Spatially resolved positron annihilation spectroscopy on friction stir weld induced defects

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Abstract
A friction stir welded (FSW) Al alloy sample was investigated by Doppler broadening spectroscopy (DBS) of the positron annihilation line. The spatially resolved defect distribution showed that the material in the joint zone becomes completely annealed during the welding process at the shoulder of the FSW tool, whereas at the tip, annealing is prevailed by the deterioration of the material due to the tool movement. This might be responsible for the increased probability of cracking in the heat affected zone of friction stir welds. Examination of a material pairing of steel S235 and the Al alloy Silafont36 by coincident Doppler broadening spectroscopy (CDBS) indicates the formation of annealed steel clusters in the Al alloy component of the sample. The clear visibility of Fe in the CDB spectra is explained by the very efficient trapping at the interface between steel cluster and bulk.

Keywords: (coincident) Doppler broadening spectroscopy ((C)DBS), friction stir welding (FSW), defect mapping, positron annihilation spectroscopy

1. Introduction
Friction stir welding (FSW), developed at The Welding Institute (TWI) in UK in 1991 [1, 2], produces the required heat input for joining components by the friction occurring between a rotating tool and the workpiece. The tool consists of a conical steel pin, which dips into the material, and a cylindrical shoulder which only scratches the surface with a depth of less than 0.5 mm. As the materials are only plasticized below the melting point, the weld is formed by mixing the materials due to the rotational movement of the pin, whereas almost all other established welding techniques produce a fusion zone. In this way, FSW has the advantage that problems like pore formation or hot cracking can be avoided, as no high temperature gradients or a liquid phase with the subsequent solidification process occur [1, 3, 4].
In addition, FSW is very flexible concerning the materials to be welded. It is possible to even join different materials with thicknesses up to 75 mm, which is outstanding [2, 3]. In most cases, the mechanical properties exceed those from other welding techniques, or possibly even those of the base material, especially with regard to the ductility [3, 4].

However, during the welding process high pressures along the joint line are induced by the enormous forces of several kN [2, 3], which are needed to press the tool into the material. In accordance with this, previous investigations of FSW by x-ray spectroscopy at the ESRF (European Synchrotron Radiation Facility) in Grenoble demonstrated the high stresses in the joint area [5]. The stress distribution showed a certain asymmetry between the advancing and the retreating side. This was also observed by the insertion of markers into the components in order to reconstruct the flow. The procedure was conducted at the University of South Carolina [6]. Still, there is much research going on in order to characterize the flow and the temperature distribution within the weld.

In this work, FSW was examined by the non-destructive method of (coincident) Doppler broadening spectroscopy ((C)DBS). Several previous experiments showed the feasibility of DBS for the detection of open volume defects.
as for instance, the indirectly probing of the $\gamma/\epsilon$ martensitic phase transformation by detecting the correlated defect concentrations [7]. Furthermore, DBS was used in order to characterize the microstructural evolution of electron beam welded Al alloys as a function of aging conditions [8].

DBS benefits from the high defect sensitivity of the positron diffusing through the solid. The positron preferentially is trapped by the lowered potential at open volume defects, such as atomic vacancies or vacancies elastically bound to dislocations, due to the missing repulsive charge of the core. Finally, the positron annihilates with an electron, with, in general, two 511 keV $\gamma$-quanta being emitted in opposite directions in the center-of-mass system.

However, in the laboratory system, a deviation from 511 keV is observed due to the Doppler shift caused by the initial longitudinal electron momentum. Thereby, the momentum of the thermalized positron can be neglected. Consequently, the slow conduction electrons produce a small Doppler broadening of the 511 keV peak, whereas the tightly bound core electrons result in a large Doppler shift of several keV. Therefore, it is possible to draw conclusions about the electron momentum distribution at the annihilation site by evaluating the shape of the annihilation line. Since the core electrons are missing in open volume defects, the number of positrons annihilating with the low momentum conduction electrons is increased, and thus the 511 keV line appears narrowed compared to that of an unaffected material. Hence, it is possible to gather information about the defect concentration from the shape of the annihilation line. For the characterization of the line shape, the so-called S-parameter is used, which puts the area of a central region in relation to the total area under the peak. The higher the defect concentration in the sample, the larger the value of the S-parameter [9].

In order to obtain information about the chemical vicinity, the annihilation line has to be measured by two Ge-detectors in coincidence, so that the background can be suppressed by several orders of magnitude [10]. Thus, the unique element specific signature becomes visible in the outer regions of the 511 keV peak where the count rate is low. For this reason, CDBS was applied to study low amounts of foreign elements in a base metal. For instance, Nagai et al could identify the formation of Cu- and vacancy clusters in Fe–Cu alloys after irradiation by fast neutrons [11] or in layered metallic samples, a Sn layer buried in an Al matrix could be detected with unprecedented sensitivity [12]. Similar to the results of Nagai et al, the detection of Mg-vacancy complexes with CDBS, after the material was quenched at different temperatures, lead to the assumption that these Mg aggregations are responsible for cracking in resistance spot welded AA5754 alloys [13].

In the present work, two FSW samples have been examined: an Al alloy and a material pairing of steel and an Al alloy. DBS has been used to map the defect distribution in the first sample in order to investigate the microstructure of the weld itself, the heat affected zone (HAZ) and the thermo-mechanical affected zone (TMAZ) [4]. The existence of the TMAZ is unique for FSW: in this region, the material is not only influenced by the heat from the process as it is the case in the HAZ, but is also plastically deformed. However, unlike in the center of the weld, recrystallization does not take place in the TMAZ of Al alloys. In both regions, the HAZ and the TMAZ, the microstructure and the mechanical properties of the material are changed. Thus, the TMAZ/HAZ is known to be the region where fracture preferentially appears. In addition to the examination by DBS, CDBS has been applied to the second sample. The goal was to identify the proportion of steel, which had been stirred into the Al alloy and vice versa.

2. Experiment

2.1. Sample preparation

Both studied samples were prepared at the Institute of Machine Tools and Industrial Management (IWB) at the Technical University of Munich (TUM). For the first sample denoted as Al/Al, a pin with a length of 5.0 mm and a shaft diameter of 5 mm was moved through a single piece of the aluminum alloy AlSi8Cu3. A cross-section with the weld being located in the lower left corner was cut out from the material. The dimensions of the cross-section are given in figure 1, which shows a photograph of the sample. There the red dotted line indicates the shape of the cross-section of the weld. The region with the broader width was caused by the shoulder of the FSW tool which dipped 0.2 mm into the material.

The second sample, denoted as Al/Fe, was a material pairing of common steel S235 and the aluminum alloy Silafont36. In order to joint these two materials, a rather unusual technique had to be applied in order to avoid a strong abrasion of the pin, which is also made of steel: the Al piece has been put on top of the steel and the pin with a length of 4.9 mm and a shank diameter of 5 mm was lowered into the aluminum until its tip reached the steel. In this position, where the pin had only little contact with the steel and the shoulder dipped 0.3 mm into the Al alloy, the feed motion was started. To clarify this process the pin is also shown in figure 2, illustrating the assembly of the sample. The area
that is bordered by the elliptic dashed line is where a mixture of both materials and hence the joint zone is expected. In addition, annealed pure Fe served as a reference material.

2.2. (Coincident) Doppler broadening spectrometer

The following experiments used the CDB spectrometer \[14\] at the high-intensity positron source NEPOMUC (NEutron induced POsitron source MUniCh) \[15\]. An average photon count rate of 32 000 counts s\(^{-1}\) was measured by each Ge-detectors with the beam diameter being set to 1.5 mm. During the measurements, the energy resolution and the stability of the electronic devices were supervised by recording the Be\(^7\) 477.6 keV \(\gamma\)-line. The evaluation of the full-width at half-maximum (FWHM) yielded an energy resolution of 1.32 keV at 477.6 keV.

To reduce surface effects caused, for example, by the polishing of the sample, the maximal positron implantation energy of 30 keV was chosen for all measurements presented in this paper. According to the Makhovian implantation profile, this leads to a maximal positron implantation depth of several \(\mu m\) \[16\]. The measurement time was set to 6 h per CDB spectrum in order to obtain a peak to background ratio better than \(10^5:1\), whereas for the usual DB measurements, each position on the maps was measured for 40 s and in the line-scans for 50 s.

The calibration of the spectrometers’ length units into millimeters, enables the comparison of a certain position on the map with that on the sample. This procedure ensures that the length scales in the graphs presented in the following sections, directly correspond to those of the samples. Thereby, the spatial accuracy was \(\pm 4.7\%\) in \(x\)-direction and \(\pm 8.8\%\) in \(y\)-direction.

3. Measurements and results

In the first series of measurements, we investigated the defect distribution in the Al/Al sample by DBS, followed by the examination of the elemental composition at selected points on the Al/Fe sample by CDBS. The positions of the measurement points on the joint line were chosen by a preceding two dimensional scan of the sample by DBS.

3.1. Al/Al sample

First, the S-parameter was recorded with a step size of 1 mm, in order to obtain the two dimensional map of the defect distribution which is shown in figure 3. By comparing the actual position of the weld on the sample with that of the region with decreased S-parameter, it is clear that the dark blue area in the lower left corner of the figure corresponds to the weld. Thus, at the weld, the defect density is significantly lower, whereas in the zone that is directly attached to the weld, overall, the defect density is higher. The S-parameter of the untreated sample is statistically distributed around the mean value of 0.521 60(2). The contour plot is not symmetric, although the pin features a rotational symmetry.

In order to investigate the structure of the weld in more detail, two line-scans with a step size of 0.25 mm were carried out. The tracks of both scans are marked in figure 3. The
results of these measurements are represented in figure 4, where the solid black lines guide the eye. Just as the horizontal scan, also the vertical scan is restricted to smaller values of \( y \) and \( x \), respectively, because there, the S-parameter is dominated by edge effects.

Considering the horizontal scan on the left hand side in figure 4, at first glance, the \( x \)-position of the minimum of 4.5 mm matches the actual position of the center of the weld at \( x = 4.7 \) mm. The latter is marked by the vertical blue dashed line. The two thin black lines delimit the width of \( \Delta x = 4.3 \) mm of the conical pin at \( y = 1.3 \) mm, where the dashed line was considered as the center of the pin. From that, it is obvious, that the region with the increased defect density (vertical red line) is not in direct contact with the pin but is strongly deteriorated by it. Therefore, this region can be identified as the TMAZ. Furthermore, the graph shows that the defect distribution on both sides of the pin is asymmetric.

On the right-hand side of figure 4, the vertical line-scan at an \( x \)-value of 2.8 mm shows a minimal S-parameter at about \( y = 1.55 \) mm (marked by the blue dashed line). It is obvious, that the width of the minimum does not correspond to the actual length of the weld in \( y \)-direction on the sample. Starting from the minimum at \( y = 1.55 \) mm, the S-parameter first increases smoothly with increasing \( y \) before it jumps to higher values near \( y = 2.65 \) mm (thin black line). Thus, even in the \( y \)-direction, the defect density is not constant in the joint zone. The defect concentration is minimal at the shoulder of the tool. Still in the weld, the maximal defect concentration is reached at about \( y = 4.25 \) mm.

3.2. Al/Fe-sample

In a first step, a two-dimensional defect map of the sample, the material pairing, was recorded by DB, whereby the whole cross-section indicated in figure 2 was investigated. Since Al and Fe have very different S-parameters, it is possible to distinguish Al and Fe containing materials roughly by a DBS scan as demonstrated in figure 5. Due to an offset in positive \( y \)-direction of about 0.9 mm, the length of the Al alloy component in \( y \)-direction does not correspond to the real height of the Al alloy given in figure 2. However, this does not matter for the interpretation of the mixing process. The track along which the pin was moved and therewith the joint zone can clearly be seen between \( x = 5.5 \) mm and \( x = 10.4 \) mm and is marked by the double arrow (compare figure 2). At these two positions, the steel clutched at the pin and hence, was dragged into the Al alloy. Therefore, a mixture of both materials is expected in the region in between. Consequently, the measurement points to be investigated by CDBS were chosen within this region and are marked by numbers in figure 5. However, figure 5 demonstrates, that also the steel was affected by the movement of the pin, although the pin only scratched its surface. However, it is not possible to distinguish by DB measurements whether this impact is caused by defects inserted by the pin or by Al getting stirred into the steel. Position No. 1 serves as a reference of the untreated Al alloy for the CDBS measurements.

Generally, the aim of the investigations by CDBS was to identify the percentage of positrons annihilating with core
Figure 4. FSW Al/Al sample: line-scan in the $x$-direction with $y$ fixed at 1.3 mm (left). Line-scan in the $y$-direction with $x$ fixed at 2.8 mm (right). The meaning of the vertical lines is explained in the text.

Figure 5. 2D scan of sample Al/Fe, the material pairing, which shows the S-parameter laterally resolved with a step-size of 1 mm in each direction. The FSW tool entered the sample from the bottom and was moved in $z$-direction. The double arrow marks the width of the track of the FSW tool, which corresponds to the tool diameter of 5 mm quite well. The two jointed components are clearly distinguishable by the different values of the S-parameter. The numbers indicate positions which were investigated further by CDBS, with No. 2 to 4 being in the clearly visible joint zone. No. 1 is the position of the Al alloy reference measurement.

electrons of iron in the aluminum component and vice versa. In order to do this, each spectrum had to be normalized to the same total intensity. Statistics was improved by mirroring the red-shifted part ($E < E_0 = 511$ keV) of the peak at 511 keV onto the blue-shifted ($E > E_0 = 511$ keV) one. Division of all spectra by the reference curves obtained for the Al alloy (position 1), clearly reveals the element specific features of the second reference curve of pure annealed iron.

These so-called ratio curves obtained at the positions 2–5, are presented in figure 6. The red curve corresponds to pure annealed iron and thus runs under all other curves for $E < 512$ keV as its defect concentration is very small. The reference curve of the Al alloy measured at position 1 is represented by the black line with a constant ratio of 1.0. Except curve 5, all other measured positions show a significant Fe signature. Thus, no iron is detected in the vicinity of position 5, which leads to the conclusion that the steel is not transported that far up by the pin. Therefore, it is clear that the iron signature of position 4 is less distinct than the signature of position 3 which is located in the region where
Figure 6. Ratio curves for the positions on sample Al/Fe (as indicated in figure 5) examined by CDBS. These were obtained by dividing all recorded Doppler broadened 511 keV lines by the reference spectrum of the Al alloy (position 1). The uppermost curve represents the elemental signature of pure, annealed iron. All ratio curves located in the joint zone show the clear signature of Fe. For these curves, a fit by a linear combination of the two reference curves of Fe and Al alloy was carried out and is represented by the solid lines.

The solid lines represent least-squares fits of the measured points using a linear combination of the two reference ratio-curves $R_{Fe}$ and $R_{Al}$. In order to determine the free parameter $p$ of the fit function $R_{fit}(E) = pR_{Fe}(E) + (1-p)R_{Al}(E)$, the least-squares fit had to be weighted by the errors of the particular point. Furthermore, Doppler shifts smaller than 3 keV and larger than 9 keV were not considered: for energies $E > 512.3$ keV, the measured intensities are low, so that the uncertainties become rather large. The ratio-curves for $E < 514$ keV are dominated by the annihilation radiation of conduction electrons that do not carry element specific information. Despite the fact, that steel and the aluminum alloy consist of several alloying elements, and the fit is a linear combination of only iron and the Al alloy, the fits are in excellent agreement with the data. The physical interpretation of the fit parameter $p$ is the percentage of positrons that annihilate in iron. As curve 3 shows a clear signature of annealed iron, and $p$ amounts to 34.9%, the formation of annealed steel clusters in the Al alloy is very probable. The lower intensities of curves 4 and 2 are reproduced very well by the smaller values of $p$ of 20.1% and 14.9%, respectively.

4. Discussion

The investigation of the distribution of open volume defects in the Al/Al sample, for example vacancies, showed a distinctly decreased defect density within the weld, with an adjoining region of a generally increased defect density. This result is interpreted as follows: previous examinations of the temperature during welding by the IWB yielded temperatures up to 500 °C at the shoulder of the tool [17]. Consequently, annealing takes place in the joint zone, since the annealing temperature of Al alloys in general is about 400 °C [18]. This effect reduces the defect concentration in the material, thus explaining the high strength of FSWs observed during tensile tests.

However, an increase of the defect concentration within the weld can be observed in the vertical line-scan. This observation matches very well with the educated guess made in FSW technique that implies that the majority of the heat input is produced by the friction due to the shoulder of the tool and not by the pin itself [3]. Thus, a negative temperature gradient is expected in $y$-direction which, in turn, leads to a positive defect concentration gradient, as annealing strongly depends on temperature. In addition, the rotation of the pin has to be considered, which, to some extent, destroys the original structure of the material and hence, induces defects. Therefore, the welding process can be understood...
as a superposition of both deterioration and annealing. In addition, a minimal temperature (300 °C) is required to initiate annealing, which explains the sudden increase of the defect density at a certain distance from the shoulder in the vertical scan. Thus, here the temperature must have been fallen beneath this minimal temperature and the deterioration of the material prevailed.

In horizontal direction, the same effect arises for larger distances from the pin, too: the temperature has dropped under the on-set temperature of annealing, but the material is still deteriorated by the rotational movement, which increases the defect concentration. Thus, the TMAZ could be clearly located by DBS and the discussed results concerning the microstructure agree very well with those obtained by previous investigations [4]. In accordance with x-ray diffraction results, also DBS showed an asymmetry in the defect distribution on the advancing and the retreating sides of the pin. In the first case, the track speed is parallel to the feed speed; in the second case, these two velocities are antiparallel.

The investigations of the material pairing by CDBS enabled detection of steel clusters in the Al alloy, owing to particular properties of the positron. The positrons become trapped at the interface between the Al alloy and the steel cluster where a high defect density can be expected due to the lattice mismatch. Hence, a large fraction of the positrons annihilates with core electrons of iron which explains the high intensity of the ratio curve corresponding to position 3. Furthermore, the positron only annihilates with an electron of Fe, if the attractive potential caused by the positron affinity to Fe is large enough to trap the positron. By approximating this potential by a three-dimensional spherical well, a minimum number of about 12 Fe atoms for α-Fe is obtained, in order to ensure the trapping of positrons. Given the distinct iron signature, steel clusters have to be present at position 3. However, the coexistence of intermetallic phases in the sample cannot be ruled out with these results only. The considerably lower percentage of positrons annihilating with core electrons of iron at position 4 suggests the absence of an Al/steel interface and thus against steel clusters in this region. This observation is supported by the image in figure 7, which was taken during an additional examination of this sample by Scanning Electron Microscopy (SEM). At a distance larger than 1 mm from the steel/Al alloy boundary, almost no steel clusters are visible. The clear iron signature and the results of DBS even suggest that the detected clusters consist of annealed iron. However, an examination of the sample by Energy Dispersive X-ray (EDX) Spectroscopy could not prove this assumption because of the very high iron content of 235 of about 98.3%, other elements like Mn with a percentage of 1.4% are not detectable even in the steel component itself.

In addition, EDX showed, that only a minor amount of the Al alloy was stirred into the steel component. Therefore, it can be ruled out that the annihilation of positrons with a large number of core electrons of Al is responsible for the low relative intensity of the ratio curve at position 2. On the contrary, it is generally known in positron physics that ratio curves can be influenced in this way by defects. This is understandable, if it is kept in mind that a large defect density causes a sharpening of the annihilation line and thus, most positrons do not annihilate with core electrons at all, but with the slow conduction electrons. This leads to a very low intensity $I(E)$ and hence to a small $I(E)/I_{Al}(E)$ ratio for the high momentum region of the peak. Compared to position 1, the reference measurement for $I_{Al}(E)$, an increased defect density is expected at position 2 from the examinations of the defect distribution at the tip of the pin in sample Al/Al. To obtain a deeper insight into the influence of various kinds of defects, sophisticated ab-initio calculations would be helpful, in order to suppress the contribution of defects on the shape of ratio curves [19, 20].

5. Conclusion and outlook

In this work, two FSW samples were investigated by DBS; one sample, an Al/Fe material pairing, was also studied by CDBS. We were able to map the defect distribution with a spatial resolution down to 0.25 mm in the first sample where annealing of the material was clearly observed. With growing distance from the shoulder, annealing diminishes and the deterioration of the material caused by the stirring of the pin prevails. These results match very well with assumptions of FSW technique, which implies the existence of a thermo-mechanical affected zone with a greatly deformed microstructure. In order to benefit from this observation, future examinations with positrons should be carried out for samples consisting of materials with known annealing temperatures. The prior knowledge possibly allows to draw conclusions about the temperature distribution at the pin during the welding process which is hardly accessible experimentally. The further examination of the TMAZ and
HAZ would be highly interesting for process engineering since jointed components predominately fail in this region.

The ratio curves obtained by CDBS at the joint of steel and Al alloy show a clear signature of pure, annealed iron. These normalized CDB spectra can be excellently fitted by a linear-combination of the reference curves from iron and Al alloy and hence, suggest the formation of steel clusters (≥98.3% Fe) due to the friction heat. In order to minimize the influence of defects on the ratio curves and to compare the results with those of complementary examination techniques applied to FSW, such as XRD or the insertion of markers, further investigation should deal with ‘conventionally’ produced FSWs. These are welds, which result from joining two workpieces positioned side by side. In this case, the examination of the chemical vicinity on the two different sides of the weld at different depths, would lead to a better understanding of the differences in the mixing process between advancing and retreating sides of the pin.

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