

# Optimization of Ultrasonic Powder Coatings on the Surface of Treated Materials

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**Abstract:** By generalizing the theory of contact of solids for the case of triple contact of working bodies, the powder particle, the treated surface and the problem of establishing an optimal value for the amplitude of ultrasonic exposure, at which the coating process proceeds most efficiently, were solved. The relevance of such a solution is directly determined by the need to obtain high-quality coating. Analytical expressions of the desired values for the physical parameters of the materials of the objects participating in the working process were obtained. Possible limitations regarding the most important characteristics of the installation were analyzed. Approximate evaluation of the optimal number of working bodies for the resonant chamber was derived. The basic time of the process duration was estimated for the total treatment of the chosen surface by working bodies.

**Keywords:** coating; ultrasonic oscillations; resonant chamber; roughness; deformation; hardness; working bodies; ball treatment; elasticity



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## 1. Introduction

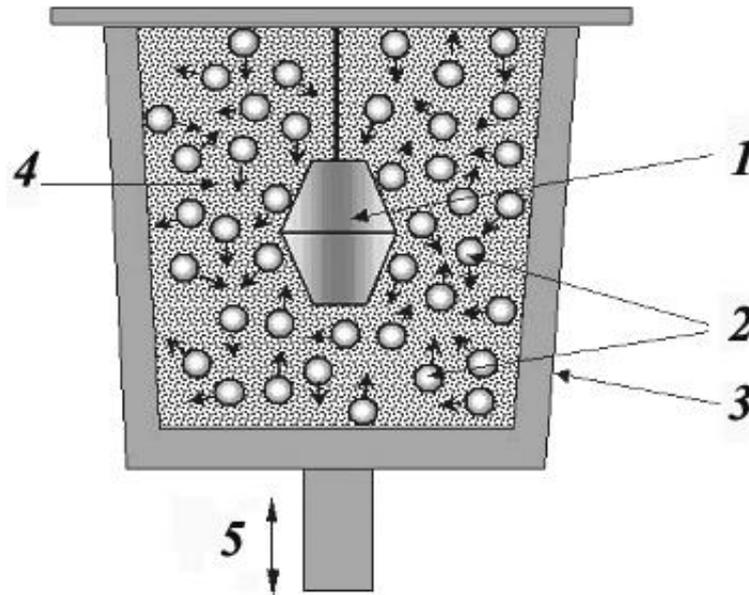
Current coating methods, such as galvanic, spraying, and chemical and physical deposition, have a number of limitations regarding the types of materials used, which is mainly due to the incompatibility of coatings and matrix materials. For example, it is extremely difficult to apply a refractory coating on the surface of a material having a lower melting point or on ceramic material. Other disadvantages are related to the need to maintain a certain temperature or to control the pressure and composition of the gas atmosphere when applying coatings, which requires expensive laser, arc or vacuum equipment [1–3].

The method of applying nanocoatings on the surface of a product using free working bodies accelerated by the ultrasonic vibrations of the walls of the resonance chamber is an example of a close combination of physical and chemical processes, and the quality of the final product depends entirely on the optimal course used. The fact is that the strength of a coating is determined by the adhesive forces of intermolecular attraction, which are enhanced as a result of the occurrence of a chemical bond between the adhesive and the substrate, and the necessary depth of introducing nanoparticles into the substrate is dictated by the physics of the interaction of the working bodies, the coating particles and the surface of the processed product. The choice of optimal conditions for the expected implementation of each of the parties to this process is a necessary step in obtaining the maximum efficiency of the technology under consideration.

The considered method possesses a number of attractive features that may make it useful for many practical applications. First, this method can provide not only a rapid formation of fine-grained structure in the coated surface layers similar to what shot peening treatment does [4–7] but also coating and armoring. Second, this method allows the armoring of metal surfaces with particles of other metal or ceramic materials, which can be capable of significantly changing the characteristics of the metal surfaces. Third, the coating can be carried out under atmospheric pressure and room temperature by using simple and cheap ultrasonic-based devices, depending on the operation conditions. Fourth, this method may work with different kinds of surfaces that can be treated.

## 2. Materials and Methods

The essence of the considered method of applying metallic, intermetallic or ceramic coatings to the surface of a treated material, using the energy of ultrasonic vibrations, is easiest to explain using Figure 1. Metal or ceramic balls (diameter from 0.1 to 3 mm) and metal or ceramic powder are placed inside a resonant chamber, the walls of which vibrate at the frequencies of the ultrasonic range.



**Figure 1.** Coating on the surface of a product: 1—treated surface; 2—working bodies in the form of steel balls; 3—resonance chamber; 4—coating material in powder form and 5—connection to the transducer.

The energy of the vibrations of the chamber walls is transmitted to the balls, which begin to randomly fly and collide with each other, as well as with the surface of the processed product placed inside the resonance chamber. As a result of intensive bombardment of this surface, part of the particles fixed on it or captured by the flying balls is driven into the surface layer. Therefore, a coating or layer of composite material having a nano- or microstructure is formed on the surface. The main feature of this approach is that it has no restrictions in the choice of coating materials and substrates, and by controlling the composition of the gas phase during coating, it is possible to form on the surface of the metal not only a metallic or intermetallic coating, but also a coating or a composite layer consisting of oxides, nitrides or carbides.

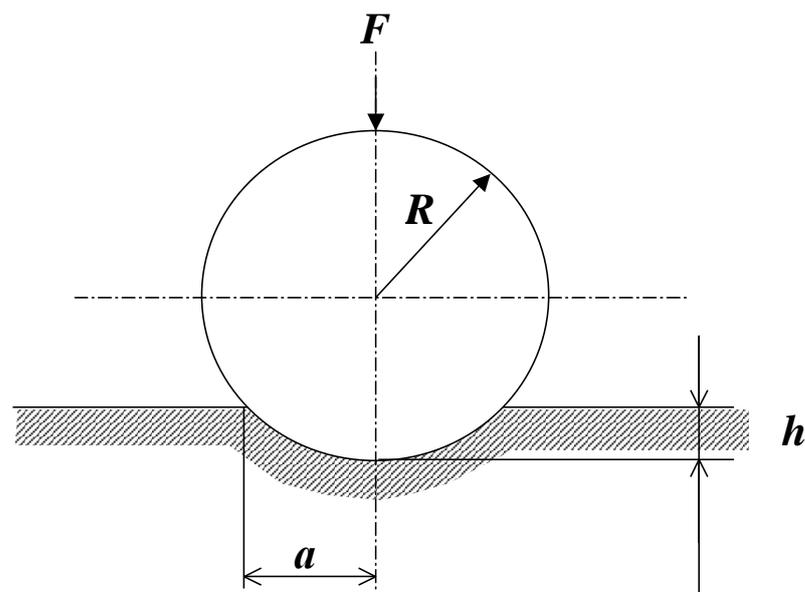
In this work, the method of obtaining the basic relationships among the main characteristics of the process of applying powder coating using ultrasound is based, first of all, on a correct understanding of the physicochemical essence of the interactions taking place. This makes it possible to obtain simple formulas for estimating the values of the most important parameters and to develop practical schemes for the implementation of the technology in question, which is capable of ensuring maximum efficiency when obtaining the finished products.

## 3. Results

This work is devoted to solving the problem of establishing the optimal values of the parameters of the nanocoating process. The urgent need for this work is dictated by the need to obtain high-quality coating. A detailed visual representation of the process under consideration, presented in Figure 1, explains the principle of applying a layer of powder material to the treated surface using the method of impact via working bodies accelerated by ultrasound. However, in order to carry out specific calculations, more complete details

of all interactions are necessary to take into account a number of important assumptions justified by the physical essence of the described processes. This scheme is shown in Figure 2. First of all, this figure more or less realistically represents a powder in the form of solid nanoparticles (balls) of mass  $m$  and radius  $R$  from the coating material, which are in a free suspended state in the resonance chamber, or are placed near its walls or held on them by glue. The process of introducing these balls into the treated surface can occur when the ball in contact with it experiences an impact from the working body flying from the side of the wall of the resonance chamber. In this case, the force that this working body produces can be represented as the action of some external force  $F$  on the ball. To obtain specific expressions describing the parameters of the task, we will proceed from the law of conservation of energy in a system consisting of a working body moving in the form of a solid ball with a mass  $M$  and a radius of  $R_0$ , a ball of the coating material standing motionless against the wall, and the wall itself, into which the ball of the coating material must be pressed to a depth  $h \equiv h(t)$  depending on time  $t$ . The optimal value of this depth  $h_{\text{opt}}$  is determined by the requirements of the strength of the applied coating, its thickness, esthetic features, etc. Its value can be expressed in terms of the dimensionless coefficient  $\kappa$ , specified by the above requirements, with the help of the following formula:

$$h_{\text{opt}} = \kappa R \quad (1)$$



**Figure 2.** Scheme depicting coating on a treated surface by pressing the impact force of a working body into it, which is a powder in the form of a hard ball from the coating material:  $R$ —ball radius of coating material;  $a$ —residual trace radius of implementation;  $h$ —depth of residual trace of implementation; and  $F$ —the force used to insert the ball.

As a result of pressing the ball into the surface by the force  $F$ , the potential energy  $U$  of the contacting bodies arises as follows:

$$\partial_h U = F \quad (2)$$

In the case of pressing the ball into a flat surface, the solution of the contact problem of these bodies obtained in the work [4] leads to the following result:

$$U = \frac{2}{5D} \cdot \sqrt{h^5 \cdot R} \quad (3)$$

where  $D = \frac{3}{4} \cdot \left\{ \frac{1-\sigma^2}{E'} + \frac{1-\sigma^2}{E} \right\}$ ,  $\sigma$  is the ball's Poisson ratio,  $E$  is the modulus of elasticity of the ball, and the stroke indicates the corresponding parameters characterizing the surface of the wall.

The complete picture of the indentation process is characterized by the fact that before the start of the contact, the energy of the system in question is equal to the kinetic energy  $T$  of the ball, which is the working body:

$$T = M\omega^2 \xi_{m0}^2 / 2. \tag{4}$$

Here,  $\omega$  is the frequency, and  $\xi_{m0}$  is the amplitude of ultrasonic exposure on the walls of the resonant chamber that accelerates the working bodies.

The total energy in the system is equal to the sum of the kinetic energy of the moving bodies, which can be written as  $(m + M) (\partial_t h)^2 / 2$ , and the potential energy (3). By virtue of the law of conservation of energy using (3) and (4), one can obtain the following:

$$M\omega^2 \xi_{m0}^2 = (M + m) (\partial_t h)^2 + \frac{4}{5D} \cdot \sqrt{h^5 R}. \tag{5}$$

At the moment when  $\partial_t h$  turns to zero, there is obviously a maximum convergence of the ball to the surface, when there should be  $h = h_{opt}$ . Using (1), we can rewrite Equation (5) at this point in time in the following form:

$$M\omega^2 \xi_{opt}^2 = \frac{4}{5D} \cdot \kappa^{5/2} R^3. \tag{6}$$

Here,  $\xi_{opt}$  is the optimal value of the amplitude of ultrasonic exposure, at which the powder particles of the coating material are pressed to the required depth into the treated surface.

In previous work [4], the following relationship has been derived between the microhardness  $H$  of the treated surface and its physical characteristics:

$$H = \frac{1}{2\pi D} \tag{7}$$

which is obtained in approximation when the properties of the surface and the ball pressed into it are not very different. Using (7), which is based on (6), it is possible to obtain for  $\xi_{opt}$  the formula describing the dependence of the optimal amplitude of ultrasonic exposure on the parameters of the material of the treated surface and the properties of the nanopowder used:

$$\xi_{opt} = \frac{2\sqrt{2\pi H} \kappa^{5/4} R^{3/2}}{\sqrt{5M} \cdot \omega} \tag{8}$$

From Equation (8), it follows that with an increase in the mass of the working body, the optimal value of the amplitude of ultrasonic exposure decreases. This is due to the fact that up to a certain limit of the value of this mass, the working body is accelerated by the walls of the resonant chamber to a speed of  $\omega \xi_{opt}$ , regardless of the magnitude of this mass. Therefore, a working body with a higher mass acquires a higher value of kinetic energy. Additionally, since the value of this energy is fixed to drive a particle of nanopowder to a given depth, then with an increase in mass, a decrease in the speed  $\omega \xi_{opt}$  is required, and, therefore, a lower amplitude value  $\xi_{opt}$ .

On the other hand, Equation (8) shows that the optimal value  $\xi_{opt}$  of the amplitude increases with increasing microhardness of the treated surface. It corresponds to simple physical considerations that the harder the surface, the more difficult it is to embed nanoparticles into it, and a more vigorous external influence is required. It is also clear that the greater the radius of nanoparticles, the greater the amplitude of ultrasound is needed to introduce them to a set depth. As the frequency increases, the speed increases, with which

the working body flies away from the wall of the resonance chamber. Therefore, with increasing frequency, the value of the optimal amplitude decreases.

The optimal number of working bodies is the result of a compromise between two contradictory conditions. On the one hand, their number cannot be too small since this significantly increases the processing time. On the other hand, a large number of working bodies leads to the fact that, when colliding with each other, they interfere with the normal course of the coating process, which also increases the processing time and worsens the quality of the final product.

To estimate this parameter, consider the motion of a working body in the case when there are  $N$  pieces in the space with a characteristic size  $l$  between the walls of the resonance chamber and the treated surface. With the velocity  $v(t)$ , one can use the following equation [4,8,9]:

$$d_t v = -\Omega \cdot v(t) \tag{9}$$

where  $\Omega = N R_0^2 v(t)/l^3 \equiv q v(t)$  is the frequency of ball collisions.

The solution of Equation (9) gives the following expression for the velocity  $v_s$  of the working body at the moment of contact with the nanoparticle of the embedded coating:

$$v_s = \omega \xi_{m0} \exp(-q l) \tag{10}$$

From (10), it can be seen that the essence of the compromise formulated above is the following approximation:

$$q l \leq 1 \tag{11}$$

Hence, the estimate for the optimal value of the number of  $N_{opt}$  working bodies for a resonance chamber, the free volume in which, for the span of the working bodies, is approximately equal to the value of  $l^3$ , is followed from the evident inequality:

$$N_{opt} \leq l^2/R_0^2 \tag{12}$$

The optimal surface treatment time with the working bodies is achieved in accordance with the condition when the entire surface area is covered with an appropriate amount of material. It means that at a given value of the treatment area  $S$ , it is necessary to sink into the surface at least  $N_0$  nanoparticles, the total cross-sectional area of which is  $S$  at the level of depth into the treated surface. It corresponds to the case when all nanoparticles lie next to each other without intersecting, which, of course, is unlikely. Therefore, in reality, the number of embedded nanoparticles should be taken twice as much. In fact, this number corresponds to the number of impacts of the working bodies necessary for the qualitative coating of the treated surface with a material in the form of nanopowder.

For the depth of implementation that is approximately equal to the radius of the nanoparticle  $R$ , we have the following equation:

$$\pi N_0 R^2 = 2S, \text{ that is } N_0 = 2S/\pi R^2 \tag{13}$$

These nanoparticles are driven by  $n_0$  working bodies flying at a distance  $l$  to the surface of the processed product from the walls of the resonant chamber, which accelerate the working bodies. The distance that a working body will travel between two consecutive collisions is obviously equal to the value of  $2l$ . Moving with a speed of about the value of  $\omega \xi_{m0}$ , the working body will spend the process time  $t_s$  equal to the following:

$$t_s = \frac{2l}{\omega \xi_{m0}} \tag{14}$$

During this time,  $2n_0$  nanoparticles will be embedded in the surface. Additionally, it will take the time  $t_{\text{opt}}$  to drive  $N_0$  nanoparticles:

$$t_{\text{opt}} = \frac{N_0}{2n_0} t_s = \frac{2Sl}{\pi\omega\xi_m R^2 n_0} \quad (15)$$

Equation (15) offers an expression for the optimal processing time of a given surface with a selected number of working bodies. It can be seen that this time is inversely proportional to the square of the radius of the nanoparticles and the number of working bodies. In the case when the optimal parameter values are selected, that is  $n_0 = N_{\text{opt}}$ , we have the following, with the help of (12) and (8):

$$t_{\text{opt}} \approx \frac{S}{\pi R l} \frac{R_0^2}{R^2} \sqrt{\frac{5M}{2\pi H R}} = \frac{l}{2R} \sqrt{\frac{5M}{2\pi H R}} \quad (16)$$

At its core, Equation (16) gives a rough estimate of the time required for the ultrasonic treatment of the chosen surface, thus providing useful guidelines for practical application. The actual duration of the resonance chamber is determined, of course, by means of processing quality control, when the process is stopped after achieving the desired result.

#### 4. Discussion

The connections between the main parameters of the process of applying powder coating using ultrasound, which have been derived in the present work, have a satisfactory agreement with the results of the experiments performed in [4,7], where all the values of the required characteristics necessary for assessments are given. Meanwhile, the development of this area of research leads to the identification of new effects that require their scientific description and evaluation analysis. More recent studies using this method have revealed [10,11] that it can obtain ultra-fine-grained and nanocrystalline structures at the surface layers of the treated samples within a relatively short time. Further development of research in this area has been accompanied by a significant expansion of areas using the studied approach. For example, in Ref. [12], the improvement in low-cycle fatigue behavior of some alloys was discovered after ultrasonic shot peening at room temperature. Fatigue crack initiation in the untreated samples was found on the surface, whereas it was in the subsurface in the ultrasonic shot peening-treated samples. In the review [13], the research area that covers the development and/or control of surface properties in order to improve the strength of metallic materials, such as controlling the fatigue fracture toughness, fretting damage, delayed fracture, stress-corrosion cracking, and hydrogen embrittlement, was examined. Another possibility was studied in Ref. [14]. The model was built in this work to study the peening and pitting of metallic materials resulting from non-spherical cavitation-bubble collapse near the materials. Bubble reentrant jet impact and shock wave emission from the jet impact and from the collapse of the remaining bubble ring can induce permanent micro-deformation, pitting, and residual stresses, which modify the roughness of the material and harden it through pre-stressing. These effects were investigated for different bubble–material standoff distances.

Thus, the widespread use of ultrasonic shock methods makes the simple formulas obtained in this work especially relevant, which allow us to not only make estimates of the necessary characteristics of the process, but also to use the inter connection of the parameters to develop practical technological schemes.

## 5. Conclusions

From the results of the above calculations and estimates, the following conclusions can be derived:

1. The obtained ratios show that the size of powder particles lies in the range from a few units to ten nanometers in the case when the frequency of ultrasound is in the operating range from 18 to 25 kHz, and the amplitude of the displacements of the walls of the resonance chamber does not exceed 15  $\mu\text{m}$ . This means that for the operating parameters commonly used in standard ultrasonic equipment, the optimal size of coating particles is in the nanoscale range of values.
2. The coating technology under consideration allows one to work with a surface of a very complex shape. It is necessary to take into account the fact that for high-quality processing of recesses, it is necessary to use working bodies, the size of which is less than the radius of curvature of the surface of this part of the product.
3. The above expressions for the optimal values of the number of working bodies and the time of surface treatment give only approximate guidelines in the establishment of the technological process. Their exact values are determined in practice from the conditions used to obtain the required quality of coverage.

In general, the resonant chamber can be in motion over the treated surface or the treated surface can shift relative to the chamber, making it possible to proceed to a wider area of treatment. Thus, the considered method makes it appropriate for practical use in many different ways and practical schemes can be realized with effective technologies.

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## References

1. Suryanarayana, C. Mechanical alloying and milling. *Prog. Mater. Sci.* **2001**, *46*, 1–184. [[CrossRef](#)]
2. Lü, L.; Lai, M.O. *Mechanical Alloying*; Kluwer Academic Publishers: Boston, MA, USA, 1998; p. 276.
3. Grigorieva, T.F.; Barinova, A.P.; Lyakhov, N.Z. Mechano-synthesis of nanocomposites. *J. Nanoparticle Res.* **2003**, *5*, 439–453. [[CrossRef](#)]
4. Abramov, V.O.; Abramov, O.V.; Sommer, F.; Gradov, O.M.; Smirnov, O.M. Surface hardening of metals by ultrasonically accelerated small metal balls. *Ultrasonics* **1998**, *36*, 1013–1019. [[CrossRef](#)]
5. Abramov, O.V. *Ultrasound in Liquid and Solid Metals*; CRC Press, Inc.: Boca Raton, FL, USA, 1993; p. 493.
6. Abramov, O.V. *High-Intensity Ultrasonics: Theory and Industrial Applications*; OPA: Singapore, 1998; p. 692.
7. Komarov, S.V.; Son, S.H.; Hayashi, N.; Kasai, E.; Kaloshkin, S.D.; Abramov, O.V. Development of a novel method for mechanical plating using ultrasonic vibrations. *Surf. Coat. Technol.* **2007**, *201*, 6999–7006. [[CrossRef](#)]
8. Lighthill, J. Acoustic Streaming. *J. Sound Vib.* **1978**, *61*, 391–418. [[CrossRef](#)]
9. Bird, R.B.; Stewart, W.E.; Lightfoot, E.N. *Transport Phenomena*; John Wiley & Sons, Inc.: New York, NY, USA, 2002; p. 895.
10. Wu, X.; Tao, N.; Hong, Y.; Xu, B.; Lu, J.; Lu, K. An investigation of surface nanocrystallization mechanism in Fe induced by surface mechanical attrition treatment. *Acta Mater.* **2002**, *50*, 4603–4616.
11. Tao, N.R.; Sui, M.L.; Lu, J.; Lu, K. Surface nanocrystallization of iron induced by ultrasonic shot peening. *Nanostruct. Mater.* **1999**, *11*, 433–440. [[CrossRef](#)]
12. Kumar, P.; Mahobia, G.S.; Singh, V.; Chattopadhyay, K. LCF life improvement of biomedical Ti-13Nb-13Zr alloy through surface nanostructuring. *Mater. Res. Express* **2019**, *6*, 125413. [[CrossRef](#)]

13. Soyama, H. Surface mechanics design of metallic materials on mechanical surface treatments. *Mech. Eng. Rev.* **2015**, *2*, 14-00192. [[CrossRef](#)]
14. Chahine, G.L.; Kapahi, A.; Hsiao, C.T.; Choi, J.K. Coupling bubble and material dynamics to model cavitation peening and pitting. *J. Fluid Sci. Technol.* **2016**, *11*, JFST0023. [[CrossRef](#)]

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