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Dissimilar friction stir welding of aluminum alloys reinforced with carbon nanotubes

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Abstract:
This chapter is devoted to studying the possibility of incorporating carbon nanotubes (CNTs) as reinforcing fillers in dissimilar metal matrices joints produced by friction stir welding (FSW), as well as the impact of this incorporation on the microstructural and mechanical properties of these joints. Carbon nanotubes are extensively used as a reinforcing material in nanocomposites, due to their high stiffness and strength. FSW is a solid-state welding process of joining aluminum and other metallic alloys and has been employed in the aerospace, rail, automotive, and marine industries. Recently, friction stir processing (FSP), a derivative method of FSW, has been employed as an alternative for the production of metal matrix composites (MMCs). In this work, the process parameters were optimized in order to achieve nondefective welds, with and without the addition of CNTs. Two main cases were studied: (1) FSP was optimized by changing the tool rotational and travel speed as well as the number and direction of FSW passes, and (2) a Taguchi design scheme was adopted to further investigate the FSP in relevance to three factors (number, direction of passes, and tool rotational speed). Mechanical behavior was studied, and the local mechanical properties of the produced MMCs were compared with their bulk counterparts and parent materials. More specifically, the measured mechanical properties in the micro- and nanoscale (namely hardness and elastic modulus) are correlated with the microstructure and the presence of fillers.

DOI: 10.1515/psr-2015-0010

1 Introduction

Aluminum alloys play a major role in today's manufacturing processes due to their properties, such as good corrosion and wear resistance as well as their high strength-to-weight ratio. Nevertheless, aluminum alloys tend to show low weldability while using conventional welding methods, and as a result severe adhesive wear may be observed. This drawback can be overcome by the incorporation of ceramic particles into the metal matrix. Most of the methods for producing metal matrix composites (MMCs) reported in literature, [1–4] require the melting of the metal matrix and hence result in solidification-related drawbacks. In order to solve these issues friction stir processing (FSP), a derivative method of friction stir welding (FSW), can be applied.

FSW is a solid-state welding method that was invented at The Welding Institute (TWI) of the UK in 1991 [5]. Welding takes place as a nonconsumable rotating tool travels down the joint line of metal plates (see Figure 1). The tool consists of a specially designed pin and an appropriate shoulder, which plays a major role in the heat input, and subsequently in the quality, of the welds and their mechanical properties (see Figure 1). This technique eliminates most of the drawbacks of fusion welding such as porosity, inclusions, and solidification cracks, as no melting of the material occurs.
Figure 1: Schematic drawing of friction stir welding.

Friction stir welding can also be used to weld dissimilar alloys. Jonckheere et al. [7] studied the welding of AA2014-T6 with AA6061-T6 and observed that the temperature is relative to the amount of each alloy in the weld nugget. Amancio-Filho et al. [8] studied the mechanical properties of the welding between AA2024-T351 and AA6064-T4, concluding that the welds fail in the area which is annealed by the heat generated by the FSW tool, while Steuwer et al. [9] focused on the residual stresses of the welds of AA5083 with AA6082. Furthermore, Tanaka et al. [10] studied the welding of mild steel with AA7075-T6 and the strength of the weld in relation to the intermetallic compounds’ thickness and welding parameters. Regardless of the formation of layers thinner than those of the resolution of FE-SEM IMC, the specimens failed in the AA7075/mild steel weld nugget. Murr et al. [11] studied the welding of AA1100 with AA6061, reporting that the weld zones’ microstructure includes some degree of dynamic recrystallization, while in the welding of AA6061 with Cu the dissimilar aluminum/copper system manifested a more complex microstructure. Further work by Murr et al. was conducted on intercalation vortices and related microstructural features in the friction stir welding of dissimilar metals, such as AA2024 with AA6061 and AA6061 with Cu, at various rotational and transverse speeds [12]. They observed that the mixing or vortex-like intercalations at the aluminum/copper weld zone is a manifestation of the solid-state flow, facilitated by dynamic recrystallization in the welds. Also, Cavaliere et al. studied the welding of AA2024 with AA7075, and both the grain structure and precipitates’ distribution differences originated by the process were investigated in depth via optical microscopy [13].

Friction stir processing is a promising technique for producing MMCs, and it is mostly used to modify and improve the microstructure, mainly near the surface (see Figure 2). By incorporating the appropriate micro or nanoparticles as fillers into the matrices, enhancement of specific material properties, especially of those that depend on the surface microstructure, is achieved. These particles are introduced into the material’s near-surface layers, resulting in localized microstructural modification. As a consequence, FSP technique is emerging to be a very effective solid-state processing technique that can provide localized modification, as well as control of the material microstructure, in the near-surface layers of the processed metallic component. Potential applications of aluminum alloys reinforced with several nanoparticles may include (but are not limited to): high performance structural components requiring high durability, automotive engines and high speed machinery parts requiring high wear and thermal resistance, armor and protection components with the ability to absorb high energy impacts and thermal shocks, aeronautic and aerospace components requiring functionally graded material properties, as well as electronic and optical instruments.

Figure 2: Schematic drawing of (a) FSP cover pass using pin-less tool (b) FSP pass.

Some of the advantages of FSP and FSW are listed below:

a. FSP and FSW are green processes, as the heat generation originates from friction and plastic deformation, in the absence of toxic fumes, radiation and noise, characteristics that classify FSP as a green and energy-saving technique.

b. FSP and FSW are short-route, solid-state processing techniques with one step processing that achieve microstructural refinement, densification, and homogeneity.

c. The microstructure and mechanical properties of the processed zone can be accurately controlled by optimizing the FSP tool design, FSP parameters and the application of active cooling/heating.

d. The depth of the processed zone can be optionally adjusted by changing the length of the tool pin, with the depth ranging from several hundred micrometers to tens of millimeters; it is difficult to achieve a manually adjusted processed depth using other metalworking techniques.

e. FSP and FSW are versatile techniques with a comprehensive function for the fabrication, processing, and synthesis of materials.

f. FSP and FSW do not alter the shape and size of the processed components.
In comparison to unreinforced aluminum alloys, aluminum matrix composites reinforced with ceramic particles exhibit higher strength and stiffness, improved tribological characteristics, as well as increased creep and fatigue strength [14–19]. Metal matrix composite materials are mainly being produced using aluminum alloy substrates reinforced with SiC [20] and Al₂O₃ particles [21], as well as copper substrates reinforced with particles of boron carbide [22]. Furthermore, surface hybrid nanocomposites have been produced and studied; results show improved hardness and mechanical properties when using aluminum alloys substrate and reinforcement of graphite/Al₂O₃ [23]. Carbon nanotubes have been used as reinforcement in the metal matrix as well. Liu et al. applied FSP on AA1016, which had surface holes filled with multiwalled carbon nanotubes [24]. The resulted MMC had superior mechanical properties when compared to the corresponding base metal. Lim et al. produced an AA7075 matrix multi-walled carbon nanotubes (MWCNT) reinforced composite via single pass FSP using a thin AA6111 plate as a cover for the MWCNT filled groove [25]. The distribution of nanoparticles in the metal matrix was inhomogeneous, with multiple FSP passes being a potential solution to that issue. Sun et al. applied FSW with the addition of particles (before friction stir welding, SiC particles were incorporated between two copper plates) [26]; this resulted in the improvement of the mechanical properties of the Cu joints. The same technique was performed by Byung-Wook et al. [27] and Bahrami et al. [28] who improved the mechanical properties of AA5083 and AA7075 joints, respectively, using SiC particles as a reinforcing material. Bahrami et al. [29] also studied the influence of the tool pin design on the distribution of the SiC powder in the weld nugget of AA7075 and the resulting mechanical properties. Although the threaded tapered tool pin resulted in the lowest average microhardness value, the most uniform particle distribution was achieved. Recently, Pantelis et al. [30] incorporated SiC nanoparticles in the weld nugget of dissimilar Friction Stir Welds of AA5083-H111 with AA6082-T6. The results showed improvement of the mechanical properties of the welds as compared to the their reinforcement-lacking counterparts without nanoparticle reinforcement.

2 Experimental procedure

Rolled plates of AA5083-H111 and AA6082-T6 with dimensions of 200 × 100 × 3 mm³ were used as parent materials. Their chemical composition is shown in Table 1. The CNTs used as reinforcing material were produced via chemical vapor deposition (CVD). They had an average diameter of 50 nm and length over 10 μm as presented in Figure 3.

<table>
<thead>
<tr>
<th>Table 1</th>
<th>Chemical composition of 5083-H111 and 6082-T6 Al alloys (wt.%).</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Si</td>
</tr>
<tr>
<td>5083-H111</td>
<td>0.37</td>
</tr>
<tr>
<td>6082-T6</td>
<td>1.07</td>
</tr>
</tbody>
</table>

Figure 3: SEM micrograph of the CNTs.

The machine used in the procedure was a custom-made friction stir welding machine of the Shipbuilding Technology Laboratory of the School of Naval Architecture and Marine Engineering of the National Technical University of Athens. The FSW tool was made of heat treated steel “SVERKER 21” (at 61 HRC) and consisted of a flat shoulder with a diameter of 23 mm and a cylindrical left-handed threaded pin with a diameter of 6 mm and height of 2.8 mm.
During the experiments the welding was conducted parallel to the rolling direction of the plates. The nanoparticles were incorporated into half grooves machined at the joining faces of the plates. Figure 4 depicts the groove with dimensions of 180 mm length, 1 mm width and 2.5 mm depth that was created when the plates were rigidly connected on the clamping arrangement. The CNTs after being mixed with ethanol to become more manageable were pressed tightly into the groove. The central line of the rotating pin was aligned along with the groove.

![Figure 4: Geometric characteristics of the machined grooves.](image)

After friction stir welding was completed, the specimens were cut at a transverse section in regard to the welding direction, and buffing and polishing was conducted. In order to observe the specimens’ microstructure, they were etched with “modified Poulton’s reagent” and macroscopically observed using an optical stereoscope (Leica MZ6). The specimens with the best particle distribution were then selected and observed using an optical microscope (Leica DMILM) and a scanning electron microscope (Jeol JSM 6390LV) equipped with the energy dispersive spectroscopy system Oxford Inca Energy 250. The software “Leica Application Suite” was used in order to measure the grain size of the optimum specimen.

A Wolpert Wilson 402MVD microhardness tester was utilized in order to measure the longitudinal Vickers microhardness distributions of the welds. The load applied was 300 g. The average longitudinal microhardness measurements were made along two lines residing 1 mm and 2 mm under the surface.

Tensile specimens were machined perpendicular to the welding direction according to the ASTM E 8M-04 standard. An MTS hydraulic mechanical testing machine of 100 kN maximum load was employed in order to conduct the tensile tests. The elongation was measured with an Epsilon ±25 mm extensometer at a deformation speed of 0.5 mm/min.

Nanoindentation testing was performed using a nanomechanical test instrument, which allows the application of loads ranging from 1 to 30,000 μN and records the displacement as a function of applied loads with high load (1 nN) and displacement resolutions (0.04 nm). The nanomechanical test instrument employed in this study is equipped with a scanning probe microscope (SPM), in which a sharp probe tip moves in a raster scan pattern across the sample surface, using a three-axis piezo positioner. In all nanoindentation tests, a total of 10 indents are averaged to determine nanomechanical values for statistical purposes, with a spacing of 50 μm, in a clean area environment with 45 % humidity and 23 °C ambient temperature. In order to operate under closed loop displacement control (DC), feedback control option was used. The nanoindentation tests were conducted with a Berkovich (three-side pyramid) diamond indenter (average radius of 100 nm) with 40 s loading and unloading segment time separately and 3 s of holding time at various penetration depths. All nanoindentation measurements were performed 2 mm under the welding substrate, longitudinally and in transverse directions (in the middle of the weld nugget), at the cross-section of the welds. Prior to indentation, the area function of the indenter tip was calibrated in fused silica, a standard calibration material [31]. The surface of the samples prior to conducting the nanoindentation measurements, was prepared by grinding and polishing.

3 Results and discussion

To the best of the authors’ knowledge, there has been no study related to the FSW of dissimilar aluminum alloys with the addition of CNTs. In this work, the optimization of welding of the dissimilar aluminum alloys AA5083-H111 and AA6082-T6 with incorporation of CNTs was studied by changing the tool rotational and travel speed as well as the number and the direction of FSW passes, which affect the distribution of the nanoparticles in the weld nugget. Also, the present work focuses on the mechanical properties of the optimum joint by tensile and indentation methods. The results were then compared to an identical joint without reinforcement. Nanoindentation was used to elucidate the effect of reinforcing fillers on the mechanical properties taking into account the microstructure. Two series of experiments were conducted. The choice of parameters for the first experiment lies on the authors’ experience from past experiments with other nanofillers incorporated into metal matrices, whereas the parameters of the second one were determined by Taguchi analysis. Both of them are analysed in the following paragraphs.
3.1 First series of experiments

First, the optimum parameters of welding AA5083-H111 to AA6082-T6 without carbon nanotube addition were determined by altering the rotational speed from 600 to 1180 RPM, the traverse speed from 60 to 85 mm/min and the tool tilt angle from 0 to 4°. These parameters are presented in Table 2. It should be mentioned that the resulting specimen was also used for comparison purposes.

<table>
<thead>
<tr>
<th>Passes (mm)</th>
<th>Rotational speed (RPM)</th>
<th>Traverse speed (mm/min)</th>
<th>Shoulder diameter (mm)</th>
<th>Tool pin height (mm)</th>
<th>Tool Pin diameter (mm)</th>
<th>Tool pin shape</th>
<th>Tilt angle</th>
<th>Tool pin penetration Depth (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>750</td>
<td>85</td>
<td>23</td>
<td>3</td>
<td>6</td>
<td>cylindrical</td>
<td>3°</td>
<td>2.50</td>
</tr>
</tbody>
</table>

An FSW test with CNT addition was conducted adopting the above parameters (test 5 on Table 3), but it presented defects such as porosity and severe CNT agglomeration. According to literature [32], a more uniform nanoparticle distribution in the weld nugget can be achieved by increasing the number of FSP passes. Also, various studies conducted in the Shipbuilding Technology Laboratory regarding friction stir processing, indicated that alternating the direction of the FSP passes as well as increasing the rotational speed of the tool can have a positive impact on the distribution of the nanoparticles. Based on this feedback, the authors concluded in the conditions presented in Table 3 for the various friction stir tests that they conducted. The same tool was used in all experiments, namely one with a cylindrical left-hand threaded pin. This was because the material flow created by this specific pin, results in very good material mixing. The transverse speed, tool tilt angle and tool pin penetration depth were also kept constant at 85 mm/min, 3°, and 2.50 mm respectively, because alternating the rotational speed is sufficient a variable in order to change the weld pitch. The theoretical volume fraction ($V_f$) of the carbon nanotubes in the weld nugget, which is calculated by equation (1) according to literature [22], was 13.8% in every experiment:

$$V_f = \frac{\text{Area of groove}}{\text{Projected area of tool pin}} \times 100$$

Table 3 Welding parameters of AA5083-H111 with AA6082-T6 with nanoparticle reinforcement.

<table>
<thead>
<tr>
<th>Nº of specimens</th>
<th>Number of passes (mm)</th>
<th>Rotational speed (RPM)</th>
<th>Direction of passes</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>3</td>
<td>750</td>
<td>++ +</td>
</tr>
<tr>
<td>2</td>
<td>2</td>
<td>750</td>
<td>+ +</td>
</tr>
<tr>
<td>3</td>
<td>2</td>
<td>1180</td>
<td>+</td>
</tr>
<tr>
<td>4</td>
<td>3</td>
<td>1180</td>
<td>++ +</td>
</tr>
<tr>
<td>5</td>
<td>1</td>
<td>750</td>
<td>+</td>
</tr>
<tr>
<td>6</td>
<td>2</td>
<td>750</td>
<td>++</td>
</tr>
<tr>
<td>7</td>
<td>2</td>
<td>1180</td>
<td>+</td>
</tr>
<tr>
<td>8</td>
<td>1</td>
<td>1180</td>
<td>+</td>
</tr>
<tr>
<td>9</td>
<td>4</td>
<td>750</td>
<td>++ ++</td>
</tr>
</tbody>
</table>

In all the tests the + and − symbols denote that the AA5083-H111 is located at the advancing and the retreating side, respectively. It should also be noted that, for all the experiments, before the welding process the grooves were covered by one FSP pass with a flat pinless tool 23 mm in diameter, using the same conditions of the welding, in order to prevent the CNTs from being ejected out of the groove during FSW.
3.1.1 Macrostructural study

The macrographs of the cross section of all the specimens (except specimen 9) are presented in Figure 5. Specimen 9 presented a massive groove-like defect, and as a result it could be neither buffed nor polished. By observing the micrographs at Figure 5(a) and (d), it is concluded that the application of three passes in the same direction resulted in a better distribution of the particles and exhibited lower particle agglomeration levels than the application of less as well as more passes. It is also observed that the increase of the rotational speed created a more homogeneous distribution and a thinner flow arm. The application of two passes of the opposite direction creates an heterogeneous distribution as there were large particle-rich and particle-free areas (specimens 2 and 3 presented at Figure 5(b) and (c), respectively). Furthermore intense agglomeration is observed, mainly in the advancing side. The presence of formed agglomerates near the heat affected zone after the application of one and two passes (specimens 2, 5, 6, 7, 8), can mainly be attributed to the left-handed screw pin, which directed the material downwards at the center and then upwards near the heat affected zone [33]. In fact, the application of the third pass of the same direction (specimen 4 – Figure 5(d)) improves the distribution, in comparison to the two-pass experiments, as only a small area consisting of agglomerated particles can be observed in the upper area of the retreating side. The weld nugget of specimen 4 is the best formed weld nugget compared to all the other specimens.

![Figure 5: Optical stereoscopy macrographs of the cross sections of the specimens after etching. Test: (a) 1, (b) 2, (c) 3, (d) 4, (e) 5, (f) 6, (g) 7, (h) 8.](image)

After the observation of the macrographs for all the specimens, it is concluded that specimen 4 presents the most uniform macrostructural morphology, and taking into account the absence of defects, it comprises the optimum weld with nanoparticles; therefore it was further studied, as shown in the next paragraphs.

In Figure 6 the macrostructure of the weld nugget of the specimen without reinforcement is presented for comparison purposes. The absence of defects, a large flow arm, and an onion ring structure can be observed. The microstructure of the weld nugget is presented in Figure 6(b). Good material mixing is observed in both micrographs and the weld nugget (WN) microstructure characterized by small and equiaxial grains with a grain size of $\sim 13 \, \mu m$.

The reduction of the grain size of the weld without reinforcement in the WN, compared to that of the untreated base metals (5083: 26 $\mu m$ and 6082: 40 $\mu m$), is a result of the dynamic recrystallization which, according to the literature [34, 35], took place during FSW.

By comparing Figure 5(d) and Figure 6(a) it can be concluded that the material mixing appears to be better in Figure 5(d) (note that the carbon nanotubes appear as dark areas under the optical microscope). That is expected because of the higher number of passes.
3.1.2 Microstructural study

In Figure 7 areas of the weld nugget of specimen 4 are observed by means of optical microscopy. In Figure 7(a) the lower half of the weld nugget, which is poor in CNTs, is depicted. It is observed that both the AA6082 and AA5083 are mechanically mixed into the metal matrix. In Figure 7(b) and (c) the upper half of the weld nugget, which is rich in carbon nanotubes, is observed. It is also observed that the CNTs are arranged in bands into the aforementioned metal matrix. Also, some agglomeration can be noted, depicted with arrows in Figure 7(b), (d) and will be further analysed with the assistance of scanning electron microscopy and energy dispersive spectroscopy. In Figure 7(d) the CNT bands (dark areas) are shown in higher magnification.

Finally, the weld nugget consisted of equiaxial grains with an average grain size of \( \sim 6 \) \( \mu \)m. The smaller grain size that is observed in this specimen in comparison to the one of the specimen without CNTs is, according to the literature, a result of the pinning effect [27, 28] that CNTs have had in this region, thus impeding grain growth.

In Figure 8 secondary electron SEM images of areas of the weld nugget are presented. In Figure 8(a) the particle poor region of the lower half of the weld nugget is presented. The mixing of the two aluminum alloys is without pores or defects. In Figure 8(b) the aforementioned CNT bands are presented. In Figure 8(c) and (d) clusters formed by agglomerated CNTs are observed.
In Figure 8, secondary electron SEM micrographs of several areas of the weld nugget of specimen 4: (a) 1000× magnification; (b) 3000× magnification, (c) 6500× magnification; (d) 20000× magnification.

In Figure 9, a backscatter electron SEM image of the CNT bands of Figure 8(b) as well as the EDS graphs of the two distinctive areas (CNTs and metal matrix) are presented. In the metal matrix, no carbon content was measured, thus confirming the arrangements of CNTs in specific bands. The measured magnesium is an alloying element of both the AA6082 and AA5083.

In Figure 10, a backscatter electron SEM image of the CNT cluster of Figure 8(c) as well as the EDS graphs of the distinctive areas (CNTs and metal matrix) are presented. The agglomeration of CNTs in specific areas that was observed by the optical and secondary electron SEM images is supported by the fact that a high amount of carbon is measured in the aforementioned cluster.
It should be mentioned that the chemical composition of the elements arising from the EDS analysis is not listed, because carbon is a low atomic weight element and the results present a large rate of error.

### 3.1.3 Microhardness study

The microhardness mean average of the base metal AA6082-T6 was $\sim 105$ HV0.3, while this of the AA5083-H111 was $\sim 80$ HV0.3 (used as reference values). As observed in Figure 11, the microhardness of the optimum weld with nanoparticles was higher compared to this of AA5083-H111, for two different distances from the surface. Nevertheless lower microhardness values were observed at 1 mm from the surface, especially in the retreating side, because of the flow arm of the specimen which had larger grains than the weld nugget and was not as rich in particles. In the other welding zones (thermo-mechanically affected zone, heat affected zone) the microhardness distribution was almost the same at all distances from the shoulder surface with the exception of the 6082 HAZ, which extends further as we get closer to the surface due to the annealing effect that takes place.

As shown in Figure 12, the microhardness distribution of the weld without the addition of nanoparticles at a distance of 2 mm from the surface presented lower values (an average 70 HV0.3) in the weld nugget compared to these of the specimen with CNTs at the same distance from the surface. This is a consequence of the fact that, according to the Hall–Petch relation, the hardness increases as grain size decreases [28], and in fact it was observed that the composite weld nugget consisted of smaller grains than the weld nugget without reinforcement. The lowest values were observed in the heat affected zone, at the side of AA6082-T6 ($\sim 54$ HV0.3) due to the dissolution of the strengthening precipitates, as referred in the literature [36].
3.1.4 Tensile tests

The mechanical properties of the created welds are determined via several metallurgical mechanisms (dissolution and precipitation of secondary phases, dynamic recrystallization) as well as the material flow during FSP. Three tensile specimens were created using the welding conditions of specimen 4. The representative stress-strain curves for that specimen, as well as for the one without reinforcing particle addition, are presented in Figure 13. Table 4 shows the average values for the elastic modulus, yield stress, ultimate tensile strength (UTS), and percentage of elongation. The values of all the magnitudes dropped with nanoparticle addition. That is because all the specimens failed in the weld nugget, probably due to the fact that the CNTs get tangled during the stirring action of the tool resulting in severe agglomeration, as confirmed by the SEM micrographs and the EDS analysis. On the other hand, the unreinforced weld failed in the HAZ. Failure took place exactly at the location of the minimum hardness value that is in the HAZ of 6082 aluminum alloy.

![Figure 12: Microhardness distribution (2 mm under the surface) of the welds, with and without CNTs.](image)

![Figure 13: Stress-strain curves for the welds with and without CNT reinforcement.](image)

<table>
<thead>
<tr>
<th>Specimen</th>
<th>Elastic modulus (GPa)</th>
<th>Yield stress (MPa)</th>
<th>UTS (MPa)</th>
<th>Elongation (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>with CNTs</td>
<td>70 ± 0.05</td>
<td>128 ± 2.8</td>
<td>155 ± 2</td>
<td>0.95 ± 0.1</td>
</tr>
<tr>
<td>without CNTs</td>
<td>65 ± 0.1</td>
<td>140 ± 3.6</td>
<td>193 ± 2</td>
<td>3 ± 0.05</td>
</tr>
</tbody>
</table>
3.1.5 Fractography

The fracture surface of the weld of specimen 4 is presented in Figure 14. It is situated at the WN. Although an overall brittle fracture morphology can be observed (Figure 14(a,b,d)), there are some small areas that are characterized by the presence of small dimples, indicative of ductile fracture (Figure 14(c)). The large dark colored cavities that are presented in Figure 14(a) and (b) may be CNT clusters. These may act as sites that trigger the fracture.

![Figure 14: Backscatter electron SEM micrographs of the fracture plane at the WN of the specimen 4 (c, d) Further magnification of the areas shown with arrows.](image)

3.1.6 Mechanical properties measured by nanoindentation Basic theory

Nanoindentation tests involve the contact of an indenter with a material surface and its penetration due to a specified load or depth. According to the Oliver and Pharr (O&P) method, the nanoindentation hardness (nanohardness \( H_{\text{nano}} \) hereafter) can be determined as a function of the final penetration depth of indent by \[37\]:

\[
H = \frac{P_{\text{max}}}{A(h_c)}.
\]

where \( P_{\text{max}} \) is the maximum applied load measured at the maximum depth of penetration \( h_{\text{max}} \), \( A(h_c) \) is the projected contact area between the indenter and the specimen. For a Berkovich indenter \( A(h_c) \) can be expressed as a function of the contact indentation depth \( h_c \) as

\[
A(h_c) = 24.54^2 + a_1 h_c + a_2 h_c^{1/2} + \ldots + a_{16} h_c^{1/16}.
\]

The contact indentation depth, \( h_c \), can be determined from the following equation:

\[
h_c = h_{\text{max}} - \varepsilon \frac{P_{\text{max}}}{S}.
\]

where \( \varepsilon \) is an indenter geometry constant equal to 0.75, for Berkovich indenter, \( S \) is the contact stiffness which can be determined as the initial slope of the unloading curve at the maximum penetration depth, i.e.

\[
S = \left( \frac{dP}{dh} \right)_{h = h_{\text{max}}},
\]
Based on Sneddon’s [38] elastic contact theory, the reduced elastic modulus, \( E_r \), is given by the following expression:

\[
E_r = \frac{S}{2\beta} \sqrt{\frac{\pi}{A(h_c)}}
\]

where \( \beta \) is a constant that depends on the geometry of the indenter. For the Berkovich indenter, \( \beta \) is equal to 1.167 [38, 39]. The Elastic modulus of the specimen, \( E \), can be calculated as follows:

\[
\frac{1}{E_r} = \frac{1 - \nu^2}{E} + \frac{1 - \nu_i^2}{E_i}
\]

where \( \nu, E \) are the Poisson ratio and the elastic modulus of the specimen, respectively, and \( \nu_i, E_i \) are the Poisson ration and elastic modulus of the indenter, respectively (for a diamond indenter, \( E_i \) is 1140 GPa and \( \nu_i \) is 0.07).

**Nanohardness and elastic modulus distribution**

Nanoindentation tests were performed in order to study the deformation mechanism of the composite (AA5083-H111/AA6082-T6 with CNTs) in the weld nugget and to measure the elastic modulus and nanohardness values of the different weld areas. Measurements were performed across the weld zone 2 mm under the shoulder surface and perpendicular to the middle of the weld nugget, at the cross-section of the welds.

The mean average of nanohardness (\( H_{\text{nano}} \)) distribution across the weld was calculated to be lower than that of the parent materials, mainly at the side of AA6082-T6 parent material. Decreased values were observed in the HAZ, at the side where AA6082-T6 was located, due to the dissolution of the strengthening precipitates, as discussed in the literature [36]. However, the values were found to increase in the WN because of the presence of CNTs, but they were still found to be lower than, if not equal to, the nanohardness of AA5083-H111 parent material (Figure 15).

The overall micro- and nanohardness distributions (Figure 11 and Figure 15) across the weld showed almost the same trend: hardness reduction from the BM, starting from the HAZ, reaching a local minimum at the HAZ from the side of AA6082-T6 and a rise to higher values in the SZ. However, comparing the two distributions (Figure 11 and Figure 15) in the weld nugget, the hardness results were significantly different in the weld nugget, taking into account the hardness values of parent materials as reference points. More precisely, the microhardness values were found to be slightly higher (8 %) than the microhardness values of the AA5083-H111 parent material, whereas the nanohardness values were found to be lower than or equal to the nanohardness values of the AA5083-H111 parent material.

**Figure 15:** Nanohardness distribution of the weld with the addition of CNTs obtained from nanoindentation experiments at 400 nm maximum penetration depth (2 mm under the surface).

This divergent behavior can be attributed to the size of the probed volume during indentation. During microhardness testing, the volume of the deformed plastic region is sufficiently large (the diameter of the residual impression is \( \sim 85 \mu m \)), as compared to the grain size, and the deformation is influenced by both the grain
boundaries and the presence of reinforcing particles. On the other hand, the plastic zone during nanoindentation, which is related to the nanohardness values, is much smaller (the diameter of the residual impression is \( \sim 1.5 \mu m \)). Details and discussion of the volumes of plastic and elastic deformation under contact indentation loading have already been presented elsewhere [40]. In order to reveal the reinforcing mechanisms and the testing method’s dependence on the hardness values, it is necessary to establish the relations between the micro- and nanohardness for both particle reinforced and unreinforced joints. Further investigation should be conducted in order to elucidate this phenomenon, which is beyond the scope of this chapter.

Also, it is critical to explain that the probed volume for the elastically deformed region is not the same as that of the plastically deformed region. The size of the elastically deformed region is larger than that of the plastically deformed region. This is also supported from the results presented in Figure 16 and discussed below. It is depicted in Figure 16 that the welding process results in lower elastic modulus values in the side of 6082 HAZ. However, the values were found to increase in the WN because of the presence of CNTs. The mean average elastic modulus, measured with tensile tests, of both base metals was 71.7 GPa. The reference lines illustrated in Figure 16 correspond to the elastic moduli, measured by tensile tests, of the specimens with and without CNTs. It is evidenced that the addition of CNTs results in higher elastic modulus (the elastic modulus increased up to 15%) in the SZ, equal in the advancing side and lower in the HAZ from the retreating side, compared to the elastic moduli of the parent materials.

![Figure 16: Elastic modulus distribution of the weld with the addition of CNTs obtained from nanoindentation experiments at 400 nm maximum penetration depth (2 mm under the surface).](image)

3.2 Second series of experiments

All the specimens of the first series of experiments failed in the weld nugget, suggesting that the chosen conditions could possibly be improved. As a result the authors proceeded to use the Taguchi method in order to further optimize the welding parameters of the first series of experiments.

The most significant process parameters of FSP are the following:

1. rotational speed of the tool (rpm);
2. welding speed or transverse speed (mm/min);
3. number of processing passes;
4. tool geometry: (a) pin profile, (b) tool shoulder diameter, \( D \) (mm), (c) pin diameter, \( d \) (mm), (d) \( D/d \) ratio of tool, (e) pin length (mm), (f) tool inclination angle (°).

Taguchi addresses quality in two main areas: offline and online quality control. The most important difference between a classical experimental design and a Taguchi-method-based robust design technique is that the former tends to focus solely on the aspect of the quality characteristic while the latter takes into consideration the minimization of the variance of the characteristic of interest. Although the Taguchi method has drawn much criticism due to several major limitations, it has been able to effectively solve single response problems.

In the second series of experiments the same FSW parameters as in the first series were altered, namely the rotational speed, number of passes, and direction of passes whereas the transverse speed, tool tilt angle, and...
tool pin penetration depth were kept constant at 85 mm/min, 3 °, and 2.50 mm, respectively. The theoretical volume fraction of the carbon nanotubes in the weld nugget was also 13.8% in every experiment. In total, 8 FSW joints of AA5083-H111 with AA6082-T6 were produced following the Taguchi scheme presented in Table 5. Throughout the Taguchi statistical analysis, only 24 (8x3) experiments for L8 orthogonal arrays are needed for microstructural observation and mechanical tests.

Table 5 Welding parameters of AA5083-H111 with AA6082-T6 with nanoparticle reinforcement.

<table>
<thead>
<tr>
<th>No. of specimen</th>
<th>Number of passes (mm)</th>
<th>Rotational speed (RPM)</th>
<th>Direction of passes</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1</td>
<td>1500</td>
<td>+</td>
</tr>
<tr>
<td>2</td>
<td>2</td>
<td>1500</td>
<td>+ +</td>
</tr>
<tr>
<td>3</td>
<td>3</td>
<td>750</td>
<td>+ + +</td>
</tr>
<tr>
<td>4</td>
<td>3</td>
<td>1180</td>
<td>+ + +</td>
</tr>
<tr>
<td>5</td>
<td>3</td>
<td>1500</td>
<td>+ + + +</td>
</tr>
<tr>
<td>6</td>
<td>3</td>
<td>1500</td>
<td>+ + + +</td>
</tr>
<tr>
<td>7</td>
<td>3</td>
<td>1180</td>
<td>+ + + +</td>
</tr>
<tr>
<td>8</td>
<td>2</td>
<td>1500</td>
<td>+ + + +</td>
</tr>
</tbody>
</table>

In all the tests the + and − symbols denote that the AA5083-H111 is located at the advancing and the retreating sides, respectively. It should also be noted that, as in the first series of experiments, before the welding process the grooves were also covered by one FSP pass with a flat pinless tool 23 mm in diameter in order to prevent the CNT nanoparticles from being ejected out of the groove during FSW, using the same conditions of welding.

3.2.1 Macrostructural study

In Figure 17 the optical macrographs of the Taguchi optimized experiments are presented. All the specimens present a large flow arm that appears light colored and is therefore characterized by a low CNT concentration. Also, by observing the micrographs it is concluded that increasing the machine’s rotational speed to 1500 RPM improves the nanoparticle distribution in the weld nugget. Despite the second specimen’s large flow arm, it presents the least agglomeration and appears to have the best microstructure of all the specimens, presenting the typical “onion ring” structure of friction stir welding. This will be further examined by means of optical and electronic microscopy, microhardness measurement, and tensile testing.

Figure 17: Optical stereoscopy macrographs of the specimens after etching. Test: (a) 1, (b) 2, (c) 3, (d) 4, (e) 5, (f) 6, (g) 7, (h) 8.
3.2.2 Microstructural study

In Figure 18 the microstructure of several areas of the weld nugget of specimen 2 can be observed by means of optical microscopy. The left side, right side, and center of the weld nugget of Figure 17(b) are presented in Figure 18(a,b), and (c), respectively. The weld nugget appears to be rich in CNTs and presents the typical onion ring structure, consisting of alternating bands with different amounts of nanotubes. That can also be observed in higher magnification in Figure 18(d).

![Figure 18: Optical micrographs of several areas of the weld nugget of the specimen 2: (a) the left side of the wn, 100× magnification; (b) the right side of the wn, 100× magnification; (c) the center of the weld nugget, 100× magnification; (d) alternating bands, 1000× magnification.](image)

In Figure 19(a) and (b), secondary electron SEM images of the center of the weld nugget are presented. The mixing of the two aluminum alloys is without pores or defects. The CNT agglomeration observed in the first series of experiments is absent, and the CNTs are indistinguishable in the metal matrix as a result of the homogeneous distribution that was achieved.

![Figure 19: Secondary electron SEM micrographs of (a) the center of the weld nugget, 1000× magnification, (b) the center of the weld nugget, 3500× magnification.](image)

3.2.3 Microhardness study

In Figure 20 the longitudinal microhardness distribution of specimen 2 at two different distances, 1 and 2 mm from the tool surface, is presented. The microhardness values in the weld nugget are lower in the first case because of the presence of the flow arm.
In the first set of experiments, they are also lower, ranging from 60 HV0.3 to 80 HV0.3, compared to the equivalent of specimen 4 of the first series of experiments, ranging from 70 HV0.3 to 90 HV0.3 (see Figure 11), but they present a flatter graph with fewer low and high peaks. This is due to the more homogeneous nanotube distribution in the weld nugget, which can also be characterized by the absence of agglomeration, as mentioned in Section 3.2.2. In the other welding zones (TMAZ, HAZ), the microhardness distribution is in correlation with the corresponding ones of the first set of experiments.

In Figure 21 the longitudinal microhardness distribution of specimen 2 is compared with the equivalent of the specimen without CNTs, both taken at 2 mm from the tool surface. The values of the reinforced specimen are slightly higher than the equivalent of the unreinforced one.

3.2.4 Tensile tests

As can be observed from Table 6 and Figure 22, the average values for the ultimate tensile strength (UTS) and percentage of elongation are improved in comparison to the equivalent values of the first set of experiments (see Table 4 and Figure 13). Despite that, they are still inferior compared to the equivalent of the unreinforced specimen, which failed in the heat affected zone. It should be noted that the reinforced specimens failed at the weld nugget as it can be observed in Section 3.2.5 as well. On the other hand, the values of the elastic modulus and the yield stress of the reinforced specimens are very close, and within the error limits, to the equivalent of the unreinforced ones. These results demonstrate that the carbon nanotube addition has a negative impact mainly on the plastic (and not in the elastic) behavior of the material. Further investigation should be conducted regarding this phenomenon.
### Table 6 Mechanical properties (average values of three tests for each case) of the welds, with and without the addition of CNTs.

<table>
<thead>
<tr>
<th>Specimen</th>
<th>Elastic modulus (GPa)</th>
<th>Yield stress (MPa)</th>
<th>UTS (MPa)</th>
<th>Elongation (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>With CNTs</td>
<td>70 ± 0.1</td>
<td>130 ± 6.43</td>
<td>177 ± 12</td>
<td>1.4 ± 0.64</td>
</tr>
<tr>
<td>Without CNTs</td>
<td>65 ± 0.1</td>
<td>140 ± 3.6</td>
<td>193 ± 2</td>
<td>3 ± 0.05</td>
</tr>
</tbody>
</table>

### Figure 22: Stress – strain curves for the welds with and without CNT reinforcement.

#### 3.2.5 Fractography

The fracture surface of the weld of the specimen 2 is presented in Figure 23. It is situated at the WN. Although it presents an overall brittle fracture morphology (Figure 23(a,b,d)), there are some small areas that are characterized by the presence of small dimples, indicating ductile fracture (Figure 23(c)). The brittle fracture morphology in combination to the fact that the specimens fractured at the weld nugget, indicates that the CNT addition lowered the ductility of the material in the weld nugget region.

### Figure 23: (a, b) Backscatter electron SEM micrographs of the fracture plane at the WN of the specimen 2, (c, d) further magnification of the areas shown with arrows.

#### 3.2.6 Mechanical properties measured by nanoindentation

In Figure 24 the longitudinal nanohardness distribution of second specimen at 2 mm under the shoulder surface, at the cross-section of the weld, is presented. Nanohardness values were extracted from nanoindentation
load-unload curves; the experimental set-up and method are presented above in Sections 3.1 and 3.2.5, respectively. The mean average of nanohardness ($H_{\text{nano}}$) distribution across the weld was found lower than this of the parent materials, mainly at the side of AA6082-T6 parent material. Decreased values were observed in the HAZ, at the side where AA6082-T6 was located, due to the dissolution of the strengthening precipitates, as referred in the literature [36]. However, the values were found to increase in the WN because of the presence of CNTs, but were still found to be lower than the nanohardness of AA5083-H111 parent material (Figure 24). Comparing the nanohardness distributions of the first and second sets of experiment (Figure 15 and Figure 24) the same trend is observed. However, in the case of the second set of experiments the microhardness values were found to reach the microhardness values of the AA5083-H111 parent material, in the SZ, whereas the nanohardness values were appear to be lower than the nanohardness value of AA5083-H111 parent material.

Concerning the elastic modulus, local values measured by nanoindentation, are presented in Figure 25. The welding process results in lower elastic modulus values at the side of 6082 HAZ. Comparing the elastic modulus distributions of the first and second set of experiments (Figure 16 and Figure 25) the same trend is observed. In both cases: the values were found to increase in the WN up to 15% compared to the elastic moduli of the parent materials (71.7 GPa) because of the presence of CNTs.

Figure 24: Nanohardness distribution of the weld with the addition of CNTs obtained from nanoindentation experiments at 400 nm maximum penetration depth (2 mm under the surface).

Figure 25: Elastic modulus distribution of the weld with the addition of CNTs obtained from nanoindentation experiments at 400 nm maximum penetration depth (2 mm under the surface).
4 Conclusions

The aim of this work was to study the incorporation of carbon nanotubes in friction stir welding joints of the dissimilar aluminum alloys AA5083-H111 and AA6082-T6, as reinforcing fillers. This was accomplished by changing the tool rotational and travel speed as well as the number and the direction of FSW passes, which mainly affect the distribution of the nanoparticles in the weld nugget. Also, the present work focuses on the evaluation of the mechanical properties of the optimum joint by microhardness measurements as well as tensile and indentation methods.

- The best distribution of CNTs was achieved after the application of two passes of the opposite direction at 1500 RPM rotational speed. This led to the formation of a relatively uniform distribution of CNTs as shown from both optical and scanning electron microscopy.

- The hardness in the weld nugget in micro level was slightly improved (≈7 %), whereas in nano level was slightly lower (≈15 %) in comparison to the weld without reinforcing particles. This divergent behavior can be attributed to the difference between the probed volume during microhardness measurement and nanoindentation.

- From the tensile tests, the elastic modulus and the yield stress of the reinforced specimens are very close, and within the error limits, to the equivalent of the unreinforced ones. Also the elastic modulus of the reinforced specimen (70 GPa) is similar to the equivalent of the parent materials (71.7 GPa for both the 5083-H111 as well as the 6082-T6). From the nanoindentation experiments, the elastic modulus of the weld nugget was found to increase up to 15 % compared to the elastic moduli of the parent materials because of the presence of CNTs. That shows a trend towards slight improvement of the elastic behavior of the stir zone of the material in nano level after the incorporation of CNTs.

- The ultimate tensile strength and the percentage of elongation could not be improved. The reinforced specimens failed in the weld nugget, while the unreinforced ones failed in the heat affected zone. This can be attributed to the geometry of the CNTs. The length of the CNTs (10 μm) is very large compared to their diameters (50 nm). This fact combined with their very high strength and the stirring action of the tool results in the formation of agglomerates. Consequently, the dispersion of CNTs in the metal matrix has a negative impact in the plastic behavior of the material, however this should be further investigated.

Acknowledgment

This research was supported by the EU FP7 Project “Enhancing structural efficiency through novel dissimilar material joining techniques” (SAFEJOINT) under Grant Agreement No. 310498. Financial support for author D. A. Dragatogiannis through a PhD scholarship granted by Research Committee of the National Technical University of Athens (NTUA) is gratefully acknowledged.

This article is also available in: Charitidis, Nanomaterials in Joining. De Gruyter (2015), isbn 978-3-11-033960.

References
