced, since the intrinsic time naturally increases for the solid–liquid system. In Eq. (15), where the "load" is transferred to the activation energy, this fact is taken into account by the quantity $\ln\,(D_{\text{sol}}/h)$.

Averaging of Eq. (15) over the particle size gives

$$\bar{z} = \bar{z}_0 + \bar{z}_1 + \bar{z}_n\,, \tag{17}$$

for a sphere

$$\bar{z}_0 = 0.3\,\frac{R^2}{h}\,, \tag{18}$$

$$\bar{z}_1 = -\,\frac{T(\tau)\,R^2 \ln \dfrac{D_{\text{sol}}}{h}}{2hT_0A}\left(\ln\frac{a^2+R^2}{a^2} - \frac{2}{3} + \frac{2a^2}{R^2} - \frac{2a^3}{R^3}\arctan\frac{R}{a}\right), \tag{19}$$

$$\bar{z}_n = \frac{(-1)^{n+1}\,T^{n+1}(\tau)\,R^{2(n+1)}\ln^{n+1}\dfrac{D_{\text{sol}}}{h}}{2hn\,T_0^{n+1}\,A^{n+1}}\left[\frac{3}{2\,(n-1)\,R^2\,(a^2+R^2)^{n-1}} - \right.$$

$$\left. -\,\frac{3}{2\,(n-1)}\int\limits_0^R \frac{dr}{(a^2+r^2)^{n-1}} - \frac{1}{a^{2n}}\right]; \tag{20}$$

for an infinite plate

$$\bar{z}_0 = \frac{R^2}{6h}\,, \tag{21}$$

$$\bar{z}_1 = -\,\frac{T(\tau)\,R \ln \dfrac{D_{\text{sol}}}{h}}{2hT_0A}\left(R\ln\frac{a^2+R^2}{a^2} - 2R + 2a\arctan\frac{R}{a}\right), \tag{22}$$

$$\bar{z}_n = \frac{(-1)^{n+1}\,T^{n+1}(\tau)\,R^{2n+1}\ln^{n+1}\dfrac{D_{\text{sol}}}{h}}{4hn\,T_0^{n+1}\,A^{n+1}}\left[\frac{R}{(n-1)\,a^2\,(a^2+R^2)^{n-1}} + \right.$$

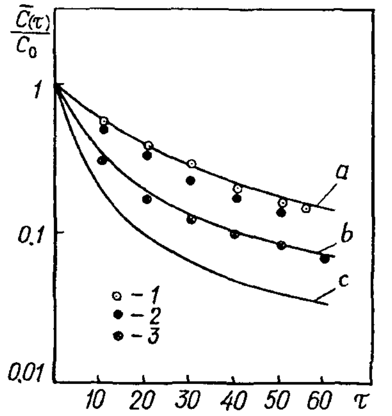
$$\left. +\,\frac{2n-3}{2\,(n-1)\,a^2}\int\limits_{-R}^{+R} \frac{dr}{(a^2+r^2)^{n-1}} - \frac{2R}{a^{2n}}\right]. \tag{23}$$

Work [13] contains the solution of Eq. (11) under the boundary conditions

$$C\,(z, 0) = 0\,, \tag{24}$$

$$C\,(0, \tau) = \varphi\,(\tau)\,. \tag{25}$$

Condition (24) corresponds to absorption (adsorption) of a substance by a solid body; this is the inverse problem in comparison with extraction [14], and it can easily be transformed into the problem of extraction [13].



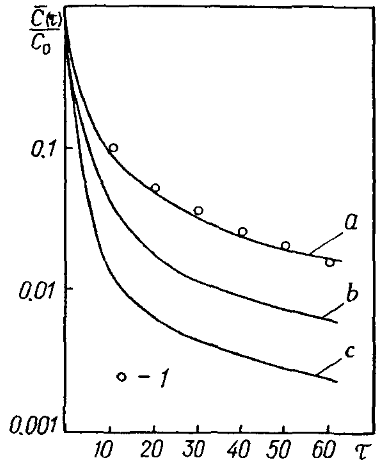


Fig. 1. Relation $\bar{C}(\tau)/C_0 = f(\tau)$ for particles of a prepressed sunflower oil cake with $R = 3$ mm in extraction without SHF-heating (conventional process) [a) calculation; 1, 2) first and second experiments under the same conditions] and with SHF-heating [b) calculation at $\text{Po}_0 = 5.8$; 3) experiment at $\text{Po}_0 = 5.8$; c) calculation at $\text{Po}_0 = 10$]. $\tau$, min.

Fig. 2. Relation $\bar{C}(\tau)/C_0 = f(\tau)$ for a raw soybean petal with $R = 0.275$ mm in extraction without SHF-heating (conventional process) [a) calculation; 1) experiment] and with SHF-heating [b) calculation at $\text{Po}_0 = 2.44 \cdot 10^{-2}$; c) calculation at $\text{Po}_0 = 10$].

In condition (25) the function $\varphi(\tau)$ can be determined from the following considerations. Physically, the limit $z \to 0$ means an infinitely large rate of diffusion, i.e., particles are saturated infinitely rapidly by the solution from the surrounding medium, and $\varphi(\tau) = C_0$ is the initial concentration. Taking this into account and regarding the relaxation time in [13] as the time of impregnation $m\tau$ before the extraction proper, we obtain

$$\frac{C(\bar{z}, \tau)}{C_0} = 1 - \frac{(1 - m)}{m} \exp\left(-\frac{\bar{z}}{m\tau}\right), \quad 0 \le m \le 1, \quad 0 \le \tau \le \infty. \tag{26}$$

When $m \to 0$, the impregnation is "instantaneous," which is quite justifiable in extraction by low-viscosity solvents, and from Eq. (26) we have

$$\frac{C(\bar{z}, \tau)}{C_0} = 1 - \exp\left(-\frac{\bar{z}}{\tau}\right). \tag{27}$$

When $m \to 1$, $C(\bar{z},\tau) \to C_0$, i.e., all the time is expended on impregnation (there is no extraction as yet).

Equation (27) corresponds to the physical picture that when $\tau \to 0$, $C(\bar{z},0) \to C_0$ and when $\tau \to \infty$, $C(\bar{z},\infty) \to 0$.

In our works [4, 6] we presented experimental data on extraction by means of extraction gasoline: extraction of particles of a prepressed sunflower oil cake with $R = 3$ mm in an SHF-field at a frequency of 2450 MHz and without it, and of a raw soybean petal without an SHF-field. The experiment on extraction of a soybean petal is distinctive in that it was carried out without blocking the surfaces of petals by each other. The blocking in a layer can reach 50%, but generally it can hardly be determined, because petals are arranged randomly in a layer. Each petal (a plate) was extracted separately (as one particle), which, strictly speaking, is necessary in order to use the equations for $z$. For the particles of the prepressed oil cake, which have a spherical shape (in a first approximation), blocking of the outer surface is not so topical.

As seen from Figs. 1 and 2, the theory is confirmed satisfactorily by experiment. In an SHF-field the process is accelerated considerably (severalfold)) to attain the same residue of the substance.

The use of an SHF electromagnetic field brings extraction, in particular of vegetable oils, to a qualitatively new level.

In Fig. 1 curve b corresponds to an experment on extraction in an SHF-field at $\mathrm{Po}_0 = 5.8$. At this value of $\mathrm{Po}_0$ the power of the heat source on the particle surface was determined by the formula $w_0 = \mathrm{Po}\lambda_h t_0/R^2$. The quantity $\lambda_h = 0.09$ W/(m·K) was calculated by the formulas from [15], and then $w_0 = 3.19 \cdot 10^6$ W/m$^3$. At the density of the oil cake saturated with the solvent $\rho_{o.c} = 0.89$ ton/m$^3$, which in turn was determined by the additivity law at the density of the perfectly dry substance of the oil cake 1.38 ton/m$^3$ [15] and of hexane 0.6 ton/m$^3$ [6], we obtain $w_0 = 3.59 \cdot 10^6$ W/ton. Then for an industrial extractor with a capacity of 350 ton/day the established (net) power with respect to extractable material (oil cake) ($4.05 \cdot 10^{-3}$ ton/sec) will be equal to $3.59 \cdot 10^6 \cdot 4.05 \cdot 10^{-3} = 14.5$ kW. At $\mathrm{Po}_0 = 10$ (curve c in Fig. 1), we correspondingly obtain 25.1 kW. It is clear that the growth of the net power is directly proportional to the number $\mathrm{Po}_0$, while the decrease in the residue of the substance has a complex dependence determined by Eqs. (17)-(20), (27) for a sphere and Eqs. (17), (21)-(23), (27) for an infinite plate.

In Fig. 2 curve b corresponds to intensification of the process at $\mathrm{Po}_0 = 2.44 \cdot 10^{-2}$. The quantity $\lambda_h = 0.079$ W/(m·K) [12], and then $w_0 = 1.4 \cdot 10^6$ W/m$^3$. For $\rho_{pet} = 0.7$ ton/m$^3$, we obtain $w_0 = 2.0 \cdot 10^6$ W/ton. At the same capacity of the extractor the net power is equal to 8.1 kW.

An attempt at a sharp increase in the rate of extraction (curve c in Fig. 2) (the residue of the substance lies in the region of thousandths of a unit) at the number $\mathrm{Po}_0 = 10$ (which means an increase in this number by three orders of magnitude) leads to a net power of 3320 kW, which obviously is unacceptable. This large power is naturally associated with evaporation of the solvent in intense boiling of it over the entire volume of the apparatus, which is not expedient for extraction.

An approach to this problem should be based on a comparison of the decrease in the extracted-substance residue in the solid phase or the increase in the extractor capacity with the increase in energy consumption. The relationships obtained in this work allow one to do this. For example, a comparison of curves a and b in Fig. 2 shows that for soybean seeds with a percentage of oil of 20% an oil residue in a grist of 0.5% can be attained in a period of 15 min in SHF-heating, while in traditional extraction it can be attained only in a period of 40 min, i.e., the capacity of the extractor can be increased by a factor of 2.7. This is very important at a net power of SHF-heating of 8.1 kW.

Naturally, the total energy consumption in SHF-heating depends on the efficiency of the installation. In the field of extraction so far now there are no corresponding technical solutions; however, installations with a power of several tens of kilowatts have already been created for other purposes.

## NOTATION

$D$, $D_{sol}$, coefficient of diffusion in the liquid and in the solid, respectively; $\delta$, elementary translation of a molecule; $\tau_0$, period of oscillations of a molecule about the equilibrium state; $W$, $W_{sol}$, activation energy of diffusion in the liquid and in the solid, respectively; $k$, Boltzmann constant; $T_0$, $T$, initial and current temperature, respectively; $X = r/R = x/R$, dimensionless coordinate; $x$, $r$, current coordinates; $R$, radius of the sphere or half of the plate thickness; $R_1$, $R_2$, first and second numerical values of $R$; $\mathrm{Po}_0 = w_0 R^2/\lambda_h t_0$, initial value of the Pomerantsev number; $t_0$, initial temperature, °C; $w_0$, power of the heat source on the particle surface; $\lambda_h$, thermal conductivity; $b = 1/\lambda_{lim}^2$, coefficient of the parabola; $\lambda_{lim}$, limiting depth of penetration of the SHF-energy into the particle; $\mathrm{Fo} = a_h \tau/R^2$, thermal Fourier number; $a_h$, thermal diffusivity; $\tau$, time; $c$, speed of light in a vacuum; $f$, frequency of the oscillations; $\varepsilon'$, $\varepsilon''$, real and imaginary parts of the complex dielectric permittivity; $\delta_d$, angle of dielectric losses; $C$, concentration; $\overline{C}$, concentration averaged over the particle size; $C_0$, initial concentration; $z$, intrinsic time of diffusion; $v$, rate of propagation of electromagnetic oscillations in the dielectric; $m$, fraction of time for impregnation; $\rho_{o.c}$, density of the oil cake; $\rho_{pet}$, density of the petal; $d = D_{sol}/D$.

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