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Evolution of Cavitation Activity During Ultrasonic Nanostructuring of Magnesium

N. Brezhneva[*,†,§], N. V. Dezhkunov[‡], S. O. Mazheika[*],
A. Nenashkina[†] and E. V. Skorb[†]

[*]Faculty of Chemistry, Belarusian State University
Leningradskaya Street 14, 220030 Minsk, Belarus

[†]ITMO University, Lomonosova Street 9
191002 St. Petersburg, Russia

[‡]Research and Development Unit
Belarusian State University of Informatics
and Radioelectronics
P. Brovki Str. 6, 220013 Minsk, Belarus
[§]brezhNY@bsu.by

In this paper we focused on the evolution of transient cavitation activity during the sono-chemical treatment of magnesium aqueous suspensions. We have investigated the non-linear behavior of the cavitation activity related to the hydrogen released in the reaction of magnesium with water. Ultrasound modifies magnesium particles leading to the formation of nanostructured $Mg(OH)_2$ phase (brucite) resulting in the chemical and sonochemical impacts on magnesium.

Keywords: Transient cavitation; sonochemical nanostructuring; magnesium particle; hydrogen storage.

1. Introduction

Nowadays, ultrasonic (US) processing provides new possibilities in the synthesis of new nanomaterials, structuring solid surfaces for creating materials with the desired properties with proper control at the microlevel and nanolevel.[1,2] Among the benefits of sonochemical method, one could mention an ability to create non-equilibrium conditions during the synthesis in a specific manner. For example, the collapse of cavitation bubbles is accompanied by creating local areas of extremely high temperatures

(ca. 5000 K) and pressures (ca. 500 bar)[3] while the system treated by US remains on macrolevel at ambient conditions. The type of cavitation that deals with the phenomenon of bubble collapse on the final stage of the bubble cycle is called the transient cavitation.[4] Moreover, microbubbles that appeared during the sonochemical treatment of aqueous solutions can appear as microcontainers with free radicals and active ions that are appropriate species for the synthesis of nanostructures and modification of solids.

§Corresponding author.

Sonochemical treatment of metals and alloys surface leads to an enhancement of different materials' properties in comparison with non-treated original materials. For instance, the activation of electrocatalytic water splitting by Al–Ni alloy,[5,6] improvement of the Ti biocompatibility,[7–9] and formation of active aluminum surfaces[10] were observed. Recently, the use of sonochemically treated Mg for reactive hard templating has been demonstrated.[11] In addition, an interest to the Mg activation is caused by the perspectives of its application as a hydrogen storage material due to the light weight of and large content of hydrogen in Mg hydride structure.[12]

In this paper, we present experimental results on the evolution of the cavitation activity during US treatment of Mg aqueous suspensions.

2. Experimental

Aqueous suspensions of Mg powder (Alfa Aesar; 99.8%; −325 mesh) with different concentrations (0.003, 0.007 and 0.01 g/mL) were prepared by adding the appropriate amount of powder to 100 mL of distillated water.

In the experimental setup schematically shown in Fig. 1(a), the sonotrode was connected to an ultrasonic generator UZG 55-22 (20 kHz). The cavitation activity was registered with the hydrophone connected to a cavitometer ICA 5D and placed under the sonotrode tip at a distance of 1 cm. The probe at the hydrophone end acted as a waveguide coupling the acoustic signal from the cavitation zone to the piezoceramic plate. The electronic block of the cavitometer allowed displaying and processing the electric signal produced by the piezoelectric probe in response to the acoustic stimulation.[13]

3. Results and Discussion

During the sonochemical treatment of magnesium the following reactions occur:

$$H_2O = H^+ + OH^-, \tag{1}$$

$$Mg + 2H_2O = Mg(OH)_2 + H_2. \tag{2}$$
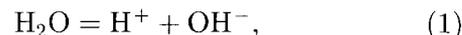
The reaction (1) is attributed to water sonolysis and the reaction (2) is related to the modification of Mg during sonication. Magnesium is affected by the chemical and sonomechanical impacts resulting in the etching of the metal surface and formation of the porous structure and magnesium hydroxide.

The transient cavitation is the main factor of US treatment when cavitation bubbles reach their critical size and violently collapse [Fig. 1(b)] producing extreme conditions with local areas of high temperatures.[3] For aqueous suspensions with different contents of Mg particles, various dependencies of cavitation activity were detected.
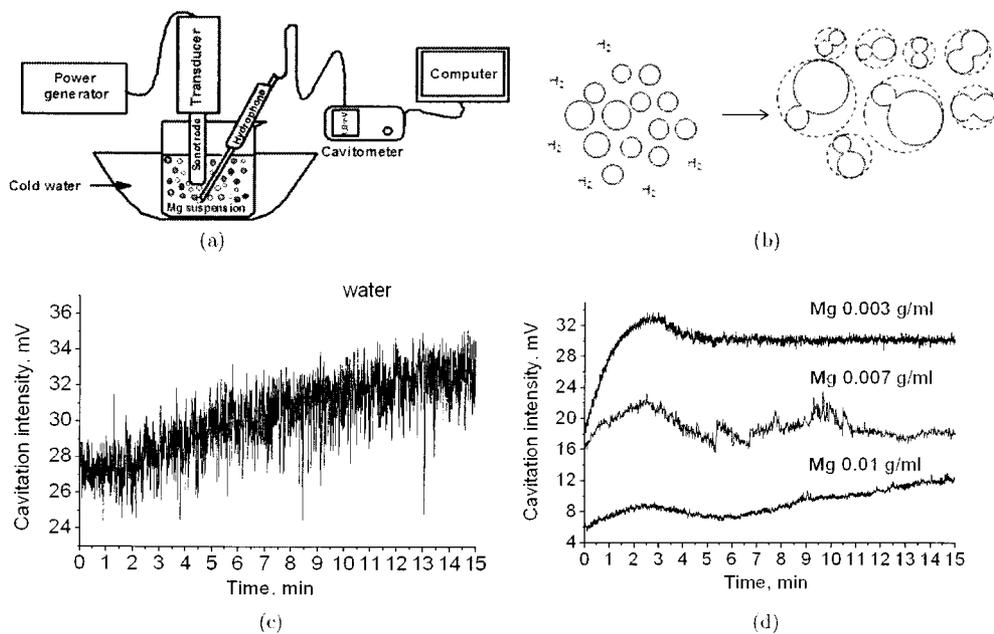

Fig. 1. (a) The setup for cavitation intensity measurements during the sonochemical treatment of Mg aqueous suspensions; (b) the evolution of the cavitation bubbles size; (c) and (d) the cavitation intensity during the US treatment of pure water and the Mg suspensions of different concentrations, respectively.

In the case of 0.003-g/mL suspension, the cavitation intensity increased till the third minute of treatment [Fig. 1(d)]. This fact can be connected with the formation of small bubbles that detach from the metal particles' surface and contribute to the cavitation intensity. During the US treatment of this suspension, the number of small bubbles increases and the cavitation intensity reaches its maximum. The increase in the amount of small bubbles can result in production of big bubbles by the possible coalescence mechanism [Fig. 1(b)] responsible for the decrease in the cavitation intensity. This phenomenon can be explained as the screening by the large bubbles the cavitation intensity on its way to be detected with hydrophone. After reaching its maximum, the intensity of cavitation further decreases and reaches the equilibrium value, when the rates of formation and destruction of large cavitation bubbles get equal.

Interestingly, when the amount of Mg is larger, the cavitation activity oscillations can be noted. In the case of 0.007-g/mL and 0.01-g/mL suspensions [Fig. 1(d)], after the stage of decrease in the cavitation intensity, the cycles of enlargement–reduction in the cavitation activity are observed and the increase in the concentration of the reactive metal leads to the formation of more hydrogen. We assume that the gas evolved by this chemical reaction influences the formation and collapse of cavitation bubbles. Gaseous hydrogen has the highest diffusivity in comparison with other gases.[14] H_2 bubbles released to liquid can generate more efficient cavitation than other dissolved gases due to its high diffusivity observed in water.[14]

Thus, in our case the *in situ* production of H_2 due to the chemical reaction of magnesium particles with water can affect the size of cavitation bubbles. These hydrogen bubbles can influence the bubble size through diffusion of gas molecules into the bubbles, leading to their enlargement and growth of the partial gas pressure inside the bubble making it more difficult to be collapsed by the external pressure. The following increase in the cavitation activity can be related to destruction of the larger bubbles and to the reformation stage of the smaller bubbles.[15] In the case of pure water such effect of cavitation intensity oscillations was not observed [Fig. 1(c)].

4. Conclusion

The analysis of transient cavitation activity in aqueous suspensions of Mg particles was carried out. The cavitation intensity increase can be attributed to the formation of small bubbles. The further decrease of cavitation intensity can be related to the formation of large bubbles by coalescence of small bubbles or the hydrogen diffusion produced during the chemical reaction of magnesium with water.

Acknowledgments

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