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Feasibility of using contactless electromagnetic cavitation for steel composite manufacturing

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Abstract

This work investigates the feasibility of using contactless electromagnetic sonication for dispersing and refining a strengthening phase in iron and steel for ferrous metal matrix composite (MMCs) production. An oscillating pressure field is generated using superposition of alternating (0.13 T) and steady magnetic fields (up to 8 T). The processing employs a static floating-zone technique and is crucible-free. This approach allows to reach around 0.8 MPa pressure oscillation amplitude that is sufficient to initiate acoustic cavitation in high melting temperature liquid metals. The viability of various reinforcement dispersion, both ex and in situ, using this electromagnetic sonication is explored in the context of oxide dispersion strengthened (ODS) steel and high modulus steel (HMS) production.

Introduction

Metal matrix composites (MMCs) are an important material class providing a high degree of tailorability to fulfil various specific structural and functional requirements. However, production of high melting point metal composites, such as Fe- and Ni-alloys, is mostly limited to solid state routes due to the high reactivity of the matrix. While the liquid state approach is economically more attractive, an additional problem is flotation and agglomeration of the reinforcement phase. Ultrasonic cavitation, which is a violent implosion of cavities within the liquid generating highly energetic events, has been used extensively to solve these problems for light MMC, e.g. aluminium and magnesium, processing [1].

However, the conventional route of introducing cavitation directly is not possible in molten steel due to a rapid dissolution and erosion of any sonotrode. As a solution, an innovative set-up based on a static floating-zone melting has been developed. By superposition of a steady axial magnetic field to induced azimuthal currents, cavitation has been successfully generated in various low melting temperature liquid metals (aluminium, tin, zinc) [2] as well as in iron and steel [3] in a contactless, crucible-free way.

While sonication of ferrous melts has been demonstrated, reinforcement particle dispersion in these liquids is not as straightforward. Composites can be produced by two methods: ex situ and in situ [4]. With ex situ approach the reinforcement phase is prepared first and then combined with the matrix phase without any reaction taking place while with in situ composite manufacturing, the reinforcing phase is synthesized in the matrix by chemical reactions between the elements.

Liquid iron is a rather extreme environment – the high melting temperature together with irons' property of generally reactive wetting of various ceramics [5] limit available reinforcements that could be used for ex situ processing while at the same time supporting composite production in situ. Therefore, two straightforward research questions emerge: can

ultrasonic cavitation of ferrous melts 1) facilitate particle wetting and, thus, dispersion with ex situ approach (while keeping the reinforcement phase stable) and 2) provide a significant refinement of precipitates with in situ approach?

This work tries to provide answers to these questions by investigating the viability of using cavitation for distributing reinforcing phase, both ex and in situ, in the context of oxide dispersion strengthened (ODS) steel and high modulus steel (HMS) production. The composites are produced using electromagnetic sonication by superposition of alternating and steady magnetic fields (up to 8 T). This corresponds to 0.8 MPa oscillating pressure amplitude. The aim is to show an overview of various investigated reinforcement-matrix pairs and provide an initial starting point for selecting the most prospective pairs for future experimental work.

Working principle and experimental processing

To investigate the feasibility of particle dispersion and subsequent production of composites by contactless sonication, different high melting point matrix-particulate systems have been examined. The chosen matrix materials are stainless steel grade 1.4404 and pure iron. Oxide particles (ZrO_2, CeO_2, Al_2O_3) have been investigated in context of ex situ ODS steel production, while different carbides and borides (TiC, ZrC, TiB_2, ZrB_2) have been used for in situ HMS fabrication. In both approaches the reinforcements are added using a *master alloy*. The particles are mixed together with the matrix powder and compressed in an electrically conducting tablet. Reinforcement amount in the end product corresponds to around 0.6 wt.%.

The produced samples are cylindrical with dimensions of 20×20 mm. The molten region is extracted and the specimens are processed with the conventional metallographic steps. Optical light (Leica DM6000 M) and scanning electron microscopes (SEM; Zeiss EVO 50) are used for microstructural analysis and energy-dispersive X-ray spectroscopy (EDX; Bruker XFlash detector 4010) is used for elemental analysis.

A sketch of the experimental set-up and the relevant electromagnetic fields is shown in Figure 1 (a). A compressed tablet (master alloy) (1) consisting of the matrix material and reinforcement powder is placed in between two supports (2) which are made of the same matrix material (iron or steel). These are melted together with the tablet creating the composite. To prevent oxidation, the melting is done in a 5 mbar argon atmosphere. The processing is enclosed by a quartz tube (3) from the sides. A 4-turn induction coil (4) is used for melting the workpiece. Melting is controlled by water-cooled copper plates (5) from top and bottom.

The induction coil generates an alternating magnetic field, b_{AC} (blue), that induces a current, j_{in} (purple), in the azimuthal plane of the workpiece. When the workpiece is liquid, a steady magnetic field, B_z (green), is applied axially either by an electromagnet (pole shoes marked as (6)) or by a superconducting wide-bore magnet system at the Dresden High Magnetic Field Laboratory (HLD-EMFL). The electromagnet can provide up to 0.8 T magnetic field, while the superconducting magnet can reach 19 T. The static magnetic field interacts with the induced currents creating a radially oscillating body force, F_{osc} (red), in the melt which in turn produces a time varying pressure. The achievable pressure amplitude, p_A, that directly creates cavitation, can be estimated as $p_A = b_{AC}B_z/\mu_0$ (μ_0 is the magnetic constant) [2]. Melting and processing is done with I_{AC} = 1300 A at f_0 = 14 kHz. The induced magnetic field has been directly measured and corresponds to b_{AC} = 0.13 T at the centre of the coil.

A sign of cavitation is a distinct acoustic signal that consists of a variety of harmonics ($n\,f_0$, n = 2,3…), sub-harmonics (f_0/m, m = 2,3…) and ultra-harmonics ($n\,f_0/m$, n = 2,3… and m = 2,3…) of the fundamental driving frequency f_0. In general, the onset of the phenomenon is marked by a broadening of the spectral lines, increase of the broadband noise and appearance of sub- and ultra-harmonics [6]. This emitted acoustic signal is recorded by piezo elements (7) attached to the copper plates. A representative power spectrum of recorded cavitation activity in iron is presented in Figure 1 (b).

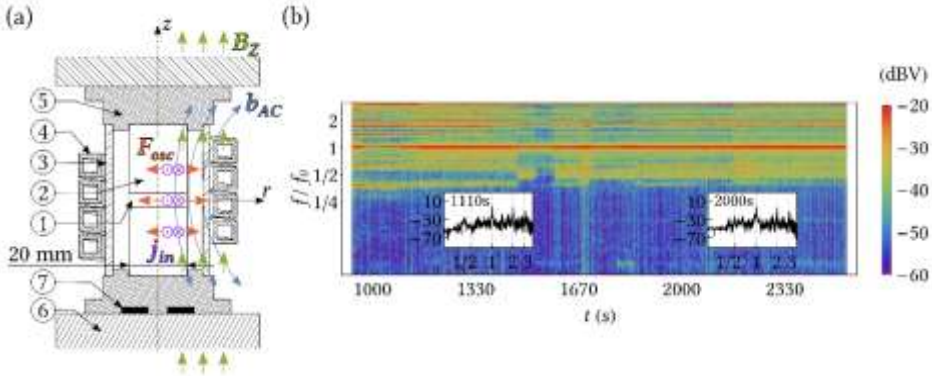

Figure 1: Sketch of the experimental set-up and working principle in (a): (1) tablet containing the powder and matrix mixture, (2) supports from the matrix material, (3) quartz tube, (4) induction coil, (5) water-cooled copper shielding plates, (6) electromagnet poles, (7) position of the piezo elements that record the emitted sound. Spectrum from cavitation signal is shown in (b): sonification of Fe-ZrC composite by applying 0.8 T strong magnetic field. The insets at 1110 and 2000 seconds present the instantaneous signal demonstrating varying intensity of cavitation activity. The background noise slightly increases at later stages of processing, together with appearance of $f_0/2$ peak.

This two dimensional spectral image clearly demonstrates the dynamics of the cavitation activity during the whole experiment. The presented figure is obtained by applying a 0.8 T magnetic field during melting of a Fe-ZrC composite. The shown time range represents processing of a fully molten state. The insets at 1100 and 2000 seconds show the instantaneous signal. Various characteristic peaks of cavitation, e.g. $f_0/3$ and $2f_0/3$ can be clearly identified at the start. During the melting, the background noise increases and the spectral lines widen. At 2000 seconds an additional $f_0/2$ peak can be observed. In general, such cavitation onset has been obtained with various degrees of intensity with all the presented matrix-reinforcement pairs. It must be noted that the observed spectral recordings correspond to the so-called "stable" cavitation [7]. The employed acoustic recording arrangement dampens the high frequency signal that is usually associated with the much more intense "inertial" cavitation regime. This has to be improved to obtain a complete understanding regarding the achieved cavitation via the contactless route.

Results and discussion

Ex situ processing

As oxides are chemically more stable, the expected behaviour is low reactivity, but weak wettability. In this case, the purpose of cavitation is to improve the wetting that would aid with the dispersion of the stable particles within the matrix. While in light alloy melts cavitation provides conditions for the unwetted particle surfaces to be cleaned forcing the melt in crevices, with the investigated matrices this was not observed. Selection of representative results of ex situ oxide reinforcement behaviour in ferrous melts is shown in Figure 2.

Dispersion of Al_2O_3 in Fe is shown in (a) with low (left side, 0.8 T) and high (right side, 6 T) magnetic field processing. Alumina structures appear black in the micrograph, while the iron matrix is light grey. The high stability translates to a bad wettability. However, application of cavitation of any intensity does not improve the end product. In all the recorded cases similar behaviour was identified – the small particles tend to agglomerate in clusters which are removed to the periphery of the sample during solidification. This also has been observed in steel matrix.

ZrO_2 in steel is shown in (b) with no (left) and high (right, 5 T) magnetic field processing. In both cases large agglomeration (black structures, enlarged in the insets) can be observed and the particles are pushed to the edges of the matrix – cavitation, irrespective of its intensity, does not improve dispersion of the zirconia in the steel grade used.

CeO_2 in steel is demonstrated in (c) with no (left) and high (right, 5 T) magnetic field processing. Similar to other oxides, ceria particles are, most likely, stable in ferrous melts and are not well wetted by them. While some small clusters have been identified deeper in the matrix, the particles mostly can be found on the edges of the matrix as well as agglomerated around inner cavities. The same behaviour has been observed in iron matrix.

Why cavitation does not improve the dispersion of these reinforcements can be explained as follows. According to the Rayleigh–Plesset equation, the response of cavities to an applied oscillating pressure depends on the fluid properties. With some simplifications and for very small bubbles with radius R_0, the minimum transient cavitation pressure-threshold, P_T, can be found as $P_T \approx 4\gamma/3\sqrt{3}R_0$ [6], i.e. for small bubbles the surface tension, γ, is dominant. Essentially, a cavitating bubble will have much stronger response to the same applied oscillating pressure field in aluminium (γ = 914 mN m^{-1}) than in iron (γ = 1872 mN m^{-1}). In addition, the mutual attraction force between two unwetted particles is also proportional to the surface tension [8]. Therefore, while cavitation can be achieved, 1) with the same oscillating pressure it might not be as potent as in light metals and 2) the forces holding the particle clusters together are much larger in ferrous melts.

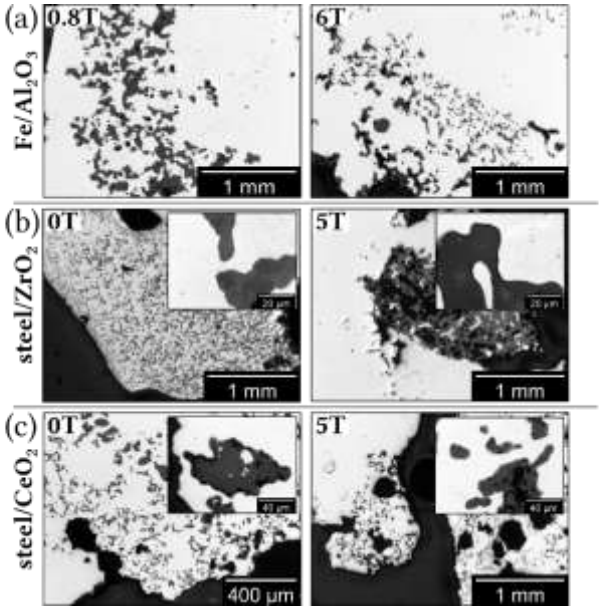

Figure 2: Micrographs of various oxide particle behaviour in ferrous matrices depending on applied magnetic field strength: (a) Al_2O_3 in pure iron processed with 0.8 T (left side) and 6 T (right side); (b) ZrO_2 in steel processed without cavitation (left) and with 5 T (right) magnetic field (high intensity cavitation); (c) CeO_2 in steel processed without cavitation (left) and with high intensity cavitation (right). Insets in (b) and (c) show enlarged clusters of the reinforcement. The reinforcing phase is dark against the grey (matrix) background.

In situ processing

For most of the refractory carbides and borides a dissolution in ferrous melts is expected. In this case, the reinforcing phase is created in situ by precipitation – during solidification the reinforcements precipitate from the hypo- or hypereutectic alloy. Cavitation, by means of dynamic nucleation, provide conditions for improved refinement and dispersion of this phase [9]. Selection of representative results of in situ carbide and boride precipitate refinement in ferrous melts is shown in Figure 3.

Figure 3 (a) demonstrates addition of ZrB_2 particles to iron. The sample is processed with either 0.8 T (left) or 6 T (right) strong fields. In both cases a layer of precipitates that are tens of microns large appear at the top and bottom of the sample. Insets show this phase enlarged. While with stronger cavitation the clustering is slightly smaller, it is not fully reduced and there is no significant refinement. Similar structures were also observed with ZrC. The middle of the

specimen has limited amount of particles due to the growth and subsequent removal as larger structures. This has to be prevented to use zirconium compounds in composite manufacturing.

Figure 3 (b) shows TiC particle dispersion in the middle of a steel matrix processed with either 0.8 T (left) or with 5 T (middle) strong magnetic field. With weak cavitation, some large clusters of primary TiC particles are formed as can be seen on the left side and in the EDX map in the inset. This is completely avoided in the other sample. In the bulk of the matrix, precipitates are elongated reaching around 10 μm. In the case of 5 T the particles are more equisized, furthermore, much smaller precipitates (around 1 μm) can be recognized. The EDX map on the right side shows titanium distribution of the micrograph in the middle. Similar refinement has also been observed in pure iron [3].

Figure 3 (c) shows results of TiB_2 particle dispersion without cavitation (left) and processed with 8 T strong magnetic field (middle). Without cavitation strong agglomeration of TiB_2 phase occurs and clusters as seen in the left micrograph are found throughout the middle of the matrix. Application of 8 T magnetic field destroys these particle clusters and the TiB_2 precipitates (black contrast in the micrograph; EDX map on the right shows titanium distribution) are evenly dispersed in the matrix. In pure iron primary particle clustering without cavitation has not been observed, however, the precipitate size and microstructure is generally improved by applied cavitation as well [3].

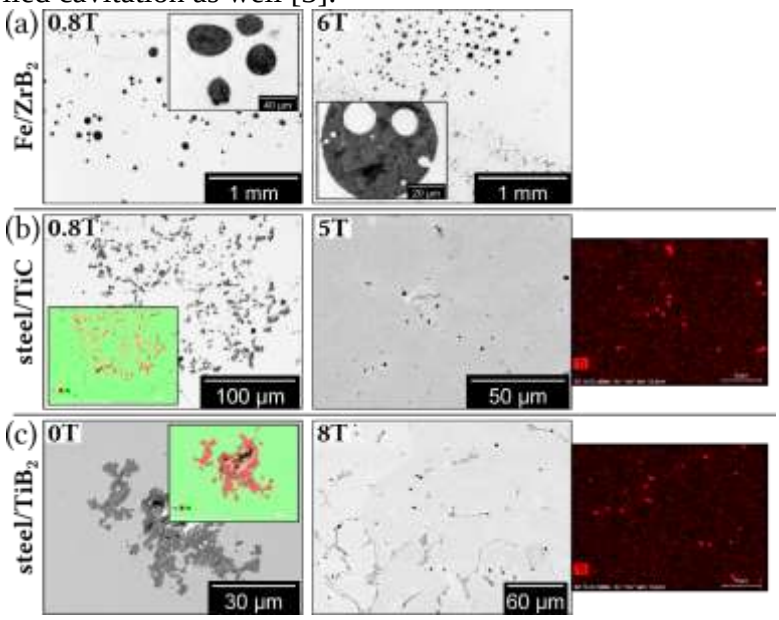

Figure 3: Micrographs of various boride and carbide particle behaviour in ferrous matrices depending on applied magnetic field strength: (a) ZrB_2 in pure iron processed with 0.8 T (left side) and 6 T (right side); (b) TiC in steel processed with 0.8 T (left) and 5 T (middle) magnetic field; (c) TiB_2 in steel processed without cavitation (left) and with high intensity cavitation (middle). Inset in (a) shows typical large particle agglomerates, while EDX maps in (b) and (c) show elemental analysis of visible reinforcements. The reinforcing phase is dark against the grey (matrix) background in the micrographs.

Conclusions

Cavitation has been clearly observed in high melting temperature liquid metals using electromagnetic forces. These forces do not have to be extreme, and a magnetic field of 3 T (0.3 MPa oscillating pressure amplitude) is sufficient to observe changes in the microstructure in some cases. However, the onset of the cavitation does not guarantee an improvement in particle dispersion, both ex situ or in situ, even with a high intensity cavitation treatment.

Oxides show a low degree of admixing due to bad wettability, and, thus, weak dispersion. High intensity cavitation, in general, does not affect the microstructure and the refinement of the added phase when the reinforcements *survive* the processing, e.g. Al_2O_3 or CeO_2. Therefore,

ex situ oxide composite production is hardly feasible via the liquid route using current processing parameters.

The investigated carbides and borides show a mixed degree of refinement. TiC and TiB_2 can be well dispersed in iron and steel for the purpose of in situ composite production. In this case, cavitation treatment reduces the agglomeration and provides a significant precipitate refinement, most likely, due to enhanced heterogeneous nucleation. However, magnetic field in the order of 3 T has to be applied to reach any clear improvement. All investigated zirconium compounds exhibit a worse dispersion independently of cavitation intensity.

While the current conclusions are not fully positive, the concept of cavitating ferrous liquids for the reinforcing phase dispersion is not fully investigated and has a strong potential. Already some easily implementable solutions have been identified for ex situ approach. Firstly, the results could be improved if a higher mixing rate is achieved. With a static magnetic field, it is difficult due to flow braking, however, the magnetic field does not have to be applied permanently. A periodic or impulse-type cavitation could provide the benefits of both, the high velocity turbulent flow regime to pull in and homogenize the particles and high impact cavitation events to disperse them. Secondly, the well-wetted yet reactive reinforcements, e.g. TiC, could also be incorporated ex situ 1) by adding dissolution limiting elements and 2) using processing times shorter than the typical reaction time.

As the overall results of the investigated systems cannot be generalized to every ferrous alloy, it is clear that material compatibility studies with different steel grades as well as iron compositions with different wettability improving alloying elements must be pursued and are recommended for further research. This will allow to identify the most prospective ceramic-metal pairs both for in situ and ex situ processing.

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