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A review of simultaneous ultrasound-assisted coal flotation

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Abstract

Ultrasound can be used both simultaneously or as a pretreatment technique for flotation to produce higher combustible recoveries, higher heat values, and lower ash data from raw hard coals. The recent research works have indicated that modifying coal surfaces, especially physical surface cleaning with the help of the cavitation process created by power ultrasound at certain frequency and time intervals, might cause significant changes in flotation responses by a thorough adsorption of reagents on coal surface, a high-speed jet of liquid is driven into the particles, and this jet may deposit enormous energy densities at the site of impact. The simultaneous ultrasonic treatment also causes significant changes in local temperatures and pressures within the slurry containing coal and a certain number of reagents at variable dosages during flotation. This treatment improves the effectiveness of reagent molecules at coal surfaces and interfaces due to their more uniform distribution in the pulp and also enhancement of the activity of the reagents used. This paper reviews the results of the recent studies and the possible mechanism of simultaneous ultrasound-assisted coal flotation.

Keywords: Coal, Flotation, Ultrasound, Simultaneous Ultrasonic Treatment, Ultrasonic Pre-treatment.

1. Introduction

Ultrasound, a sound wave propagating above the human perception limit, creates some extraordinary conditions in liquids such as cavitation [1]. The froth flotation process can be influenced by a large number of material, chemical, equipment, and operational variables. Changing one of these variables certainly significantly affects the results of flotation such as grade and recovery. Ultrasound is one of the most important treatment methods used to advance the flotation process [2-5].

The recent studies have shown that the separate elements of the flotation process could, in some cases, be favourably influenced by mechanical vibrations by the acoustic wave process or by the joint manifestation of these two physical phenomena [3]. Some applications of ultrasonic treatment in mineral processing and extractive metallurgy show that acoustic fields can produce significant positive effects on recoveries [6-8]. Previous researchers indicated that modifying solid surfaces, especially physical surface cleaning with the help of the cavitation process created by power ultrasound at certain frequency and time intervals before and during froth flotation, might cause significant changes in the adsorption of collectors on mineral surfaces, and accordingly, in their flotation responses [9-11].

While previous researchers [12-16] investigated the effects of ultrasonics before the flotation process for removal of adsorbed layers of reagents from mineral surfaces and emulsification of the reagents, the recent studies have concentrated on the effect of ultrasonic treatment during the flotation process, which is called the simultaneous ultrasonic treatment [17-24].

Chemical effects of ultrasonic treatment in a flotation system are characterized by cavitation, and are accompanied by a local increase in pressure and temperature. As solid/liquid

interactions are weaker than liquid cohesion forces, solid/liquid interfaces are more amenable to the formation of cavitation. The unsettled conditions caused at a solid/liquid interface can modify the surface properties of minerals, leading to changes in the adsorption of collectors on minerals, and accordingly, in their flotation responses. However, dispersive effects are realized when ultrasound is applied to a pulp containing a stabiliser such as a surfactant; this phenomenon concludes with the formation of an emulsion. Ultrasonic treatment can improve the effectiveness of a reagent due to a more uniform distribution in the suspension and also in enhancement of the activity of the chemicals used [25].

Various vibroacoustic effects on separate stages of the flotation process have been outlined as preparation of the pulp and reagents, formation behaviour of the bubbles, secondary and concentration in the froth layer, desorption of the reagents, and joint accomplishment of grinding and flotation [2-3]. The effects of vibroacoustics have been examined in the same study either on flotation, in general, or on the individual components of the flotation system; some devices for generating the desired high available frequency vibrations have also been discussed. It was found that by means of vibrations, it is possible

- to facilitate the attachment of mineral particles to air bubbles;

- to increase the recovery of valuable components;

- to improve selectivity, and correspondingly, achieve a rise in the quantity of valuable components in the concentrate;

- to increase the flotation rate;

- to form air bubbles with appropriate dimensions;

- to achieve a desorption of reagents from the mineral surfaces;

- to emulsify flotation reagents;

- to destroy viscous and resilent froths, etc.

Various studies haverevealed that intensive ultrasonic vibrations can effectively alter the state of a material in an acoustic field, causing dispersion, coagulation, and emulsification, of dissolution changing the rate and bringing crystallisation, about chemical conversions and accelerating multi-phase processes [26-27]. The flotation process is mainly dependent upon the state of the mineral surface, thus factors affecting the interface state cause changes during the flotation process. The

introduction of ultrasonic energy into a flotation system could produce changes in the qualitative relationships in the system and cause a change in the flotation rates [28].

Simultaneous ultrasonic treatment causes significant changes in the local temperatures and pressures within the liquid media containing solid minerals and a certain number of reagents at variable dosages during flotation. This energy can improve the effectiveness of reagent molecules at solid surfaces and interfaces due to their more uniform distribution in the suspension and also in enhancement of the activity of the chemicals used [29-30].

Power ultrasound is known to create its effect via cavitation bubbles. When power ultrasound is applied to a liquid in sufficient power, frequency, and intensity, the liquid is alternately subjected to compression and expansion forces giving rise to cavitation bubbles. Application of power ultrasound to a liquid containing a mixture of solid particles makes the bubbles collapse near a solid surface, a high-speed jet of liquid is driven into the particles, and this jet can deposit enormous energy densities at the impact site.

The interaction forces involved in the physical adsorption of a reagent molecule during the froth flotation process are weaker than the forces involved in chemical adsorption. The physical bond between the reagent molecule and the mineral surface can be easily broken by the hydrodynamic turbulence created by ultrasound. Ultrasound facilitates reagent adsorption by exposing clean surfaces, and produces highenergy centers on the mineral surface for the reagent molecule to adsorb, increasing the flotation recoveries. It is known to promote the precipitation of gas particles followed by the formation of bubble nuclei. The deposition of such stabilized micro-bubbles, particularly on hydrophobic particles, can improve the bubble-particle collision efficiency, and therefore, flotation recoveries are increased [31].

The introduction of ultrasonic energy into a flotation system could produce changes in the qualitative relationships in the system and cause a severe change in the flotation rates. Some studies indicate that sound irradiation may change the pH values, surface tensions, and oxidation-reduction potentials of flotation pulps with a certain increase in the local temperature and pressure. Previous researchers have stated that application of ultrasound for flotation of various minerals such as ilmenite, rutile, and zircon even for a short time period considerably increase their flotation

response and cause significant changes in the recovery and grade values. These improved values are believed to be due to the effective cleaning of particle surfaces from film coatings, namely slimes [32].

The interaction forces involved in physical adsorption of a reagent molecule during the froth flotation process are weaker than the forces involved in chemical adsorption. The physical bond between the reagent molecule and the mineral surface can be easily broken by the hydrodynamic turbulence created by ultrasound. In case of chemical adsorption, the interaction forces between the reagent molecule and the mineral surface are stronger and can not be broken by the ultrasonification. Increased flotation recoveries in the presence of ultrasonic treatment could be explained by cleaning and formation of micropits on mineral surfaces with the ultrasound effect [33-37].

It has been reported that the acoustic or ultrasonic pretreatment of minerals such as ilmenite, rutile and zircon, even for a short time, considerably increases their flotation response and causes significant changes in the recovery and grade values. These improved values are believed to be due to the effective cleaning of particle surfaces from film coatings, namely slimes. These studies have also shown that separation of cinnabarantimonite, coal-ash, barite-fluorite, sphaleritepyrite-chalcopyrite, galena-quartz, manganese oxides-jarosite-ilmenite-ferromolybdenite, and copper-molybden bulk concentrates could easily be achieved by froth flotation with the help of an acoustic or ultrasonic treatment [2-3].

The effects of ultrasonic treatment on flotation of colemanite-clays, magnesite-quartz, and coal-ash have also been studied [9, 19-24].

In one of the studies, the hydroacoustic cavitation (HAC) treatment of flotation has been investigated for nominal -150 μ m Illinois-basin bituminous coal. It has been reported that cavitation is produced either by flow through an ultrasonic resonator chamber or enhanced cavitation by initial flow through a cavitation chamber followed by flow through the ultrasonic unit. The results obtained have indicated that desliming substantially improves flotation [12].

The effects of ultrasonication on the hydrophobicity of oxidized pyrite have recently been reported and the flotation of an oxidized pyrite ore has been investigated. It has been mentioned that ultrasonication shows opposite effects on the hydrophobicity of oxidized pyrite by surface cleaning and further oxidation of both slightly and heavily oxidized pyrite within a very short time period [13].

Some researchers have studied the effect of ultrasonic irradiation as a pretreatment method for high-ash coal flotation and its kinetics. They applied ultrasonic treatment prior to flotation at different ultrasonic power levels and periods. Their flotation experiments were conducted based on a two-level fractional factorial design with six variables: ultrasound intensity, collector dosage, conditioning time, pH, rotor speed, and solid content. Their optimized parameters were reported as follow: ultrasound intensity, 30 W/cm²; 1500 g/t collector; 11 min conditioning time; pH = 7.5; 800 rev/min of rotor speed; and 10% of solid content. They concluded that their kinetic flotation experiments were 10% faster using the ultrasonic conditioning in comparison with the conventional conditioning [34].

Another investigation has been performed on the use of low frequency ultrasound wave with a low frequency in column flotation to remove ash and pyritic sulfur from bitumen. The effects of four parameters including the amounts of collector and frother agents, ultrasound power, and ultrasound times were investigated, and the optimum conditions for column flotation were noted as 1.5 kg/t bitumen of collector, 0.4 ppm of frother, an ultrasound power of 300 W, and an ultrasound time of 3 min under a flotation time of 5 min. They used central composite design (CCD) of response surface methodology (RSM) and multilayer feed forward back-propagation neural network for the interpretatation of their results [35].

The flotation recovery of copper sulfide tailings was studied at the El Teniente plant. They found the effect of acoustic cavitation that cleans particle surfaces and minimizes slime coatings, facilitating the action of the reagents with the help of ultrasound [38].

It has also been reported that slime particles reduce the floatability and increase the ash content of clean coal. In his work, the ultrasonic pretreatment method was used as a pretreatment method to enhance the selectivity as well as the kinetics of many minerals such as magnesite, complex sulfide ore, silica, coal, galena, and zinc. The ultrasonic pretreatment time as well as the reagent (collector and frother) dosages were optimized to achieve a maximum clean coal yield. With ultrasonic pretreatment, the clean coal yield increased for all the reagent dosages. Moreover, the clean coal yield obtained through ultrasonic pretreatment was very close to the theoretical yield calculated by the release analysis. Improvement in clean coal yield with ultrasonic pretreatment was validated with the characterization studies such as XRD, zeta potential, petrographic, and FT-IR analysis [39].

2. Use of an ultrasonic bath and a transducer

The ultrasonic application began in the present author's several studies carried out during early 1990's and 2000's for flotation of colemanite and magnesite ores separately by both simultaneous and as a pretreatment method [9, 19-24]. In one of those studies, Zonguldak hard coal samples were floated with the help of ultrasound. The ultrasonic flotation results showed that effective desliming occurred and the combustible recoveries increased while the ash data of the floated coals decreased. Figure 1 shows a photo of the system [40-43].

In another study, a plexiglass flotation cell equipped with a single frequency ultrasonic transducer (35 kHz) connected to a Sonorex type generator (200 W power) was applied for flotation of a fly ash sample. The results obtained showed that there was an improvement in the flotation rate with a decrease in the ash contents of the floated samples. Figure 2 shows some photos of the system [44].



Figure 1. Direct use of an ultrasonic bath.



Figure 2. A plexiglass flotation cell equipped with a single frequency ultrasonic transducer.

3. Use of an ultrasonically-assisted flotation cell Within the present author's other previous works [21-23], coal floatability was investigated using a specially designed Wemco-type flotation cell (1.25 L volume, 1200 rpm impeller speed) ultrasound transducers equipped with on representaive hard coal slimes from Bottrop mine (Germany). Ultrasonic power generation was provided by a $(2 \times 300 \text{ W})$ generator with various power levels (at 10% intervals) and frequency modules (25 and 40 kHz). The manufacturer of the ultrasonic equipment (Bandelin Electronic GmbH&Co. KG in Berlin) built each transducer with a 50 W power capacity and different dimensions according to different frequency requirements, i.e. 25, 40, and 25-40 kHz. The manufacturer has reported that the overall energy loss might be approximately 10%, and that the electroacoustic overall efficiency per cell might be approximately 65%. The losses are independent from frequency. During the coal flotation experiments, the ultrasonic power generator was run at up to 50% of its total output power during all stages of the process, i.e. conditioning and flotation. The flotation operation parameters were chosen as 5 min of conditioning time, 5 min of flotation time, and a variable amount of the Ekofol-440 reagent as the coal flotation collectorfrother combination. The Bottrop mine site local water used as the sample was in the original pulp form with a rough solid ratio of 8-12%, ambient temperature, natural pH, electrical conductivity, and oxidation-reduction potential of the slurry.

A photo of the system is shown in Figure 3. The results obtained indicated that ultrasonic treatment during coal flotation positively affected the quality and quantity of the properties of floated coals, while using lesser amounts of reagent than a conventional flotation system. The results showed that conditioning and aeration, and therefore, total flotation times were certainly shortened. It was observed that the froth or air bubble sizes were uniformly distributed inside the flotation cell and tended to become finer during the ultrasonic flotation tests. Reagent consumption was drastically decreased by the use of ultrasound due to surface cleaning of coal particles, which may be due to the thorough contact between the collector-frother and coal particles. Although the quality of the floated coal seemed not to be affected by the use of ultrasound, the tailing part of the flotation products did not seem to contain coal particles; this certainly caused an improved recovery of coal flotation by ultrasound. These conclusions could be attributed to the thorough surface cleaning of coal particles by ultrasonic energy given to flotation system, and this certainly gives an explanation for more positive results than the conventional coal flotation.

The effects of simultaneous ultrasonic treatment on flotation of the original and aged hard coal slimes were investigated with the aim to recover more combustible and good quality products [19]. The conventional and ultrasonically-assisted flotation tests of the original and aged coal slimes were performed using Montanol-531, a special coal flotation reagent at variable dosages. The results of the batch flotation tests revealed that simultaneous ultrasonic treatment increased the combustible recovery and lowered ash values of concentrates compared to the conventional flotation conditions despite the use of similar reagent dosages. These results were also verified by testing some physico-chemical properties of samples. temperature. the i.e. pH. oxidation- reduction potential, and zeta potential. Finally, it was concluded that a thorough surface cleaning by cavitation due to power ultrasound

might improve the flotation recovery compared with the conventional flotation conditions, even though very slight changes were also observed in some physico-chemical properties of the treated samples. The physical cleaning mechanism of ultrasonically created cavitation bubbles onto hard coal samples is suggested in Figure 4. In the conventional flotation conditions, although the density of mixture of hydrophobic coal particles, reagent, and air bubble together are lower than the water density, slimes (mostly clays) are present, and they prevent reagent adsorption on mineral surfaces, and hence, this causes a decrease in the flotation recovery. However, in the case of ultrasonic flotation conditions, cavitation bubbles occur during the process, and are replaced by clay particles due to surface cleaning by high energy transfer onto interfaces. At the same time, the reagents are much more effective under ultrasound due to thorough surface cleaning effects. This phenomenon might cause a higher adsorption of reagents to coal surfaces, and therefore, the coal recovery increases.



Figure 3. A group of simultaneous ultrasound-assisted flotation cells.



Figure 4. Suggested mechanism of ultrasound-assisted bubbles by cavitation [19].

4. Use of a refurbished ultrasonic bath and flotation cell combination

In a recent work [46], Ozkan has investigated the flotation of very finely-sized representative hard coal samples obtained from the Bottrop mine using a newly designed sub-aeration type flotation machine and cell combination refurbished from an ultrasonic bath with a 35 kHz frequency variable power levels (80-160 W). The flotation reagent was Ekofol-440 with variable dosages (40-300 g/t). The batch coal flotation results were analyzed by comparing the combustible recovery and separation efficiency values, taking the yield and ash contents of the products into account. Particle size distribution of the ultrasonically treated and untreated coals were also measured in order to understand the effects. The results obtained showed that the simultaneous ultrasonic coal flotation increased the yield and recovery values of the floated products with lesser ash values than the conventional flotation despite the us of similar reagent dosages.

Figure 5 shows a photo of the re-designed flotation machine and cell combination as well as some auxiliary equipment for all the experiments. The ultrasonic generator of the bath was run at 100% of the total output power during all stages of flotation, i.e. conditioning and aeration. Using the whole cell volume, the total energy use for each flotation trial could be calculated as 30 and 60 W/L. The flotation tests were categorized into different groups of conventional (no ultrasound) and simultaneous ultrasonic flotation experiments (with power levels of 30 and 60 W/L). While the main paramater studied was the ultrasonic power per volume, the other flotation variables were kept constant in each experiment for ease of comparison and evaluation of the results.

The purpose of these tests was to demonstrate and to clarify the yield and combustible recovery differences between the conventional and simultaneous ultrasonic flotation tests. The batch flotation results were analyzed by comparing the combustible recovery and separation efficiency values, taking the yield and ash contents of the products into account [45].



EQUIPMENT NO:

 Refurbished ultrasonic bath used as a flotation cell
Impeller of a refurbished flotation machine
Air valve

- 4. Electrical mains
- 5. Froth collection pan
- 6. Camera

Figure 5. A re-designed flotation machine and a cell combination.

4.1. Effects of ultrasound on combustible recovery and separation efficiency

The conventional and ultrasonic coal flotation results were compared according to the combustible recovery and separation efficiency values as well as the ash contents of floats and tails with increase in the reagent consumption as separate graphs in Figure 6 [46]. It could clearly be seen that increasing the reagent dosage from 40 g/t to 300 g/t drastically changed the combustible recovery from 20% to over 90% in the conventional flotation conditions; however,

separation efficiencies seemed to reach from 20% to 60%. This may be due to increase in the ash contents of both the tailing and float products collected in froths with increase in the reagent dosage. It may be said that the optimal reagent dosage could be around 150 g/t because of achieving acceptable recovery and ash contents of the flotation products.

When simultaneous ultrasonic treatment was applied, the combustible recovery and separation efficiency values were clearly affected by achieving acceptable recovery and ash contents of the products even with consumption of half the reagent dosage of the conventional conditions. The optimal reagent consumption could be seen around 80 g/t in order to achieve the acceptable results. However, varying the ultrasonic power level from 30 to 60 W/L does not necessarily

change neither the optimal reagent dosage nor the acceptable values for flotation products. The separation efficiencies were also lower than the combustible recoveries due to collection of all the coal particles together with the ash-forming minerals connected to the froths.



Figure 6. Comparison of combustible recovery and separation efficiencies (top two graphs) with varying reagent dosage against ash contents (bottom two graphs) [46].

4.2. Effect of ultrasound on particle size distribution and froth structure

In order to understand whether the simultaneous ultrasonic treatment affected the particle size distribution of the float part of the flotation products, the fractional and cumulative particle size distributions of the conventional and simultaneous ultrasonic coal flotation tests were measured by separately sieving the froth parts after the flotation tests ended. Figure 7 shows a comparison of the particle size distribution of the original feed with floated coals according to the simultanous ultrasonic treatment. It could be seen in Figure 7 that particle size distribution was certainly affected due to the simultaneous ultrasonic treatment. The initial mean particle size value (d_{50}) was known to be around 40 microns.

This value increased to around 60 microns during the conventional flotation, and 70 and 80 microns after low and high ultrasonic treatments, respectively. This may be due to the removal of a certain amount of finely-sized ash forming clays together with coals after flotation. It is interesting to note that applying and increasing the simultaneous ultrasonic treatment from 30 to 60 W/L may have caused aglomeration of coal particles in the froths despite the fact that ultrasound may cause size reduction with increasing power levels [46].

It was also observed that froth or air bubble sizes were uniformly distributed inside the flotation cell and tended to become finer during the ultrasonic flotation tests, as seen in Figure 8 [46].



Figure 7. Comparison of particle size distributions [46].



Figure 8. Comparison of froth sizes (left: conventional, right: ultrasonic).

5. Conclusions

The results of the recent studies conducted by the present author and his teammates, especially covering the further investigations on simultaneous ultrasonic coal flotation using a newly designed flotation machine and cell combination refurbished from an old ultrasonic cleaning bath equipped with a single frequency and two different power levels and a sub-aeration type old flotation machine were outlined. It was reported that the simultaneous ultrasonic treatment drastically decreased the optimal reagent dosage from 150 g/t to 80 g/t due to physical surface cleaning, therefore, achieving thorough contacts between the collector-frother and the coal particles. Within the optimal reagent dosage, over 90% combustible recovery and 70% separation efficiency values were achieved, although the rough combustible recovery was thought to be about 60-65% for the Bottrop mine.

It was commented that tailings did not seem to contain coal particles, although the quality of the floated coal seemed not to be affected by the use of ultrasound, therefore, this might explain the improved recovery of coal flotation by ultrasound. The particle size distribution was certainly affected due to the simultaneous ultrasonic treatment. While the mean particle size value (d_{50}) was around 40 microns, this value increased to around 60 microns during conventional flotation, and 70 and 80 microns after low and high ultrasonic treatments, respectively. This might be due to the removal of a certain amount of finely-sized ash forming clays together with coals after flotation. It was also interesting to note that applying and increasing the simultaneous ultrasonic treatment from 30 to 60 W/L might have caused aglomeration of coal particles in the froths despite the fact that ultrasound might cause size reduction with increasing power levels.

These conclusions might be interpreted as that the positive effects of the simultaneous ultrasonic treatment were mainly due to physical cleaning of very finely-sized ash-forming clays from coal surfaces due to the cavitational forces.

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مروری بر فلوتاسیون زغالسنگ تحت امواج مافوق صوت

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چکیدہ:

امواج مافوق صوت به صورت همزمان یا قبل از فرآیند فلوتاسیون ذرات زغالسنگ، برای افزایش بازیابی مواد سوختنی، افزایش مقدار گرما و کاهش میزان خاکستر استفاده می شود. تحقیقات اخیر نشان داده است که ایجاد تغییر در سطح ذرات زغالسنگ به ویژه تمیز کردن سطح فیزیکی آن به کمک فرآیند ایجاد حباب که توسط امواج مافوق صوت در فرکانس و فاصلههای زمانی مشخص به وجود آمده است، ممکن است علت تغییرات قابل توجه در نتایج فلوتاسیون باشد که به وسیلهی جذب کامل معرفها روی سطح زغالسنگ به دست آمده است. هنگامی که امواج مافوق صوت به دوغاب زغالسنگ اعمال می شود باعث قرار گرفتن حبابها در نزدیک سطح زغالسنگ می شود. جریان مایع با سرعت زیادی به ذرات وارد شده و این جریان ممکن است انرژی بسیار زیادی را به محل ضربه وارد کند. استفاده از امواج مافوق صوت باعث ایجاد تغییرات قابل توجهی در دما و فشارهای محلی داخل دوغاب حوی زغالسنگ و مقادیر متغیری از معرفها، در حین کند. استفاده از امواج مافوق صوت باعث ایجاد تغییرات قابل توجهی در دما و فشارهای محلی داخل دوغاب حوی زغالسنگ و مقادیر متغیری از معرفها، در حین فلوتاسیون می شود. این رفتار، تأثیر مولکولهای معرف روی سطح زغالسنگ، اتصالهای ناشی از توزیع یکنواخت آنها در پالپ و همچنین افزایش فعالیت معرفهای استفاده شده را بهبود می بخشد. در این پژوهش نتایج مطالعات اخیر و مکانیسم احتمالی فلوتاسیون زغالسنگ با استفاده از امواج مافوق صوت باست قریکی شرا فرای معرف می محل مربر می معرفهای استفاده شده را بهبود می بخشد. در این پژوهش نتایج مطالعات اخیر و مکانیسم احتمالی فلوتاسیون زغالسنگ با استفاده از امواج مافوق صوت بر سی

كلمات كليدى: زغالسنگ، فلوتاسيون، امواج مافوق صوت، امواج مافوق صوت همزمان، امواج مافوق صوت قبل فلوتاسيون.