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INDUSTRIAL APPLICATIONS OF HIGH-INTENSITY ULTRASOUND

Key words: ultrasonic equipment, molten metal, enhanced oil recovery, waste water, antibacterial nanomaterials.

Abstract: In the last two decades, an increasing interest of academy and industry in the development of enabling technologies for process intensification has been paid. The main bottle neck for scientists and engineers to apply non-conventional technologies at industrial levels, such as high-intensity ultrasound, is the scaling up. Power ultrasound is proving to be a front-runner and offers a wide range of profitable applications. Although most examples reported by patents and scientific literature are not yet an industrial reality, few applications are recognised as the best available techniques for big scale production. Eloquent examples are reviewed in this article.

Przemysłowe zastosowania ultradźwięków o wysokiej intensywności

Słowa kluczowe: urządzenia ultradźwiękowe, stopiony metal, ulepszony odzysk oleju, ścieki, antybakteryjne nanomateriały.

Streszczenie: W ciągu ostatnich dwóch dekad wzrosło zainteresowanie środowiska naukowego i przemysłowego rozwojem technologii wspomagających intensyfikację procesów. Główną przeszkodą dla naukowców i inżynierów do stosowania niekonwencjonalnych technologii na poziomie przemysłowym, takich jak ultradźwięki o wysokiej intensywności, jest zwiększanie skali. Ultradźwięki oferują szeroką gamę dochodowych zastosowań i okazują się być dobrym rozwiązaniem. Pomimo że większość zastosowań zgłaszanych w formie patentów lub przedstawianych w literaturze naukowej nie istnieje w rzeczywistości przemysłowej, niektóre z nich uznawane są za najlepsze dostępne techniki dla produkcji na dużą skalę. Konkretne przykłady zostały omówione w artykule.

Introduction

The course of any physicochemical process in a given system is determined by temperature, pressure, their changing rates, and the speed of relative movement of particular parts of the system. The kinetics of any chemical conversion can be affected by varying these parameters in the search for optimal conditions to enhance the reaction rate. This also occurs in materials manufacturing science and biological processes affecting the metabolism of micro- and macro-organisms.

Innovative technologies, such as lasers, plasma, electromagnetic fields, and high-energy particle fluxes, appreciably widen the range of means to control physicochemical processes. Compared to natural processes and standard technologies, they may substantially affect heat and mass transfer and common parameters (temperature, pressure, speed of convective flows in liquids and structural imperfection movement in solids). The use of the latest scientific advances in the processes of physicochemical transformations could appreciably contribute to the development of efficient technologies of industrial interest. In this regard, one of the most emerging technologies is high-intensity ultrasound, because it is environmentally friendly and effective in a wide range of processes [1]. Power ultrasound is already exploited by the industry and several other uses have a real chance to be applied on a large scale.

1. Technical issues in the design of industrial ultrasonic equipment

The design of *ad hoc* tailored high-intensity ultrasound reactors and a rational scaling up of lab scale facilities is not a trivial task. The construction of new generators with the development of complex oscillatory systems and the design of suitable geometries for an efficient transmission of ultrasonic waves are now a matter of extensive research. The extent of this development largely determines the efficiency of high-intensity ultrasound applications in industrial processes in view of the electrical energy required. The transducers, power supply systems, radiators, and various systems for the measurement and control of acoustic parameters are part of such installations. Typically ultrasonic units include at least five blocks, namely, an electric ultrasonic generator, an electroacoustic transducer, a waveguide system, a technological load, and a control system.

Magnetostrictive and piezoelectric effects are the two phenomena exploited by electroacoustical transducers to generate ultrasonic waves. Depending on the type of the electroacoustic transducer, magnetic or electric fields induce elastic strains in electromechanically active materials. Magnetostriction is typical for ferromagnetics and ferrites. Magnetostriction is a change in the linear and volumetric dimensions of metallic bars made of specific alloy as a result of the interaction of an external magnetic field. Although only moderately efficient, these transducers are extremely robust and reliable enabling harsh working conditions. Currently, piezoceramic transducers are the most widely used in ultrasonic technologies. The piezoelectric effect consists in strain imposed on the transducer material by external electric fields. When the piezoceramic transducer operates under optimal conditions, its electroacoustical efficiency ranges from 90 to 98%.

Aiming to achieve a high level of efficiency, the ultrasonic units have to address the following two conditions [2, 3]:

1. The system should operate at or in the vicinity of its electric and mechanical resonance (the frequency of the electric current generated by the ultrasonic generator should match the frequency of the natural resonance of the electroacoustic transducer and the waveguide system).
2. There must be effective transmission of the vibrations through the contact area between the transducer and the waveguide system.

Experimental search of the optimal waveguide dimensions is complicated and time consuming. Nevertheless, aiming to simplify and speed up the procedure to get the best waveguide parameters, besides experimental work, a computational modelling of these parameters can be performed. Modelling of resonance frequencies and waveguides geometry can be calculated by means of the finite-element method (i.e. by the commercially available COMSOL Multiphysics) [4]. This is a powerful interactive environment for modelling and calculating data that is based on partial differential equations (PDE) by the finite-element method. This software package expands the standard models which use one differential equation (application mode) and Multiphysics® Models for calculating inter-related physical phenomena. This method of calculation enables one to find the optimum shape and structure while avoiding the construction of test parts. This significantly accelerates the design of ultrasound devices and the selection of optimal processing modes.

2. Ultrasonic Action on Molten and Solidifying Metal

Ultrasonic action on molten and solidifying metal is one of the most technologically advanced ultrasonic technologies in the industry. Vibrational treatment leads to solid non-metallic and gaseous impurities removal from liquid metal and to structure refinement of the ingot in shape casting. The effect of ultrasound on the alloy structure can be summarized as follows [2, 3, 5–7]:

- Reduction of the grain size;
- Formation of equiaxed grains;
- Variation of phase distribution in terms of relative amounts, structure refinement, and mutual geometry;
- Improvement of material homogeneity; and,
- Uniformity improvement of the distribution of non-metallic inclusions.

Thus, ultrasonic treatment of molten and solidifying metals improves their mechanical properties of strength and plasticity. Ultrasonic treatment was industrially used for such metals as Al and Mg. Figure 1 shows a specific device for ultrasonic treatment of molten aluminium.

The insertion of high-power ultrasonic vibrations into molten metal leads to cavitation and acoustic streaming. Cavitation involves the formation, growth, pulsating and collapsing of tiny discontinuities or bubbles in the melted material. The discontinuities result from the tensile stress generated by the sound wave during the tensioning phase. The compression rate of this unsteady state can be so high that their collapse generates a hydraulic shock wave. The bubble is initiated and moves in liquid simultaneously with many other bubbles spaced by less than one wavelength and forming a cavitation region. The average bubble collapse pressures of ~10^4 MPa vary for low-melting metals within one order of magnitude. Besides cavitation, the propagation of high intensity ultrasonic wave involves the initiation of steady state acoustic streaming in the liquid.

Fig. 1. Ultrasonic treatment of molten aluminium

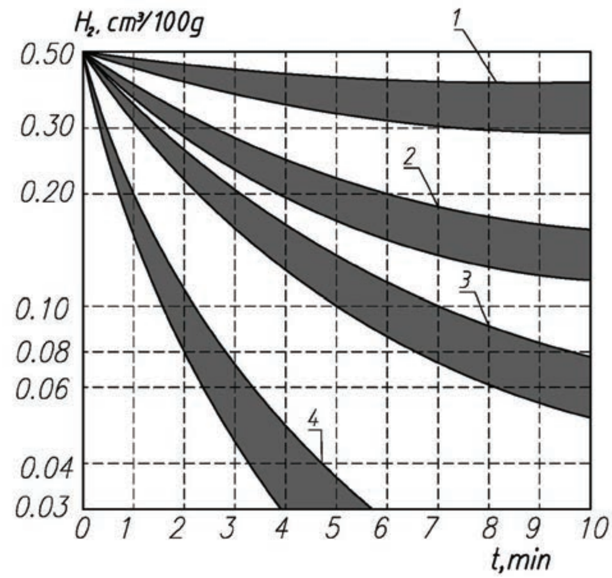


Fig. 2. Degassing kinetics for grade A19 aluminium alloy: 1 – zinc chloride process, 2 – vacuum degassing, 3 – ultrasonic degassing, 4 – ultrasonic + vacuum degassing

Ultrasonic refining of metal melts is intended for effective removal of solid non-metallic and gaseous impurities from liquid metal. This method might be recommended for the refining of light metal alloys (Al, Mg), where material defects, due to solid and gaseous impurities, must be minimised. As depicted in Figure 2, the efficiency of ultrasonic degassing is higher in comparison with melt treatment by the additions of chlorine salts and evacuation [3].

The concentration of hydrogen in aluminium alloys is reduced by ultrasound by the factor 2–3 in comparison to the untreated metal. For example, the concentration of hydrogen in the melt of grade A19 alloy is normally 0.45–0.75 cm^3/100 g of metal at a temperature 720°C. Ultrasonic treatment reduces the gas concentration level to 0.1 cm^3/100 g. Similar results were obtained during the degassing of other aluminium and magnesium alloys (grade AMg6, A14, MA8, etc.). Ultrasonic refining improves mechanical properties (characteristics of strength and plasticity, fracture toughness, and fatigue properties) of as-cast and as-worked product. It enables one to increase the characteristics of the strength and plasticity of alloys by 15–30%.

The introduction of vibrations into solidifying metal results in refining of its structures and an increase of the chemical homogeneity of the produced ingot or casting and also provides a non-dendritic structure. In solidifying metal (melt and two-phase liquid-solid zone), cavitation and acoustic streaming might produce crystal nucleation and the dispersion of growing crystals.

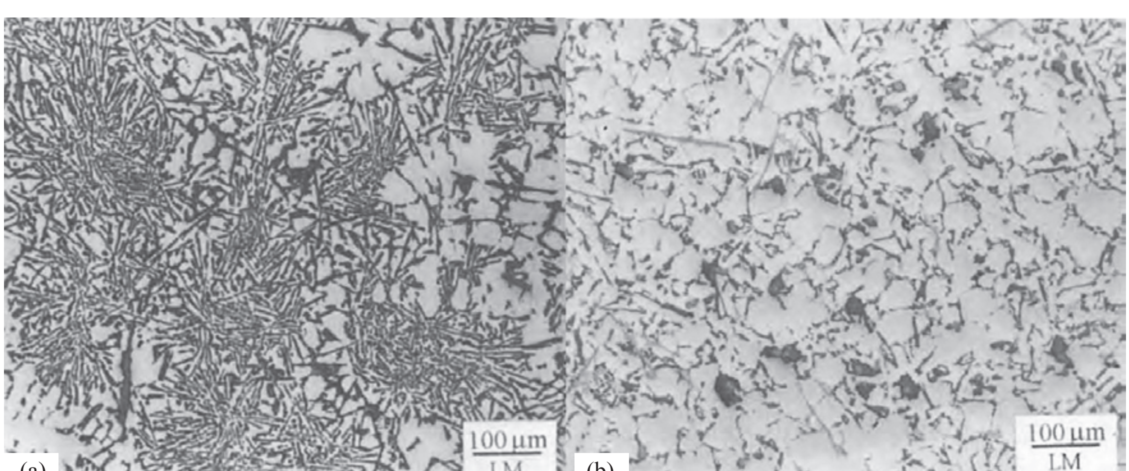


Fig. 3. Microstructure of AlSi13 alloys solidified conventionally: (a) and with ultrasonic treatment (b)

For example, in hypereutectic AlSi13 specimens prepared with the aid of convectional casting, the primary silicon crystals as hexagonal plates joined together at the centre into star-shaped particles, as they appear in cross section. The ultrasonic treatment may refine the silicon crystals providing a homogeneous cross-section. Most of the silicon plates were disconnected and broken during the ultrasonic treatment, forming spheroid crystals. This may be observed in Figure 3. Typically, ultrasonic treatment results in a plasticity increase of a factor of 1.3 to 1.6.

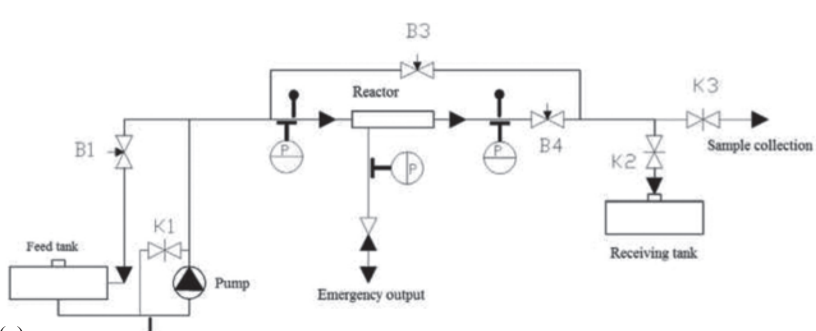
3. Ultrasonic technology for enhanced oil recovery and transportation of highly viscous oil

Another example of an industrial scale application of high power ultrasound is to enhance oil recovery. Generally, the efficiency of oil recovery from wells is less than 40% with a strong effort in the search of new enabling technologies [8]. Existing processes for enhanced oil recovery (EOR) are high-energy and labour consuming and often environmentally unfriendly [9, 10]. Thus, there is great interest in the development of more efficient technologies. Over the last few years, the use of physical EOR techniques have been reported, especially those based on ultrasonic treatment [11–15]. We have developed an ultrasound-assisted method that operates in the wellbore perforation zone with the simultaneous creation of a zone of lower pressure. The methodology is particularly useful for older wells which are in the later stages of reduced yields.

Ultrasonic treatment is one of the most promising alternative methods for affecting a fluid both under well conditions and on the surface. Laboratory and field tests have shown that acoustical oscillations initiate a variety of chemical and physical processes in oil bearing formations. Under well conditions, ultrasonic treatment can lead to such effects as increase of fluid penetration into capillaries due to the sonocapillary effect, an increase of fluid mobility, and the detachment of paraffinic and other deposits from the rock [14]. Under well and surface conditions, ultrasound can lead to viscosity reduction [16, 17]. In high viscosity oil conglomerates, the molecules are bonded to each other by intermolecular forces. The goal of ultrasonic treatment is to destroy these bonds and to bring the properties of the oil closer to what they would have been if no conglomerates were present. To study the potential of ultrasonic treatment, we have carried out the following:

- Studies of the effect of ultrasonic – hydrodynamic treatment (UHT) and chemical agents on the rheological characteristics of oil; and,
- Studies of the effect of ultrasonic treatment on the rheological properties and heavy oil production under well conditions.

Fluid from one field of Samara region (Russia) was used to investigate the effect of UHT and chemical agents on the rheological characteristics of extremely viscous oil. The fluid had the following characteristics: the density was 0.953 g/m^3, the effective viscosity at 20^{o}C was 1014 mPa*s, and the freezing point was 17°C. The fluid contained 64.05% oil, 28.6% resins, and 6.1% asphaltenes. To study the effect of UHT on crude oil and petroleum products, an experimental setup was assembled. The setup made it possible to vary the pressure at the inlet of the reactor. The maximum inlet pressure was 50 MPa, and the maximum capacity of the setup was 1200 L/h. Figure 4a shows the scheme of the UHT experimental setup. The setup consisted of a pump, a working section (reactor), a tank for the untreated oil, a receiving tank, an electric heater, an emergency discharge tank, control and stop valves, and instrumentation (pressure gages, a compound pressure and vacuum gage and thermocouples). A hydrodynamic emitter is inserted in the working chamber, as depicted in Fig. 4b. The operation of the emitter is based on the generation of oscillations in a liquid media, when the jet from the nozzle interacts with a barrier of a certain shape and size. The perturbations caused by the obstacle affect the jet base, causing auto-oscillations. In the experimental setup, we used an annular slotted nozzle, which was formed by two conical surfaces. The barrier had the



(a)

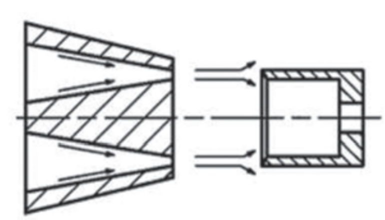

(b)

Fig. 4. a) Schematic of the experimental setup for ultrasonic – hydrodynamic oil treatment; b) Schematic of the hydrodynamic emitter in the setup for UHT

shape of a hollow cylinder, dissected along the elements. Thus, the barrier consisted of cantilever plates, arranged circumferentially. The frequencies of the oscillations, caused by that emitter, were in the range of 15–35 kHz. The electric heater in the setup was used to maintain constant temperature of the treated oil.

Experiments have shown that the use of UHT is a promising acoustic method for decreasing the viscosity of oil after its recovery. Such treatment makes it possible to reduce oil viscosity by more than 30%. In addition, treatment in the designed setup allows a chemical agent to be effectively introduced into the oil. The introduction of a chemical agent during UHT lead to a synergistic effect and cause a further reduction in viscosity, compared to the viscosity of oil after UHT only.

Aiming to study the effect of ultrasound on the rheological properties of oil under well conditions, an ultrasonic tool was placed into the perforation zone of the well. The tool was attached to the tubing and was powered with a suitable cable, attached to the outer surface of the tubing using cable bands. An ultrasonic generator was installed near the wellhead on the surface to power the downhole tool. The output power of the ultrasonic generator was 9 kW and 5 kW at the ultrasonic downhole tool. The scheme of the whole equipment set up is shown in Fig. 5.

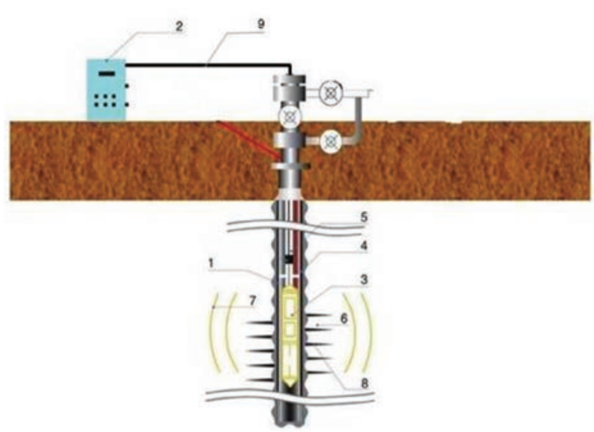


Fig. 5. Arrangement of ultrasonic equipment in the well: 1 – anchor, 2 – ultrasonic generator, 3 – downhole tool (102 mm), 4 – casing, 5 – tubing, 6 – reservoir, 7 – area of acoustic impact, 8 – perforation zone, 9 – cable

4. Experimental set up in oil well

Experiments were carried out in the producing well No. 4620 at the Demkinskoe oil field (Russia). The well had the following characteristics: 168 mm production casing, C1bb formation, perforated interval depth 1309.3–1312 m, fluid production before treatment was 1.82 m^3/day, oil production before treatment was 1.51 tons/day, bottom hole pressure (on average during the last month before treatment) was 25.6 atm, temperature was 23^{o}C, water cut was 10.3%, formation pressure was 49 atm, and the production coefficient was 0.071. The outer diameter of the downhole tool was 102 mm, and its length was 700 mm. The downhole tool contained three annular magnetostrictive transducers that generated an ultrasonic field at a frequency of 19 kHz.

Ultrasonic treatment by a downhole tool located directly in the perforation zone leads to a decrease of the viscosity of oil and simultaneously to an increase in oil production. Both effects were demonstrated during field experiments: a viscosity reduction by 16% 4 h after treatment was observed, and oil production was simultaneously increase by 26.5%. As a result, the velocity of oil rise from the well increased. Consequently, the oil, which is supplied to the pipeline near the well, was 10^{o}C hotter and the aggregated viscosity change was based on two factors: the change of the rheological properties of oil due to ultrasonic treatment and the higher temperature. Besides the increase production rate in the well, ultrasonic treatment performed inside the well can facilitate the pipeline transportation of heavy oil.

For the treatment of horizontal wells a sonochemical approach [12] should be used when a combination of acoustical and chemical treatment is used. In this case, ultrasonic treatment not only contributes to the cleaning of the perforation zone and increasing the mobility of oil, but also to the penetration of the reagent into the formation and acceleration of the chemical reaction in the porous media of the formation. In order to carry out effective treatment of horizontal wells, specially designed equipment has been developed. Any technology of EOR for horizontal wells should be based upon the following guidelines:

- The formation intervals, i.e. those which need treatment, should be based on geophysical studies.
- In order to achieve sinergistic effects, the reagents need to be injected directly into the zone of acoustic treatment.
- The treatment should be selective in that only the problematic zones should be treated, and this decreases the overall treatment time.

Taking into account the mentioned above, ground and downhole equipment for sonochemical EOR of horizontal wells has been developed [12]. The ground equipment includes an upgraded ultrasonic generator involving a unit for processing the information obtained regarding pressure and temperature in the borehole, and this information is obtained from a downhole tool. The ultrasonic generator is matched to the ultrasonic downhole tool and easily adapts to changes in the technological load by controlling the voltage and the current which goes to the downhole tool. The ultrasonic generator can work in a pulse mode and can modulate the power. The operating frequency is 15–30 kHz, and the output power is 10 kW. During the operation of the generator, the following parameters can be monitored on the display: voltage, current, work/pause, and frequency.

The downhole equipment includes a sonotrode, a system for injection of chemicals, and a probe for acquiring geophysical data (temperature, pressure, flow). In order to use this equipment in horizontal wells, the equipment complex must include a special signal cores to control the parameters of the process. The cable is an armoured polymeric tube, and the copper cores are nested within it. The cross section of the cable is represented in Fig. 6. The diameter of the hydraulic channel is 15 mm. Power up to 5 kW can be delivered through the cable. The cable can be also used for moving the ultrasonic downhole tool through the horizontal area of the well during the treatment. Apart from the injection of reagents directly to the zone of the acoustical treatment, the cable can also be used to wash and clean the horizontal area of the well with technological fluid prior to and after the treatment. The armour surrounding the cable protects it from external damage.

Fig. 6. Cross-section of the cable with hydraulic channel and power and signal cores

It has the required breaking strength and torsional stiffness to be wound onto a drum on a geophysical truck. Photographs of the wireline truck constructed specially for sonochemical treatment of horizontal wells in the transport (a) and working (b) position are presented in Fig. 7.

Fig. 7. Photograph of the wireline truck for sonochemical treatment of horizontal wells in working position

During the treatment of wells, it is necessary to continuously process the data in order to choose the appropriate treatment modes and to adjust them during the operation. Complex geophysical downhole tools, which are used during the treatment, measure the following parameters: pressure, temperature, flow of the fluid, and magnetic location of the couplings. A special cable with a hydraulic channel for the injection of technological fluids was developed. The cable includes electrical cores to power the ultrasonic equipment (1.5 mm diameter) and the geophysical probe (0.4 mm diameter).

To select the optimal design of the sonotrode, various waveguide systems were designed, modelled, manufactured, and tested. The best results were achieved when sonotrodes emitted from the sidewall were used. The operating frequency of the sonotrode used for sonochemical treatments of wells was 20 kHz.

Our preliminary work on the sonochemical treatment of horizontal wells in not sufficient to provide a full statistical analysis. However, the results obtained to date allow us to take an optimistic view on the potential of this technology. Thus far, we have treated 3 horizontal wells in sandstone reservoirs in Western Siberia. The time of ultrasonic treatment of 1 m of the formation after injection of the reagent was 15 min. Before and after sonochemical treatment of the well, geophysical studies of the well were carried out. Based on the information received, the zones for sonochemical treatment were determined. The treated area was 200--300 m long. As a result of sonochemical treatment, the production of fluid and production of oil from all three treated wells grew. On average, the production of fluid increased from 51 to 72 tons per day, and the production of oil increased from 23 to 33 tons per day.

5. Combined acoustical – chemical method for purification and disinfection of wastewater

Another application of high-power ultrasound, which has a big potential for use in industry, is the activation of reagents, used for wastewater treatment. Contaminated water from industrial sources can contain a range of pollutants, including heavy metals and oil, the latter often in the form of an emulsion. One general treatment adopted for such wastewater is coagulation, which involves the addition of aluminium salts, such as $AlCl_3$ or $Al_2(SO_4)_{3,}$ that generate aluminium hydroxide flocs. The coagulation process can also be initiated through the electrochemical generation of Al_3+ or Fe_3+ ions. These flocs absorb pollutants, which are concentrated in the material, which settles out and can be separated from the water [18]. Coagulation and electrocoagulation can also be used to break oil/water emulsions [19–21].

A valuable but so far relatively unexploited method for water remediation is the use of galvanochemistry for the generation of particles capable of removing metal ions and oil contaminants from wastewater [22]. The method is based on the use of two inexpensive bulk materials, i.e. iron and coke. Iron metal is corroded in the presence of coke which acts as the other part of the electrochemical cell due to their difference in standard electrode potential. The elements undergo intermittent contact during mechanical mixing and during this process iron becomes the anode and oxidises without the requirement for the application of an electric current (carbon is the cathode).

$$2Fe + O_2 + 4H^+ \rightarrow 2Fe^{2+} + H_2O$$

Fe^{2+} is then oxidized further and ions of Fe^{3+} are formed the majority of which is magnetic.

$$2Fe^{2+} + 1/2O_2 + 4OH^- \rightarrow 2FeOOH\downarrow + H_2O$$

There is no specific instrumentation required, and yet the technology is almost unknown outside of Russia; although, it presents a huge potential for both industrial wastewater treatment and hazardous waste remediation because of its effectiveness and low cost. The use of power ultrasound in various fields and has been widely studied [23]. It has been proven to provide process intensification in water purification through both its effect on heterogeneous systems and sonochemical reactions. The particle size of the iron oxide crystals (most of which is magnetite Fe_3O_4) generated in the galvano-reactor is around 8.5 µm, but subsequent ultrasonic treatment of these particles reduces the size to 5 µm. It is important to note that only the suspension of magnetite emerging from the galvano-reactor is sonicated in a flow system prior to the cleaning process in a separate reactor. This allows more intensive treatment of a concentrated suspension rather than treating the whole volume of the polluted water. In this way, power ultrasound can significantly increase the effectiveness of the treatment and expand its use to include the removal of oil pollution. Results are presented in Figure 8. Based on the methodology described above, a mobile system was constructed – see Fig. 9.

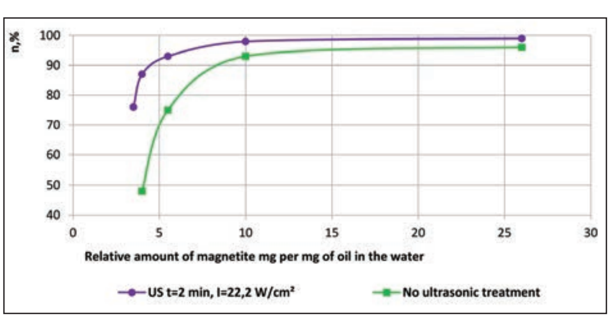


Fig. 8. Dependence of the degree of water purification (g) from the relative concentrations of magnetite and petroleum products in the contaminated water

Fig. 9. Mobile complex of ultrasonically assisted galvano--coagulation wastewater treatment. Placement of equipment inside the container

6. Ultrasonic-assisted production of antibacterial nanomaterials and textiles coating

Many efforts have been made for the application of ultrasound for the coating of various materials and fabrics with nanoparticles, in particular, textiles. The industrial potential of this application is tremendous [24]. Below, we have briefly described the procedure we have studied [25]. A method for the production of antibacterial ZnO nanoparticles has been developed. The technique combines the simultaneous treatment with ultrasonic waves and an electric current flow. By using high-power ultrasound, a cavitation zone is created between two zinc electrodes with the generation of a spatial electrical discharge in water. This discharge leads to the depletion of the electrodes and the formation of ZnO nanoparticles, which demonstrate antibacterial properties. At the end of this reaction, the suspension of ZnO nanoparticles is transported to a specially developed ultrasonic reactor in which the nanoparticles are deposited on the textile. The nanoparticles are embedded into the fibres by the cavitation jets, which are formed by asymmetrically collapsing bubbles in the presence of a solid surface and are directed towards the surface of textile at very high velocities. A SEM image of the treated fabric is shown in Figure 10. Fabrics coated with ZnO nanoparticles by using the developed method showed good antibacterial activity against *E. coli.*

We have tested the antibacterial activity against *E. coli* of the two sets of fabrics. Figure 11 shows the results of these tests. It is clearly visible that the antibacterial activity of the textile coated by the sonoplasma particles against E. coli is higher than the fabric coated with industrial NP's. This might be explained as the result of the small ZnO NPS obtained by the sonoplasma synthesis. We have repeatedly shown

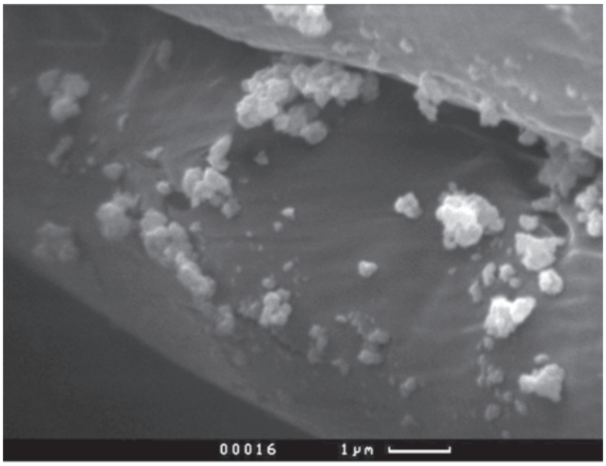


Fig. 10. SEM image of coated textile fibres. The scale bar is 1mm

that the biocidal effect is size dependent and particles with smaller size have a stronger bactericidal effect. The suspension continuously exposed to an ultrasonic field prevents the formation of particles agglomerates.

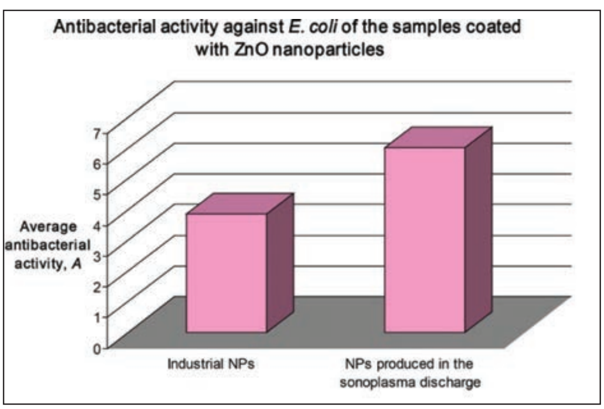


Fig. 11. Antibacterial activity against *E. coli* of ZnO nanoparticles coated fabric

Conclusions

Studies on fundamental phenomena related to high-power ultrasound propagation in liquids and solids paved the way for development of important applications of ultrasonic technologies. Ultrasonication can significantly influence heat and mass transfer in several chemical processes, modify the structure and properties of solids, and thereby interfere with interactions. This article reports a number of relevant industrial applications of ultrasound in which remarkable process intensification was documented. In general, ultrasound-assisted processes are faster, environmentally friendly, energy saving, and provide high quality products and new materials. We have shown that ultrasonic technologies can be successfully used in foundries, the oil industry, the production of bactericidal materials, and environmental protection. In spite of these relevant achievements, ultrasonic technology still remains unexploited and the industrial potential underestimated.

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