

TREATMENT OF AMMONIA WASTEWATER BY ULTRASOUND. PART I: THE INFLUENCE OF THE ULTRASOUND ENERGY ON THE ULTRASOUND BATH TEMPERATURE

Nicoleta MATEI (CIOBOTARU)^{1*}, Dan SCARPETE²

¹"Dunarea de Jos" University of Galati, Faculty of Engineering and Agronomy,
Calarasi Street, 29, RO-810017, Braila, Romania

²"Dunarea de Jos" University of Galati, Faculty of Engineering,
Domneasca Street, 47, RO-800008, Galati, Romania

*Corresponding author

e-mail: nicoleta.ciobotaru@ugal.ro

ABSTRACT

The industrial ammonia water decontamination depending on the sample temperature is monitored by this study. The treatment was conducted by the UP100S ultrasound generator (Hielscher Ultrasound Technology, Germany), operating at 30kHz frequency and acoustic power densities of 90 W/cm² and 460W/cm² respectively. The effect of sonication both on the bath temperature and ammonia removal, based on treatment time, is presented in this paper. Experiments were carried out according to different parameters, so as the sample temperature variation by ultrasonic treatment to be determined. Studied parameters were: the operating mode variation (continuous or intermittent), the additional aeration and the application of a cooling water serpentine. Based on the results, the ammonia removal efficiency is improved by the heating produced by the ultrasonic energy.

KEYWORDS: ultrasound, ammonia wastewater, acoustic cavitation, temperature, ammonia removal

1. Introduction

Among all the uses of power ultrasound, the treatment of wastewater containing toxic and complex pollutants, both from industrial and domestic sources, appears to be the most attractive field of study [1]. The advantages of this technology include the potential chemical-free and simultaneous oxidation, thermo-lysis, shear degradation, enhanced mass-transfer processes together [2].



Fig. 1. Compression and expansion cycle of ultrasound [4]

The enhancement in the processing rates is mainly due to the fact that, when ultrasound is passed through a liquid medium, it can generate cavitation phenomena due to alternate compression and rarefaction cycles [3], as shown in Figure 1 [4]. The cavitation can be a suitable technology for the degradation of wastewater streams or, at the minimum, it can be used for lowering the toxicity levels of the effluent stream so that conventional biological oxidation can be readily applicable [5].

The ultrasonic cavitation concentrates the energy and, with the collapse of bubbles, the energy is released within a tiny area, which generates a very high local temperature (around 5000 °C) and pressure (in excess of 500 atm), forming the so-called 'hot spots', which will open up widespread new chemical reaction routes and abruptly accelerate the chemical reaction rate [6].

During the treatment process, an ultrasonic generator transforms the electrical energy into other kinds of energies, as shown in Figure 2.

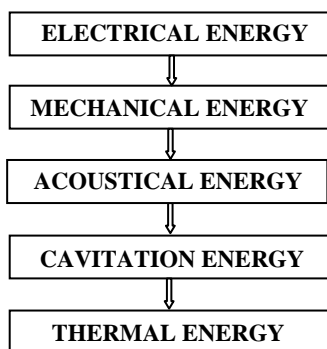


Fig. 2. The energy transformation chain during ultrasonic treatment [7]

In this work, the effect of the temperature rise due to the thermal energy emitted by the probe during sonication was assessed. The potential influence of the bulk phase temperature at ultrasonic treatment application and the optimum temperature range for different processes is discussed in many papers [8-15]. The thermal impact of continuously imploding cavitation bubbles on the surrounding liquid or sonicated matter itself depends on the physical properties of a sonication medium, such as vapor pressure or viscosity [16, 17], surface tension, gases dissolved and bulk temperature. According to many authors, the temperature increase of the sonicated liquid medium leads to an increase of the vapor pressure and a reduction of viscosity and surface tension [18-20]. With an increase in the vapor pressure of the liquid, the vapor content of the cavity increases thereby lowering the energy released during the cavitation collapse [18]. On the other hand, the reduction of viscosity and/or surface tension lowers the threshold intensity required to produce cavitation [19] and makes the effects temperature rise to be favorable. Increasing temperature results in reduction in acoustic cavitation threshold, meaning, the liquids cavitate at lower intensities [21]. Notwithstanding, Raman et al. explain that lower cavitation thresholds translate into the ease of cavity formation, thereby making higher temperatures more favorable for particle breakage [20].

The present study refers to the ammonia water decontamination by sonication and it is focused on the ultrasound influence on the heating occurring in the treated sample. This work is further continued by Part II, referring to the effect of ultrasound on ammonia removal. Considering that the separation of the ammonia-water mixture can be achieved by conventional distillation [22], heating appears to have a beneficial effect in ammonia removal. However, the separation performance is subject to the thermodynamic constraints of the system based on the volatility (boiling point) difference of the various

substances (e.g., -33.4 °C for ammonia and 100 °C for water at atmospheric pressure) [22, 23].

2. Materials and methods

2.1. Wastewater Features

This research is conducted on ammonia water decontamination by ultrasonic treatment. Several activities generate high-strength ammonia wastewater including human waste, agricultural waste and industrial effluent [24].

The sample utilized in the current paper is generated in the ion exchangers chemical industry. By washing the ammonium gas, ammonia water results as residual water. In order to carry out experiments in normal laboratory conditions, appropriate dilutions of 1:1000 were applied to the effluent. The final concentration of the sample subjected to tests was of 72.840 mg/l ammonium and 56.597 mg/l ammonia nitrogen respectively, after dilution. The volume of the sample to be treated was set at 300 ml.

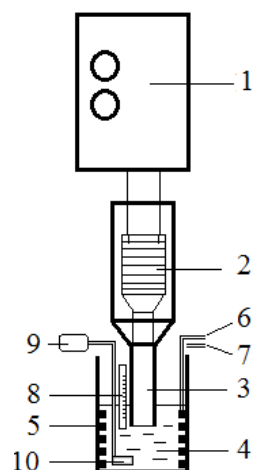


Fig. 3. Schematic representation of experimental set-up

The temperature increase was measured at room temperature by using a thermometer, which was immersed and held at the half height of the sonicated sample of ammonia water.

2.2. Experimental Set-Up

The schematic representation of the ultrasonic setup designed for the treatment of industrial ammonia water is shown in Figure 3, where: 1- electronic generator; 2- electromechanical transducer; 3- probe; 4- vessel containing ammonia water sample; 5- water cooling coil; 6, 7- hoses for cooling water in

and out; 8-thermometer; 9-aeration pump; 10-air stone.

The electric energy produced by the piezoelectric type generator 1 is converted to mechanical energy by transducer 2, further converted into acoustic energy in the form of ultrasonic waves transmitted through probe 3 to the ammonia water sample from vessel 4. This energy causes physical and chemical effects in the liquid medium to be treated and it is finally converted into heat.

In order to highlight both the effect of sonication and the effect of heating that occurs during the treatment process, the cooling water coil 5 was sunk in the vessel 4. The cooling water was taken from the current water source by hose 6 and the used water was directed to a sink by the drain hose 7. Experimental tests have been carried out both with and without water cooling coil. Ammonia water temperature is constantly monitored by the thermometer 8 immersed in the ammonia wastewater sample. To create the effect of bubbling, or the additional oxygen diffusion into the liquid, the aeration pump 9, fitted with air stone 10 (having the role of a fine spray air bubbles) were used.

The ultrasonic processor UP100H, Hielscher Ultrasonics GmbH, Teltow, Germany, was utilized to conduct the experiments. The generator is working at a 30 kHz fixed frequency, with the possibility to adjust the amplitude of the oscillating system. The amplitudes used in the treatment process were the maximum amplitudes working with each of the used probe (70 and 180 μm , respectively). Also, to each probe a different acoustic power density (ultrasonic intensity) corresponds, as follows:

- Probe MS 3 (3 mm diameter): 460 W/cm^2 ;
- Probe MS 10 (10 mm diameter): 90 W/cm^2 .

The maximum depth to which the probe was immersed in the tested liquid was 30 mm.

3. Results and discussion

For both acoustic intensities of the ultrasonic piezoelectric generator, various ways of temperature variation were registered. In the following, the dynamics of the temperature during sonication is discussed depending on the working parameters (with or without additional aeration, in either continuous or intermittent operation and depending on whether the cooling water coil is applied). Temperature values were read every 5 to 5 minutes for 60 minutes by the thermometer immersed in the solution. The constant value of temperature was noted after an hour of treatment, which is explained by the dynamic thermal equilibrium conditions of permanent transfers of heat between the treated ammonia water and the ambient air.

The initial temperature at which the readings started could vary by experiment, due to the ambient temperature of the laboratory, which was constantly monitored. However, of primary interest is the dynamics of temperature rise and the registered upper limit.

3.1. Thermal effect at higher acoustic intensity

Fig. 4 shows the dynamics of temperature depending on the operating mode (continuous or intermittent) and the additional aeration at higher acoustic intensity.

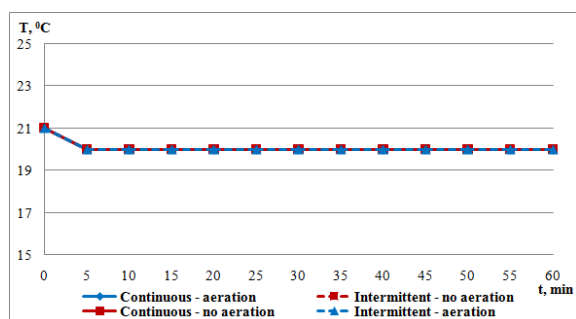


Fig. 4. Dynamics of temperature at 460 W/cm^2 acoustic intensity

The ultrasonic power of 460 W/cm^2 generates the temperature raise in the treated sample, up to a maximum constant at 34 °C, after 50 minutes, in continuous operation without additional aeration.

At additional aeration, the sample temperature reaches a maximum of 33 °C after 55 minutes, due to the air generated to the working vessel by the aeration pump attached to the set-up designed for the treatment of industrial ammonia water.

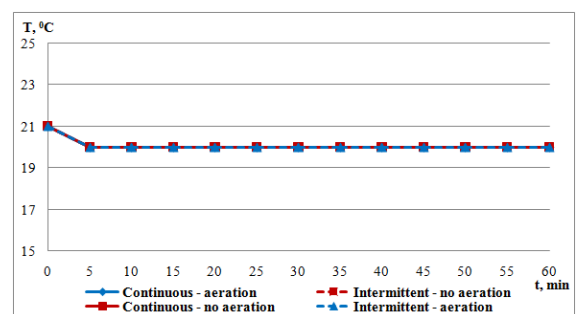


Fig. 5. Dynamics of temperature at 460 W/cm^2 acoustic intensity by cooling water coil

In the case of intermittent operation of 0.5 seconds, the temperature of the sample reaches 28 °C after 45 minutes, without aeration or 27 °C after 50 minutes with additional aeration.

Considering the role of maintaining the sample of treated water at a constant temperature, the applications where the cooling water coil was utilized recorded a decrease of 1 °C compared to the initial temperature of the water, according to Figure 5. This water temperature evolution was recorded when using the MS 3 probe in both continuous and intermittent operation. Additional aeration does not affect the studied water temperature.

3.2. Thermal effect at lower acoustic intensity

When the ultrasonic treatment is conducted at an acoustic power density of 90 W/cm², the sample heating is remarkable. The maximum constant value of 60 °C is reached after 60 minutes of continuous mode treatment, both with and without additional aeration. Intermittent operation of the ultrasonic irradiation generates a constant upper limit of 46 °C after 50 minutes without additional aeration or after 55 minutes with aeration applied (Figure 6).

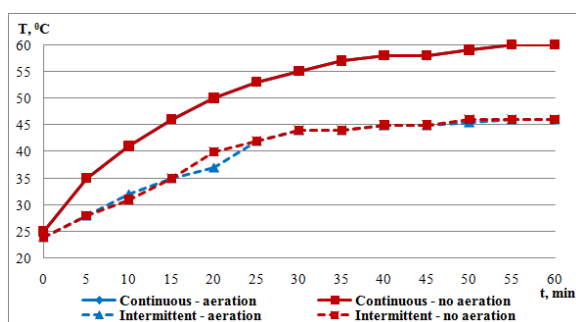


Fig. 6. Dynamics of temperature at 90 W/cm² acoustic intensity

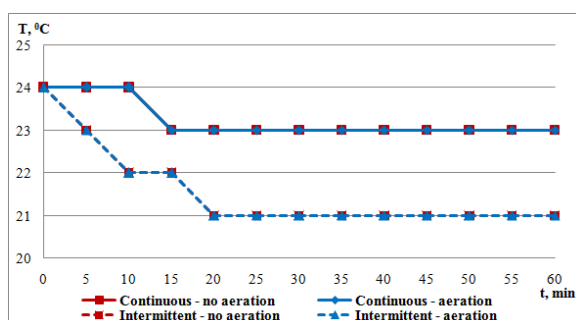


Fig. 7. Dynamics of temperature at 90 W/cm² acoustic intensity by cooling water coil

Figure 7 shows the effects on temperature when using the cooling water coil for the treatment with MS 10 probe. In intermittent operation, there is a temperature drop of 3 °C in the first 20 minutes of ultrasound treatment, followed by a temperature

maintaining to the value equal to the cooling water temperature throughout the treatment period.

In the continuous mode, the temperature remains constant, but the cooling is provided only with 1 °C, due to a much higher heating trend of the sample.

Any additional aeration has no influence on the studied water temperature.

In the second part of the study, complete results obtained in the treatment of ammonia water with a detailed discussion will be presented. Some of the objectives can be summarized as follows:

- The thermal effect in ammonia removal by ultrasonic technique is intended. In order to determine both effects of ultrasonic irradiation with heating and without heating on elimination rate, the experiments will be conducted by the addition of the cooling water coil.

- The removal efficiency according to allowable discharge limits (to natural water courses or public sewerage networks) will be determined.

4. Conclusions

The entire study refers to the determination of the optimum removal of ammonia from industrial ammonia wastewaters. In this part of the study, the influence of the acoustic power density on the liquid medium temperature by sonication was determined.

At higher ultrasonic intensity (460 W/cm²) the temperature rise is recording an upper limit of 34 °C in 60 minutes of treatment. The lower studied ultrasonic intensity (90 W/cm²) generates higher temperature rise rates of 60 °C after 60 minutes of treatment.

The beneficial effect of heating on the elimination of ammonia from liquid mediums is already known. However, to strictly observe the effect of ultrasonic treatment without additional heating effect, a cooling water coil to maintain a constant sample temperature was applied to the treatment setup.

Acknowledgements

The work has been funded by the Sectorial Operational Programme Human Resources Development 2007-2013 of the Ministry of European Funds through the Financial Agreement POSDRU/159/1.5/S/132397.

References

- [1]. P. R. Gogate *et al.*, Sonochemical reactors for waste water treatment: comparison using formic acid degradation as a model reaction, *Advances in Environmental Research*, 7, p. 283-299, 2003.

- [2]. **M. Matouq et al.**, *The kinetic of dyes degradation resulted from food industry in wastewater using high frequency of ultrasound*, Separation and Purification Technology, 135, p. 42-47, 2014.
- [3]. **V. S. Sutkar et al.**, *Theoretical prediction of cavitation activity distribution in sonochemical reactors*, Chemical Engineering Journal, 158, p. 290-295, 2010.
- [4]. **A. Mahvi**, *Application of Ultrasonic Technology for Water and Wastewater Treatment*, Iranian J Publ Health, 38, p. 1-17, 2009.
- [5]. **P. R. Gogate**, *Treatment of wastewater streams containing phenolic compounds using hybrid techniques based on cavitation: A review of the current status and the way forward*, Ultrasonics Sonochemistry, 15, p. 1-15, 2008.
- [6]. **J. Huang et al.**, *Low-MHz frequency effect on a sonochemical reaction determined by an electrical method*, Ultrasonics Sonochemistry, 2, p. S93-S97, 1995.
- [7]. **Z. Kobus et al.**, *Influence of physical properties of liquid on acoustic power of ultrasonic processor*, TEKA Kom. Mot. Energ. Roln. – OL PAN, 8a, p. 71-78, 2008.
- [8]. **V. A. Lemos et al.**, *Ultrasound-assisted temperature-controlled ionic liquid microextraction for the preconcentration and determination of cadmium content in mussel samples*, Food Control, 50, p. 901-906, 2015.
- [9]. **S. Rochebrochard et al.**, *Sonochemical efficiency dependence on liquid height and frequency in an improved sonochemical reactor*, Ultrasonics Sonochemistry, 19, p. 280-285, 2012.
- [10]. **M. Plesset**, *Temperature effects in cavitation damage*, Journal of Basic Engineering, 94, p. 559-566, 1972.
- [11]. **S. Hattori et al.**, *Influence of air content and vapor pressure of liquids on cavitation erosion*, Trans. JSME, 68 B, p. 130-136, 2002.
- [12]. **S. Hattori et al.**, *Influence of temperature on erosion by a cavitating liquid jet*, Wear, 260, p. 1217-1223, 2006.
- [13]. **B. Ondruschka et al.**, *Ultrasound in environmental engineering*, TUHH Reports on Sanitary Engineering, p. 139, 1999.
- [14]. **H. Destailats et al.**, *Applications of ultrasound in NAPL remediation: sonochemical degradation of TCE in aqueous surfactant solutions*, Environ. Sci. Technol., 35, p. 3019-3024, 2001.
- [15]. **L. Wenjun et al.**, *Removal of Organic Matter and Ammonia Nitrogen in Azodicarbonamide Wastewater by a Combination of Power Ultrasound Radiation and Hydrogen Peroxide*, Chinese Journal of Chemical Engineering, 20, p. 754-759, 2012.
- [16]. **P. V. Cherepanov et al.**, *Up to which temperature ultrasound can heat the particle?* Ultrasonics Sonochemistry, 26, p. 9-14, 2015.
- [17]. **M. Ashokkumar et al.**, *Sonochemistry*, in: Kirk-Othmer Encyclopedia of Chemical Technology, John Wiley & Sons, 2007.
- [18]. **P. R. Gogate et al.**, *Sonochemical reactors: Important design and scale up considerations with a special emphasis on heterogeneous systems*, Chemical Engineering Journal, 166, p. 1066-1082, 2011.
- [19]. **M. Goel et al.**, *Sonochemical decomposition of volatile and non-volatile organic compounds-A comparative study*, Water Research, 38, p. 4247-4261, 2004.
- [20]. **V. Raman et al.**, *Experimental investigations on ultrasound mediated particle breakage*, Ultrasonics Sonochemistry, 15, p. 55-64, 2008.
- [21]. **T. Mason et al.**, *Applied Sonochemistry: The Uses of Power Ultrasound in Chemistry and Processing*, Wiley-VCH Verlag GmbH and Co. KGaA, 2002.
- [22]. **X. Yang et al.**, *A Pervaporation Study of Ammonia Solutions Using Molecular Sieve Silica Membranes*, Membranes, 4, p. 40-54, 2014.
- [23]. **Y. A. Cengel et al.**, *Thermodynamics: An Engineering Approach*, Mcgraw-Hill College: New York, USA, ISBN-13: 978-0073398174, 2011.
- [24]. **S. H. Mirhossaini et al.**, *Effect of influent COD on biological ammonia removal efficiency*, International Journal of Environmental, Chemical, Ecological, Geological and Geophysical Engineering, 4, p. 86-88, 2010.