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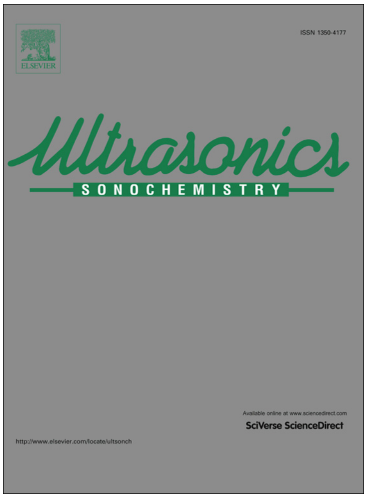

Ultrasonic cleaning: an historical perspective

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ULTRASONIC CLEANING: AN HISTORICAL PERSPECTIVE

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Introduction

Ultrasonic cleaning is nowadays regarded nowadays as a conventional technique for industry and also in both scientific and medical laboratories. Its origins date back to the 1950's and it was beginning to become established around forty years ago. In a series of reviews on the uses of power ultrasound in industry "Macrosonics in Industry" Neppiras suggested that ultrasonic energy performed a physical function in the process of cleaning which could not be obtained by any other industrial tool. He further maintained that its ultimate success depended on the selection of proper equipment and materials, a knowledge of both cavitation and chemical cleaning techniques together with process control [1]. A later review in the series dealt exclusively with cleaning and in it Bulat claimed that this was probably the commonest use of power ultrasound and one which was being improved continually [2]. Nevertheless we seldom give a thought as to why ultrasonic cleaning has proved to be so widely accepted.

In terms of its historical development it is reasonable to ask what factors have made it important ? In other words what are its advantages over more traditional cleaning methods ? To help answer these questions we can explore the alternatives that were available in the 1950's when ultrasonic cleaning first emerged as a technology. Many of the cleaning methods available then are still in use today and so if we consider these then it will become easier to appreciate the reasons why surface cleaning with ultrasound has gained such prominence.

Survey of non-ultrasonic cleaning technologies

The need for large scale and heavy-duty washing and cleaning has existed since the industrial revolution or even before. There are several different approaches to these more traditional cleaning processes but they can be grouped in terms of the ones used in each of the various types of manufacturing industries.

Heavy industry

After machining and/or assembly of individual parts most engineering products must be cleaned free of cutting oil residues and swarf. This will also be true when parts are dismantled and recycled because ingrained debris must be removed.

For degreasing the most common method in the past was immersion in a hot chlorinated solvent. In the days before health and safety concerns precluded such materials from common use these methods were certainly more effective than the use of aqueous or semi-aqueous immersion processes [3]. An alternative to total immersion is vapour degreasing where the object to be cleaned is placed in a heated vapour tank above a chlorinated solvent. The vapour combines with the grease to form droplets that fall back into the solvent tank. Vapour degreasing is ideal for reaching into small crevices in parts with convoluted shapes and also to remove more stubborn soiling. An additional benefit is that parts degreased in chlorinated solvent or vapours come out of the process dry; there is no need for an additional drying stage, as required in water based technologies.

The major drawback to such processes is of course the health and environmental problems associated with the use of chlorinated solvents such as carbon tetrachloride (CTC), tetrachloroethylene (PCE), trichloroethylene (TCE) and 1,1,1-trichloroethane (TCA) which were four of the most widely used cleaning and degreasing solvents. The history of the production and use of these four compounds can be linked to the development and growth of the synthetic organic chemical industry in the USA [4]. In the early years of the 20th century, CTC and TCE were used as a replacement for petroleum distillates in the dry-cleaning industry. The latter became the solvent of choice for vapour degreasing in the 1930s. but in the 1960s TCA became increasing popular [5]. During the 1980s environmental and safety issues led to the banning of chlorinated solvents for parts cleaning and in the 1990s, CTC was phased out under the Montreal Protocol due to its role in stratospheric ozone depletion.

It became clear that aqueous systems should replace chlorinated solvents but methods were then needed to make the water based cleaning more efficient. One route was to improve the performance of detergents for immersion cleaning and this required considerable chemical development. Mechanical methods were also required to ensure that detergent solutions would reach all parts of the surface of the object to be cleaned. Two alternatives emerged which have remained popular to this day: pressure jetting and parts washing. The two differ in that pressure jetting involves a pressurised jet of water plus detergent directed, often manually, at the item to be cleaned. In contrast a parts washer is used to clean smaller engineering items generally placed on some form of carousel contained within an enclosed cabinet. The cleaning is achieved by spraying or immersing the parts in aqueous detergent.

Food industry

In the food industry baked on deposits or residues on molds or cutting tools need to be hygienically removed. Traditional methods involve simply soaking in a water/detergent/bactericide mixture together with agitating and heating which is followed by a rinse cycle. The choice of detergent is key to this and so is the operating temperature with higher cleaning temperatures being more effective. As with industrial cleaning pressure jetting or a form of parts washing are sometimes used to help in the removal of heavily adherent material [6].

Medical instruments

More specific methods are needed for the cleaning of surgical instruments, medical implants and dental implements. The cleaning method must both remove dirt and sterilise the surface. The former can be done with an automated washer-disinfector to carry out the process of cleaning and disinfection consecutively. Generally though for full sterilisation an autoclave is required.

Clothing and textiles

Traditionally clothing and textiles were cleaned in stirred hot water with detergent. The process temperature depends on the fabric but the overall process is one of tumbling with hot aqueous detergent followed by rinsing and drying. Not much has change here except that newer detergents are produced and the washing can be done at significantly lower temperatures down to 30^{o}C.

The origins of ultrasonic cleaning

It is difficult to trace the actual “eureka” moment when ultrasound was applied to cleaning technology. The original discovery that ultrasound could be used to improve cleaning does not appear to be published as any kind of authenticated fact. Indeed it is not at all obvious why one would want to apply ultrasonic irradiation to a cleaning system. What is clear however is that by the 1950’s there were a number of companies who had developed ultrasonic cleaning systems. Amongst these in the

USA were the Bendix Corporation in Davenport, Iowa, Branson Cleaning Equipment Co., Danbury, Connecticut and Zenith Ultrasonics, Norwood, New Jersey while in the UK there were Mullard in Redhill, Surrey and Kerry, Hitchin, Hertfordshire.

In a report on the 20th Engineering, Marine and Welding Exhibition held at Olympia in London the Engineer magazine reported on a development in cleaning by Mullard Ltd [7]. The company had on show a mechanised ultrasonic cleaning plant built in conjunction with Kerry Ltd suitable for removing loose contamination (e.g. swarf, lapping compounds, oil and grease) from engineering parts. The parts to be cleaned were in baskets that passed through three tanks in succession first, through two tanks containing trichlorethylene a pre-wash tank and then an ultrasonic cleaning bath powered by a 2kW ultrasonic generator and finally through a hot vapour zone for drying. The ultrasound was at a continuously variable frequency between 10 and 30 kHz. Apart from the solvents used the basic set up is much the same as with today’s automated ultrasonic cleaners.

Ultrasound is particularly useful for surface decontamination because of two factors related to cavitation in a liquid medium:

- Above the cavitation threshold non-symmetric collapse of a cavitation bubble near to a surface results in the formation of a powerful jet directed at the surface which can dislodge dirt and bacteria. This is an effective mechanism for conventional cleaning systems operating in the 40 kHz range.
- When acoustic waves pass through the cleaning fluid acoustic streaming occurs which reduce the thickness of hydrodynamic boundary layer on any immersed surface. As a result tiny particles on the surface become more exposed to the liquid streaming which can overcome the adhesion force between particle and surface. This process becomes important in high frequency 1 MHz megasonic cleaning.

The particular advantage of ultrasonic cleaning in this context is that it can reach crevices that are not easily accessible using conventional cleaning methods. Objects that can be cleaned range from large crates used for food packaging and transportation to delicate surgical implements such as forceps. The use of ultrasound allows the destruction of a variety of fungi, bacteria and viruses in a much reduced processing time when compared to thermal treatment at similar temperatures. The removal of bacteria from various surfaces is of great importance to the food industry and can be efficiently accomplished with the combined use of sonicated hot water containing biocidal detergent [8].

For small and delicate items such as computer components, silicon wafers and printed circuit boards the method of choice is megasonic cleaning and this will be dealt with later in this article.

The development of ultrasonic cleaning – a consideration of parameters that affect efficiency

Cleaning fluid

The cleaning fluid plays an important part in determining the effectiveness of an ultrasonic cleaner. In the early days, as with conventional cleaning, chlorinated solvents were used e.g. perchloroethylene, trichloroethylene, 1,1,1-trichlorethane, carbon tetrachloride. For ultrasonic cleaning Colclough emphasised that the solvent was not only as the cleaning medium but also as an organic liquid used to transmit the ultrasonic vibrations from the transducer to the object being cleaned [9]. The fluid is required to solvate as much of the dirt as is possible and so the chemical characteristics are very important. Apart from the viscosity, surface tension and vapour pressure of a liquid it should also have good cavitating properties and Antony emphasised the importance of choosing the right solvent for each cleaning task in an article published in the first ever volume of the journal Ultrasonics [10]. For grease removal the solvents of choice were halogenated hydrocarbons and acetone but for more general cleaning of dust-contaminated components he suggested a mixture of 8-12% alcohol in water while for removing oxides and slight descaling various combinations of a solution of hydrogen peroxide, formic acid and distilled water could be used. In the same year and also in the first volume of Ultrasonics Crawford published a paper entitled “A Practical Introduction to Ultrasonic Cleaning” [11]. He concluded correctly that the rapid growth of ultrasonic cleaning has been due, at least in part, to attempts to reduce the many man-hours entailed in normal cleaning methods. Ultrasonic cleaners save time and often produce results better than any other method, ensuring a progressive future for this technique. Five years later a discussion of the scale up of cleaning can be found in the same journal [12]. The article identified the three basic processing configurations available at that time for large scale ultrasonic cleaning as in-line, carousel and tank. The importance of ultrasonic cleaning to industry is emphasised but organic solvents were still the main cleaning fluids for industry.

In the latter part of the 20th century there was a definite move away from halogenated and other organic solvents and aqueous solvents came into favour. This move was driven by environmental concerns and the effect of solvent vapours on the health of factory workers.

Temperature

The temperature of the bath is another important parameter that must be considered with ultrasonic cleaning. Temperature has an effect upon the intensity of the cavitation of the liquid. An investigation of the variation of relative intensity of cavitation with temperature was determined by chemical and erosion methods [13]. The former involved the liberation of chlorine from a saturated solution of carbon tetrachloride in water and the latter as the loss in weight of lead samples after exposure to cavitation. Niemczewski reported the cavitation intensity over a range of temperatures for 37 organic liquids and water [14]. He found that the maximum cavitation intensity of water occurs at 35°C despite the fact that most aqueous ultrasonic cleaning solutions operate best between 50 - 65℃. He suggested that this was due to the effect of reagents added to ultrasonic cleaning solutions such as acids, alkalis or detergents because these could produce a stronger cleaning effect at 60℃ than at 35℃.

Standing waves

Another factor that can influence the performance of cleaning baths is the presence of an acoustic standing wave. This can happen when a transducer at the base of a tank emits a single frequency and the wave hits the surface of the liquid and is reflected back into the tank. The resulting standing wave will produce active cavitation zones at fixed points over the depth of the bath corresponding to half-wave distances for the frequency used. This problem has been solved by cleaning bath manufacturers by inserting a circuit into the ultrasonic generator that will cause the signal that is sent to the transducer to vary slightly in frequency over a set period of time - a frequency sweep. In this way the standing wave is avoided and the sweep, with lower maximum pressure than a standing wave, will move up and down within the tank and so distribute the energy more evenly.

Power

An increase in the power fed to the transducer will produce a rise in the vibrational amplitude of the emitting surface and so it might be expected that this would increase the cleaning effect of an ultrasonic bath. But the situation is rather more complicated than this. There will be an upper limit in the vibrational amplitude above which the transducer will suffer mechanical fracture but before this occurs there will be a reduction in the vibrational energy that a transducer can transfer to the liquid. The generation and collapse of cavitation bubbles is the source of energy for cleaning but if a large number of cavitation bubbles are formed in front of the emitting surface of the transducer these can act as a barrier to the transfer of acoustic energy and dampen the power transmission to the bulk of the tank. When the emitting surface is driven at higher amplitudes the physical motion of the surface travels too fast for the bath liquid to remain in contact with it so a gap is generated between transducer and liquid and the majority of the acoustic energy is lost, this is termed decoupling. For this reason there will be a maximum amount of energy that can be transmitted efficiently into the liquid medium because of cavitation bubble shielding and “decoupling”. A good example of this effect can be found in the field of sonochemistry in the production of iodine from aqueous KI via free radicals produced by cavitation bubble collapse. In a classic example the initial iodine yield first increases in a relatively linear fashion above the cavitation threshold but then reaches a plateau for a while before decoupling sets in and the yield drops dramatically despite the increased power supplied by the transducer [15]. Generally, for any cleaning (or sonochemical) process, there will be an optimum power for maximum effect. This will depend on a range of conditions but will mean that power optimisation can lead to a considerable saving in the overall economics of the process.

Frequency

The majority of ultrasonic cleaning systems, which were developed in the 1950s operated in the range of 20 to 40 kHz. Nowadays the frequency used is almost entirely around 40 kHz. This is because 20 kHz can be heard by younger workers but 40 kHz is inaudible to all workers who use the machines although there will still be associated vibrations from the metal casings and other parts of the equipment. In the early years there seemed to be no real need to move outside of this lower frequency for cleaning but this situation changed. In 1986 McQueen compared the efficiencies of two cleaning systems one at 40 and the other at 220 kHz [16]. This revealed cases where increasing the ultrasonic frequency increased the rate of decontamination particularly in cases where the contaminants were sub microscopic (e.g. fingerprints, lubricant paste). On the other hand materials such as blood clots were removed more quickly by the lower frequency ultrasound.

The main interest in using higher frequencies has come in more recent times where it has been recognised that cavitational damage to delicate objects can be minimised at frequencies around 1 MHz. This is generally referred to as megasonic cleaning and has been adopted in the semiconductor industry where it is extremely important to avoid surface damage of silicon wafers during cleaning [17] [18]. Two types of mechanism were suggested to explain the way in which megasonic cleaners operate. The first is a direct interaction of the sound field with the attached particle, i.e. the oscillating acoustic field exerts periodic forces directly on particulate matter attached to a boundary or surface. The other is that unlike the collapse of cavitation bubbles at lower frequencies bubbles produced at high frequency are much smaller and have a tendency to resonate rather than collapse. Any microscopic air bubble present in the liquid would undergo stable, large-amplitude pulsations which cause rapid movement of the surrounding liquid as it follows the oscillating bubble boundary. Microstreaming patterns could then develop, not as intense as those induced by cavitation collapse, nevertheless sufficient to dislodge particles as small as 0.1 μm from a surface. Crum has reported investigations into megasonic cleaning which indicate that the origin of the effect may involve some cavitation activity near to the surface [19].

Measuring the performance of cleaning baths

From the very beginning of ultrasonic cleaning there has always been the question of how to determine the efficiency of a cleaning process. The configuration of the bath and other aspects of ultrasonic cleaning began to become more significant with passing years and there was a move to calibrate and assess the efficiency of ultrasonic cleaning machines.

Visual inspection

The basic requirement of any cleaning process is that it should remove contamination from a surface. It is often possible to see that an object is cleaner than when it was put in the bath and this can be performed visually or with the aid of optical magnification. The contamination can be made more visible by the addition of fluorescent dyes to the object or by viewing under ultraviolet light. This is undoubtedly the most widely used and simple method for the rapid assessment of cleaning efficiency but it cannot be regarded as accurate. Certainly such simple visual inspection cannot determine the cleanliness of areas that are hidden from sight such as crevices. Normal eyesight is also unable to detect thin biofilms or nanoparticles remaining on the surface after cleaning.

Gravimetric analysis

For small parts it may be possible to determine the removal of dirt by simply weighing them before and after cleaning. This type of test is less suitable for large items because the material removed in cleaning is only a tiny proportion of the overall mass of the object and as a consequence accuracy is compromised.

The mechanical effects of cavitation can also be determined by measuring the loss of material from a test specimen through erosion damage. If a solid piece of metal such as lead is placed in a cavitating field pitting erosion will occur after several minutes of activation. This gives a method of comparing the cavitational activity of different cleaning baths since the relative amount of sonic energy expended to achieve a particular mass loss can be obtained. A similar technique can be used using the perforation of aluminium foil of known dimension. In practice this methodology is not very quantitative because of poor reproducibility. However, apart from comparing the performance of different baths the foil test may also be used in a qualitative sense for the location of active zones within a cleaning bath. The positions where the foil is subject to maximum perforation is the zone of maximum cavitation in the bath.

Removal of deliberate soiling

A method which dates back to the very origins of ultrasonic cleaning but has undergone many developments is by cleaning an item of standard dimensions that has been deliberately soiled. In the old days this might have been graphite on ceramic surfaces or emulsion paint on metal. Here again the assessment would have been visual and it is necessary to find a "standard" dirt and a reproducible method of attaching the contaminant to the sample to be cleaned. In 1972 Pohlman suggested a suitable measuring process for the quantitative determination of the degree of cleaning [20]. The technique involves observation of the transparency of a glass plate measured by photometry before and after coating with ink and then subsequently after ultrasonic cleaning. The frequency of the cleaning bath used in the original report was 18.1 kHz and the optimal cleaning efficiency in terms of the various wave-forms fed to the transducers was obtained using half-wave modulation.

Today ultrasonic cleaning is used more and more for the cleaning of medical and dental instruments. In these situations it is necessary to have a different "standard" pollutant which reflects medical contamination [21]. A number of such pollutants are available for this purpose one of which is known as Edinburgh soil and consists of a mixture of egg yolk, horse blood and pig mucin. In the analysis of cleaning efficiency a microbiological assessment of the surface of the cleaned item will also be required.

Cavitation

Whatever method is used to determine the cleanliness of an item any user will want an objective method of monitoring the performance of the bath so that results can be reproducible. To this end a measurement of acoustic cavitation activity in the cleaning bath provides a method that allows the cleaning equipment to be set at the same level and achieve the same effects every time it is switched on. This is also a requirement in sonochemistry [22] and there are parallels in both approaches as can be seen when comparing two papers dealing with this topic from the points of view of cleaning [2] and sonochemistry [23].

In an extensive review of practical methods for the measurement and characterisation of acoustic cavitation a large number of available methods were compared [24]. In all thirteen different systems were studied including chemical dosimeters, calorimetry and hydrophones together with some additional work involving the mapping of acoustic fields. The results showed some promising correlations between the various methodologies

Power

Perhaps the simplest method of estimating electrical power consumption by a cleaning bath is to directly measure the power consumption from the electrical mains supply. While certainly this is important in terms of calculating the cost of the process for industrialists it does not take into account the electrical efficiency of the generator or transducer. The net acoustical power entering the bath can be measured by immersing a hydrophone or cavitometer. The former can be rather fragile but the latter is more robust and can be used in strongly cavitating media. Both devices convert the vibrational energy within the bath into electrical signals by the piezoelectric effect. More detailed review of the types of the methods available for the measurement of cavitation activity can be found in two recent publications [25] [26].

Calorimetry

A general method for estimating the power entering an ultrasonic cleaning bath is calorimetry. This involves the measurement of the rise in temperature of the bath liquid over a short period of time after the transducer has been switched on. This gives some estimate of the acoustic power entering the system (i.e. the acoustic energy absorbed by the solution). However it is only an estimate because it does not take into account any sonochemical degradation of the liquid or erosion of the emitting surface. In addition there is a component of heating from the surface of the transducer itself which can act as a sort of “hot plate”. Nevertheless this is a simple method of estimating input power even when a thermally insulated vessel is not used [22, 27, 28].

Chemical dosimetry

There are a number of different methods available for the measurement of acoustic energy in a bath using its effect on a chemical (usually radical) reaction. A common problem with such methods however is that the bath is normally of large volume requiring a lot of chemicals and so these dosimeters are normally used in small containers which can be dipped into various parts of the bath to determine cavitation bubble activity. These dosimeters are much more generally used for sonochemistry rather than for cleaning bath systems [29]. There are many different dosimeters available including the Fe^{2+}/Fe^{3+} dosimeter (Fricke-dosimeter) [30], terephthalate dosimeter [31], iodine dosimeter [32] and *para*-nitrophenol dosimeter (PNP-dosimeter) [33].

Conclusions

Ultrasound is particularly effective for cleaning because it is capable of dislodging and removing surface contamination in the form of inorganic dirt or microbiological material through the shock waves and jet formation that accompany acoustic cavitation bubble collapse. This type of cleaning can be used for both small and large items and can penetrate deep into crevices and cavities in the surface of an object. The major advantages have been recognised from the start of the use of ultrasonics in cleaning and include:

- Increased cleaning speed which can often be applied to assembled components without the need to break them down into individual units.
- If a frequency sweep is used to avoid standing waves in the cleaning bath all areas of an object can be reached to give uniform cleaning.
- Ultrasound generally works well with water based solvents which can be used to replace the more hazardous halocarbons.
- The micro-streaming effect induced by the jet formed on collapse of a bubble improves mass transfer from the bulk cleaning solvent to the surface i.e. provides cleaner solvent for flushing the surface.

Ultrasonic cleaning was developed many years ago but is still developing as more refined scientific and engineering applications are found requiring specialist forms of surface treatment.

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Abstract: The development of ultrasonic cleaning dates from the middle of the 20th century and has become a method of choice for a range of surface cleaning operations. The reasons why this has happened and the methods of assessing the efficiency of ultrasonic cleaning baths are reviewed