Investigation of particle size distribution of grinded amber by electropulse discharges in a liquid medium

The article presents the study results of electropulse grinding of amber in aqueous and alcoholic media at different amounts of supplied energy. Description of the electropulse grinding laboratory installation, the mechanism of the destruction process of amber particles and methods of statistical processing of experimental data are given. It was established that alcohol medium has a greater impact on the efficiency of crushing than water. Thus, under the same conditions of energy supply, in the aqueous medium the weighted average particle size of amber was \(601.6 \pm 688.9\, \mu m\), and in an alcohol medium – \(368.0 \pm 269.6\, \mu m\). In an aqueous medium, the particle size decreased to \(1/13.6\) of raw sample, and in an alcoholic medium to \(1/22.3\) of raw sample compared to the initial size of raw amber. We found that in the aqueous medium the ratio of large to small fractions is mainly the same with the coefficient of alignment of particles with a size of 1.09. In an alcoholic medium, this ratio significantly differs, with the coefficient of alignment of amber particles of a size of 1.67 with the amount of supplied energy of 125 kJ.

1. Introduction

About 20% of extracted stones are suitable for the production of amber jewelry, and the rest is melted and pressed for further jewelry needs [1]. The degree of grinding of amber determines the technological quality of the process. Among the known advanced methods of grinding substances of stone structure, such as ore, to obtain minerals and metals, one uses electropulse discharges. Efficacy of the...
method is also confirmed by its efficiency of grinding diamond-bearing ores in their final stages of processing [2]. This method can presumably be effective for grinding such a solid polymer as amber. Thus, the experiments of [3] confirmed high efficiency of grinding materials of different strength. Grinding of a material occurs due to the action of the shock wave front, which is generated by a high-voltage electric discharge in the liquid. This effect was studied by scientists in [4]. The example of crystalline silicon processing in the chemical industry [5] also confirmed the efficiency of electropulse grinding in a liquid without dust, as the presence of dust creates some inconvenience.

The work [6] investigated grinding of amber by electropulse discharge, but the grinding was carried out only in an aqueous medium. The researchers also investigated the influence of the liquid medium on the generation of the shock wave amplitude during oil well treatment [7] and the improvement of the porosity of the coal structure [8] during electropulse treatment in NaCl solutions of different concentrations. Then, the influence of the medium in amber grinding should be investigated. However, in the literature there is no information about the influence of alcohol on the efficiency of amber grinding.

In this context, it should be noted that amber contains a significant amount of succinic acid, which can be extracted for a further use in food, pharmaceutical, cosmetic and other industries. The formed amber meal is sent to high-temperature processing to obtain jewelry.

The use of electropulse discharges as a method of preliminary preparation of raw materials for extraction intensifies the process of extraction of succinic acid due to the formation of a new contact area between the phases and the increase in raw material porosity.

The purpose of the work is to establish the influence of water and alcohol medium on the efficiency of grinding substandard amber under different conditions of energy supply of electropulse discharge.

2. Materials and methods

2.1. Preparation of amber for grinding

Substandard amber stones mined in Volyn region, Ukraine, were selected for research. The amber that was subjected to grinding was selected by passing through a sieve with holes with a diameter of 10.4 mm and the rest with a hole diameter of 6 mm. The average particle size of raw amber was 8.2 mm.

2.2. Grinding of raw amber

Electropulse processing was performed using a set of electrical equipment consisting of a universal power supply that provides a stabilized DC voltage; a high-resistance charging resistance consisting of resistors; a thyristor; capacitors
with a nominal capacitance of 0.25 and 1.0 µF. In all experiments, the connecting wires did not change. The thyristor was controlled by a generator of rectangular (shifted) pulses in the manual start mode (except for the processing procedure).

The experiments were performed in special electric discharge chambers of cylindrical shape (Fig. 1). The cylindrical chamber (Fig. 1a) has the metal body 1 with a diameter of 250 mm, the grounded electrode 4 mounted in the center of the bottom at a distance of 45 mm from the high-voltage electrode 3. The body of the horizontal semi-cylindrical chamber (Fig. 1b) is made of stainless steel with a diameter of 160 mm, at the bottom of which is mounted the grounded electrode 4 having a gap of 30 mm between it and the high-voltage electrode 3.

![Scheme of electric discharge chambers](image)

Fig. 1. Scheme of electric discharge chambers: 1 – chamber body; 2 – lid; 3 – high voltage electrode; 4 – grounded electrode

The experimental installations were equipped with a pointed electrode-tip 100 mm above the surface of the chamber in the form of steel reinforcement with a diameter of 12 mm, placed in a tube of vacuum rubber with an outer diameter of 34 mm. The other protruding end had a welded metal guard and a terminal strip for connecting the cable tip from the discharge switchboard. The radius of the electrode tip was half the diameter of the armature – 6 mm.

In the cylindrical chamber (Fig. 1a), the raw material was processed with 200 pulses when the capacitance of the connected capacitor was 1.0 µF, i.e., the total amount of energy used was 250 kJ. The installation (Fig. 1b) was equipped with a capacitor with a capacitance of 0.25 µF, in which 400 discharges were generated with a total energy of 125 kJ.

The charging voltage was the same in all cases, equal to 50 kV, but different capacitors were connected to different chambers, with a capacitance of 1.0 µF in the cylindrical and 0.25 µF in the semi-cylindrical one, so the discharge power in each chamber was different.

The processing was performed in aqueous and alcoholic medium with an ethanol concentration of 96% at a mass ratio of liquid to solid phase of 15:1, respectively. Alcohol is chosen as a medium because it is a rational solvent for further extraction of amber and simplifies the use of the obtained extract [9].
In addition, water and alcohol have different physical properties, which allows for a clearer analysis of the results of amber grinding.

Amber sample of 51 g was loaded in the chamber body 1, then the chamber was filled with 760 g of liquid and closed with the lid 2. The energy accumulated in the capacitor bank was converted into the energy of the plasma of the discharge channel, which was formed between the two electrodes, 3 and 4. Then, the energy of the plasma was transferred to the liquid that surrounded it, after which a part of the energy of the liquid was expended on the work of grinding the material.

After electropulse grinding of amber, the chamber was unloaded, the obtained suspension was filtered, and this was followed by drying to equilibrium moisture. Amber dried samples were subjected to sieve analysis to determine the particle size distribution of the mixture.

2.3. Analysis of particle size in grinding products

Particle size analysis was performed by sieving the ground amber through sieves with the following hole sizes: 2500, 2000, 1500, 1000, 800, 670, 560, 450, 390, 300, 250, 200, 160, 132, 112, 90, 71, 56, 45 \( \mu m \). The sieve holes size was selected so that the modulus of the sieves set [10] was equal, \( \Delta \approx 1.21 \). As a result of sieving, the yield of each fraction was determined, on the basis of which differential particle distribution curves were constructed. Integral distribution curves were calculated by summing the yield values of the obtained fractions.

2.4. Mathematical generalization of experimental data

Mathematical description of the integral curves of amber grinding was carried out using the software Origin 8.6.

Amber grinding efficiency was described by the following indicators: the degree of grinding \( i \), the weighted average particle size of grinded amber \( d_a \), the total coefficient of fineness of grinding \( k_1 \), the coefficient of fineness of grinding of small fraction \( k_2 \), the coefficient of fineness of grinding of large fraction \( k_3 \), the coefficient of alignment of amber particles by size \( \beta \) [11].

Amber grinding efficiency is determined by calculating the degree of grinding \( i \), which is the ratio of the average particle size before grinding \( d_b \) to the average particle size of the product after grinding \( d_a \):

\[
i = \frac{d_b}{d_a},
\]

where \( d_b, d_a \) – respectively, the weighted average particle size before grinding and after grinding, \( \mu m \).

The weighted average particle size \( d_a \) of the whole grinded product was determined by the formula [12]:

\[
d_a = \frac{m_1d_1 + m_2d_2 + \ldots + m_id_i}{m_1 + m_2 + \ldots + m_i},
\]
where, \( m_1, m_2, m_i \) – amount of the product in each faction, \( \% \); \( d_1, d_2, d_i \) – respectively, the average particle size in each fraction, which was defined as the half-sum of the holes sizes of the upper and lower sieves, \( \mu \text{m} \).

The degree of dispersion of particle size of the grinded product is the standard deviation \( S \), which was calculated with the formula [12]:

\[
S = \pm \sqrt{\frac{\sum_{i=1}^{n} m_i x_i^2}{\sum_{i=1}^{n} m_i} - \left(\frac{\sum_{i=1}^{n} m_i x_i}{\sum_{i=1}^{n} m_i}\right)^2}.
\]  

(3)

If the value of the standard deviation is low, the scattering of the particle size is small and the homogeneity of the mixture, with respect to particle size, is better. The standard deviation characterizes unevenness of grinding of the product, i.e., over-grinding in small fractions and under-grinding in large [12].

The average particle size of the products, i.e., the product of which is 50\%, is calculated by interpolation using a Lagrange polynomial [13] according to the formula:

\[
d_{av} = \sum_{i=0}^{n} d_i \prod_{j=0}^{n} \frac{x - x_j}{x_i - x_j},
\]

(4)

where \( d_{av} \) – the average particle size at which the total product of the products is 50\%, \( \mu \text{m} \); \( d_i \) – the particle size of the fractions of the grinded product, \( \mu \text{m} \); \( x \) – the total product of the products which is equal to \( x = 50\% \); \( x_i, x_j \) – the value of the product of the products within which the interpolation is performed, \( \% \).

To find the average particle size of the mixture one took the range of four points, within which 50\% the total product was located.

The large fraction included the products of amber grinding the particle size of which was greater than 50\% of the average particle size in the total product.

The grinding efficiency can be evaluated by the total grinding fineness coefficient \( k_1 \), which shows the ratio of large to small particles in the crushed product [11]. If the value of the total grinding coefficient \( k_1 \) is close to one, then there are more small particles in the test product, and if the coefficient \( k_1 \) is close to zero, then there are more large particles.

The total coefficient of fineness of grinding of amber \( k_1 \), was calculated according to the formula [11]:

\[
k_1 = \frac{\int_{d_{\min}}^{d_{\max}} y_i \, dx}{(d_{\max} - d_{\min}) y_{\max}},
\]

(5)
where \( d_{\text{min}}, d_{\text{max}} \) – respectively the smallest and largest value of the particle size of the amber fractions, \( \mu m \); \( y_{\text{max}} \) – the largest total yield of amber, \( y_{\text{max}} = 100\% \); \( y_i \) – fraction yield, \( \% \).

The coefficient of fineness of grinding of amber of small fraction \( k_2 \) is calculated by the formula [11]:

\[
k_2 = \frac{d_{av}}{d_{i}} \int_{d_1}^{d_{av}} y_i \, dx \left( \frac{d_{av} - d_1}{0.5y_{\text{max}}} \right),
\]

where \( d_{av} \) – the average particle size of the fraction, which contains 50\% of the total product, is calculated by the formula (4), \( \mu m \).

The coefficient of fineness of grinding of amber of large fraction \( k_3 \) is calculated by the formula [11]:

\[
k_3 = \frac{d_{max}}{d_{av}} \int_{d_{av}}^{d_{max}} y_i \, dx \left( \frac{d_{max} - d_{av}}{0.5y_{\text{max}}} \right) - 1.
\]

Integration was performed using a trapezoidal formula.

To assess the alignment of the crushed product by size one used the coefficient of alignment of the particle size distribution \( K_p \). If the distribution has a higher coefficient of evenness of the particle size \( K_p \), the product is more aligned in size, i.e., in the mixture there are more particles of the same size. As the coefficient \( K_p \) increases, the integral curve of the particle size distribution is closer to a straight line [12]. The coefficient of alignment of amber particles by the size \( K_p \), was calculated according to the formula [11]:

\[
K_p = \frac{k_3}{k_2}.
\]

3. Results and discussion

Particle size analysis allows us to determine the yield of fractions of different sizes, which make up the whole test sample. As a result of grinding the amber under three different grinding conditions, the differential yield of each fraction was obtained, which is shown in Fig. 2. Differential curves show variations in the number of grains in individual factions.

The data of Fig. 2 show that the grinding products have an uneven distribution of particles in the total mass of the product. In the fractions with the highest yield, the curve shows maxima, and in the case of absence of particles – the curve falls
to zero. The formation of uneven distribution of amber particles can be explained by the inhomogeneity of the amber solid and the unevenness of the forces applied to the particles during grinding. It is difficult to conduct the analysis of differential characteristics of crushing, because the differential curves are more sensitive to changes in particle size distribution than the integral curves.

![Graph of particle size distribution](image)

Fig. 2. Differential distribution curves of grinded amber \(q \geq 0.95; n = 3\): 1 – aqueous medium, 250 kJ; 2 – alcohol medium, 125 kJ; 3 – alcohol medium, 250 kJ

The data in Fig. 2 indicate that the differential curves are polymodal. The differential distribution curve of amber particles obtained in an aqueous medium has 8 maxima; the differential curve of distribution of particles of amber grinded in an alcoholic medium, with the supplied grinding energy of 125 kJ, has 6 maxima, and accordingly, the distribution curve of particles in alcoholic medium with the supplied grinding energy of 250 kJ has 7 maxima. These results illustrate the discrete distribution of amber particles in the grinded mixture, regardless of the grinding medium. Discrete distribution of amber particles may indicate discrete distribution of the applied energy during grinding. The discrete set of weak points of the amber structure before grinding with different strength is also a cause for discrete particle size range of the grinded amber [14].

Comparing integral curves compared to the differential ones is the best way to graphically represent the data for particle size analysis. According to the type of integral curves and the degree of their concavity, one can draw conclusions about the prevalence of large or small particles in the sample.

The processing of amber by electropulse method in water gave a larger range of particle sizes than the processing in alcohol, under the same conditions (curves 1 and 2).
Integral curves are well described by the Langmuir equation:

\[ Y = \frac{abx^{1-c}}{1 + bx^{1-c}}, \]  

(9)

where \( Y \) – integral yield of the fraction; \( a, b, c \) – experimental coefficients of the equation; \( x \) – the particle size of the fraction, \( \mu m \).

Correlation coefficients \( R \) are close to one, which indicates a close relationship between the studied features.

Table 1 shows the calculation results of statistical parameters of grinded amber mixtures under different grinding conditions.

<table>
<thead>
<tr>
<th>Indicator</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Weighted average of particle size, ( d_a, \mu m )</td>
<td>601.6 368.0 343.2</td>
</tr>
<tr>
<td>Standard deviation, ( S )</td>
<td>688.9 269.6 231.6</td>
</tr>
<tr>
<td>Degree of grinding, ( i )</td>
<td>13.6 22.3 23.9</td>
</tr>
<tr>
<td>Total coefficient of fineness of grinding, ( k_1 )</td>
<td>0.46 0.46 0.46</td>
</tr>
<tr>
<td>Coefficient of fineness of grinding small fraction, ( k_2 )</td>
<td>0.47 0.38 0.3</td>
</tr>
<tr>
<td>Coefficient of fineness of grinding large fraction, ( k_3 )</td>
<td>0.53 0.62 0.7</td>
</tr>
<tr>
<td>Coefficient of alignment of amber particles by size, ( K_p )</td>
<td>1.34 1.65 2.31</td>
</tr>
</tbody>
</table>

Based on the data in Table 1, the following conclusions can be made: grinding of amber by electropulse method in water with energy supply of 250 kJ reduced the weighted average particle size of amber to 601.6 \( \mu m \) with the range of particle scatter relative to the weighted average of 688.9 \( \mu m \). Under the same conditions, grinding of amber in alcohol reduced the particle size to 368.0 \( \mu m \) with the range of particle scatter around the weighted average of 269.6 \( \mu m \). This indicates that the alcohol medium, in which the grinding was carried out, affected the efficiency of grinding making it greater than in the aqueous medium, which led to a decrease in amber particle size. The range of particle scatter relative to the weighted average value also decreased. Smaller values of the weighted average of amber particle size obtained by grinding in the alcoholic medium can be explained by the lower value of the dielectric constant of alcohol. The dielectric constant of alcohol is 25.3, and the dielectric constant of water is 80 [15]. Thus, due to higher amplitudes of discharge pressures formed in a liquid with lower dielectric constant, we obtained such a result of amber grinding, because the main factor deciding on the destruction of the material is the amplitude of pressure pulse, not the pulse energy.

The data from Table 1 also indicate the correlation between the standard deviation of the particles \( S \) and the coefficient of alignment of the particles in size.
As the standard deviation of the particle scatter decreases, the coefficient of particle alignment in size $K_p$ increases. This indicates a greater homogeneity in the size of the particles.

Comparing the weighted average of particle sizes of grinded amber (Table 1) obtained in the alcoholic medium, one can see that the increase in supplied energy from 125 kJ to 250 kJ leads to a slight increase in particle size. This effect can be explained by the fact that the number of electric pulses is a more influential factor for grinding than the total amount of the supplied energy. In the case of using 400 discharges with less energy (312.5 J), a better grinding result is achieved than when using 200 discharges with a four times higher energy (1250 J).

The weighted average of amber particle size does not correspond to the average particle size of 50% of the total product of the products, and this is reflected by the asymmetry of the integral curves, shown in Fig. 3. Using the Lagrange interpolation polynomial [13], we calculated the average particle size in 50% of the total product of, which was 307 $\mu$m for amber grinded in aqueous medium, 271 $\mu$m for amber grinded in an alcohol medium with an energy supply of 250 kJ, and 269 $\mu$m for amber grinded in an alcoholic medium with an energy supply of 125 kJ. Perhaps the difference between the weighted average and the average particle size in 50% of the total product can be explained by the peculiarities of the formation of the grinded products.

![Fig. 3. Integral distribution curves of grinded amber ($q \geq 0.95$; $n = 3$): 1 – aqueous medium, 250 kJ; 2 – alcohol medium, 125 kJ; 3 – alcohol medium, 250 kJ](image)

The degree of grinding indicates that in an aqueous medium the particle size of amber decreased to $1/13.6$ of raw sample (250 kJ), and in an alcohol medium
to 1/22.3 (250 kJ) and 1/23.9 (125 kJ) of raw sample, compared with raw amber before grinding.

The total fineness coefficient of amber grinding correlates with the weighted average particle size and the degree of grinding. With a decrease in the weighted average particle size of amber, the total fineness coefficient of grinding increases, which can be clearly seen in the data given in Table 1.

Coefficients of fineness of grinding of small and large fraction give idea of a ratio of small to large fractions in the total weight of a product. From the data of Table 1 one can see that, when grinding amber in an aqueous medium, the ratio of large to small fractions is preferably equal, and the coefficient of alignment of the particles in size is 1.34. A similar result is observed for the amber which is grinded in the alcoholic medium with the supplied energy of 250 kJ, where the coefficient of alignment of the particles in size is 1.65. A completely different result is observed for amber which is grinded in the alcoholic medium with an energy supply of 125 kJ. The number of small fractions is lower than that of large fractions, which is evidenced by the fineness coefficient of grinding of the small fraction equal to 0.3, against the fineness coefficient of grinding of the large fraction, which is equal to 0.7. Such a significantly different ratio of small to large fractions gave an increased coefficient of alignment of particles in size, which amounted to 2.31.

From the analysis of grinding coefficients one can conclude that, in an alcoholic medium, the supply of a greater energy leads to a slight increase in particle size, but the ratio of large to small fractions in the total mixture is preferably the same. On the contrary, reducing the amount of supplied energy leads to a slight decrease in the weighted average particle size of the mixture, but the large fractions increase more than the small ones.

Therefore, it should be noted that the fronts of high pressure waves created during the electropulse discharge in the liquid, which move across the liquid, are able to grind solid materials.

Firstly, the so-called pre-discharge stage is considered, during which a discharge channel is formed, which closes the inter-electrode gap, then – the phenomena occurring during the release of capacitor energy in the discharge channel, and finally – the pulsation of the gas bubble after discharge. In the area surrounding the discharge channel, a high impulse pressure develops, which manifests itself in the form of explosive mechanical impact on the medium. Distribution and attenuation of such shock waves depends on the hydrostatic pressure of the liquid and the energy of the discharge, which accordingly affects the performance of the mechanical action of the wave during grinding.

After the formation of the channel, a strong discharge current, reaching tens and hundreds of kiloamperes, heats the plasma at the initial stage of the discharge to a temperature of about $10^4$ K. During the flow of current in the discharge process, the plasma temperature changes slightly and decreases after the discharge.

The heat produced by plasma causes an increase in the pressure in the channel. Under the action of high pressure, the channel expands. The pressure in the channel
during the discharge process passes through the maximum: at the initial stage of the discharge, the pressure in the channel increases, despite the increase in the channel’s volume, and decreases only near the end of the discharge.

4. Conclusions

The results of the study showed that substandard amber is better ground by electropulse in an alcoholic medium than in aqueous medium with the amount of supplied energy of 125 and 250 kJ. The results of the study confirmed the effect of alcohol on the grinding efficiency of substandard amber. Due to the fact that higher amplitudes of discharge pressure are formed in a liquid with lower dielectric constant, we obtained this result of amber grinding, because the main factor in the destruction of the material is the amplitude of pressure, not its pulse energy.

As a result of electropulse grinding of amber, the best indicators characterizing the efficiency of the process were obtained in an alcoholic medium using discharges with an energy of 312.5 J compared to the discharges of 1250 J. In this case, one should pay attention to the factors that may influence this result. Perhaps the number and frequency of summed pulses had additional effects on the formation of shock-wave front, contributing to grinding of amber.

In the future, it is necessary to investigate the effect of concentration and amount of alcohol on the grinding efficiency at different amounts of energy supplied.

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