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"Comparative studies on the characterisation of ultrafine particles during welding and allied processes"

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Comparative studies on the characterisation of ultrafine particles during welding and allied processes

Summary of the final report

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Foreword

The research project "Comparative studies on the characterisation of ultrafine particles during welding and allied processes" was launched by the BG expert committee "Metal and surface treatment" of the Deutsche Gesetzliche Unfallversicherung (DGVU; German Statutory Accident Insurance), section "Hazardous substances during welding and allied processes" and has been financially sponsored by the Vereinigung der Metall-Berufsgenossenschaften (VMBG; association of metal BGs).

The following BGs are represented in the VMBG:

Berufsgenossenschaft Metall Nord Süd
Hütten und Walzwerks-Berufsgenossenschaft
Maschinenbau- und Metall-Berufsgenossenschaft

A steering committee, consisting of the heads of the prevention departments of the Metall-BGs, representatives of the social partners, the BG expert committee "Metal and surface treatment", of the DGVU and other experts accompanied the project.

The research project has been carried out as joint project of the institutes for welding and allied processes and for joining technology of the RWTH Aachen and the Fraunhofer Institute for toxicology and experimental medicine (ITEM) Hannover.

Objective

Stimulated by discussions on the determination of a specific air limit value for welding fumes during the last years and in order to gain information on type, quantity and pathogenous effect of ultrafine particles (UFP) in welding fumes, it was considered necessary within the expert committee "Metal and surface treatment", section "Hazardous substances in welding and allied processes" to carry out a characterisation of these welding fume fractions. The intention was to present and document the possible hazard potentials of welding, soldering and cutting processes.

The question arising especially concerns dosimetry. Is the mass the correct measuring quantity or is it rather number or surface? How should the surface be characterised? Which importance has the solubility of the particles? These knowledge gaps make a quantitative evaluation of the UFP with occupational medical relevance very difficult at present.

Thus, the aim of the project was to carry out a comparative study of welding fumes for different processes relevant to practice with different materials.

In general, the hazard potential of welding fume is due to the hazardous substances present in the gaseous and particulate phases, welding fume being a singular mixture.

The characterisation of ultrafine particles in view of their specific characteristics, i.e. the physical properties of the welding fumes with potentially toxicological relevance, among others the diffusion equivalent diameter of the agglomerates, number and geometric diameter of the primary particles and their "biologically active" surface was done alongside the determination of the emission rates, the chemical compositions and the collection of the respirable and inhalable fractions (A and E fractions).

The parameters relevant for the physical characterisation of the welding fumes are mass, surface, number and morphology. They are investigated in this project in order to find out if there are basic differences in particle size distribution and particle morphology in the different welding and allied processes.

Due to the synergistic effects between gaseous and particulate phases described in literature, the mass related chemical characterisation of the exposure atmosphere is not sufficient.

Proceedings

The following processes were investigated:
Manual metal arc welding with covered electrodes,
Gas-shielded metal arc welding (GSM)
- Metal active gas welding (MAG)
- Metal inert gas welding (MIG)

Tungsten inert gas welding (WIG)
Resistance spot welding (random)
Resistance spot welding with adhesive bonding (random)
Non-vacuum electron beam welding (NVEBW)
Laser welding
Laser hybrid welding
MIG soldering
Soldering and brazing
Thermal cutting processes
- gas cutting
- plasma cutting
- laser cutting

The following materials were used: non-alloy and low-alloyed metals, high-alloy metals, copper-based alloys, aluminium and aluminium alloys.
The shielding gases used were mainly mixed gases with carbon dioxide, argon, helium and oxygen.

For sampling and for the determination of the emission rates and the chemical analysis, different welding fume samplers were used. The fume boxes specified in EN ISO 15011-1 were suitably adapted to the processes investigated.

The determination of the mass size distribution was done with a low pressure Berner impactor. This sampler enables collection of particles of 10 aerodynamically different size categories on aluminium foils and subsequent gravimetrical determination. The size range extends from 0,03 to 16 µm.

For the determination of the number size distribution and for the measurement of the total number concentration, the welding fume is firstly diluted, as the number concentration of the welding fume examined is very high. For this purpose, a so-called rotation diluter is used, which is connected upstream to the Scanning Mobility Particle Sizer (SMPS) and at the same time to the Condensation Particle Counter (CPC).

In the SMPS, particles with defined electric load are classified in the electric field. Dependant on the strength of the electric field, a narrow size fraction of the aspirated welding fume is isolated and transferred to the CPC for the measurement of the particle number concentration of the isolated size fraction. The measuring range of the process extends from 0,01 to 0,5 µm according to the setting. The classification in the electric field is done on the basis of the so-called mobility diameter.

The agglomerate emission rates are calculated from the determined number concentrations and the set suction volume flow.

The collection of the agglomerate has to be done carefully in order to maintain the partly very filigree structure for further examinations in the transmission electron microscope (TEM). For this reason, the ITEM already developed a thermophoretic separator some time ago, which, due the deposition mechanism it is based on, separates representatively and keeping the structure. The particles to be collected are subjected in a precipitator to a temperature gradient in such a way that the surface of a TEM net is on a lower temperature level than the welding fume flowing over the net.

For the determination of the mean primary particle number, the agglomerates are separated by means of the thermophoretic separator and electronic images are taken by a TEM. The determination of the primary particle emission rate is determined from the identified mean primary particle number per agglomerate and the agglomerate emission rate.

The methodology described was used for all processes.

Results

Manual metal arc welding

For manual metal arc welding with covered electrodes four non-alloy and low-alloy electrodes and a high alloy rod electrode were used.
The welding fume emission rates were between 3,9 mg/s for a rutile-basic coating and 7,6 mg/s for a basic coating.

The chemical composition of these fumes corresponds to the composition of the coating and the rod: for most non-alloy and low-alloy rod electrodes, the potassium content and the iron content are higher then the sodium content. As opposed to that, the welding fume of high-alloy rod electrodes contains higher amounts of sodium, calcium and chromium(VI) compounds.

The mean aerodynamic mass median diameter is similarly high for these tests, i.e. between 0,34 and 0,45 µm. The number median of the mobility distribution is between 117 and 142 nm. The agglomerate emission rate of the welding fume particles is between 3,57 10^{11} for the basic covered high-alloy rod electrode and 7,76 10^{11} for the cellulose covered non-alloy and low-allow electrode. The highest mean primary particle number per agglomerate (91) for a simultaneously very small median value of the primary particle size distribution (7nm) is found in the fume of the rutile covered non-alloy and low-alloyed rod electrode.
The lowest value for the mean primary particle number per agglomerate (24) with the highest median value of the primary particle size distribution (15nm) at the same time is found in the fume generated by the basic covered and low-alloyed rod electrode. The calculated primary particle emission rates (1/sec) are between 1,2 10^{13} for the basic covered electrode and 5,9 10^{13} for the cellulose covered electrode.

Metal active gas welding / surfacing

With this process, non-alloy and low-alloyed wires and filler wires, high-alloyed wires and coated sample material were examined. Different shielding gases were used for the same process/material combinations, e.g. 100% CO_2, Ar/CO_2 mixtures, in order to determine their influence on the emission rate and other characteristics of ultrafine particles.
Among others, it was confirmed that the use of an Ar/CO_2 mixture instead of CO_2 as shielding gas leads to significantly lower welding fume emission rates.

As expected, the highest emission rates of 30,4 mg/s are measured during flux-cored wire welding without shielding gas with a self-shielding metal powder filler wire. Flux-cored wire welding with 82% Ar and 18% CO_2 as shielding gas also reaches emission rates of 24,8 mg/s. The lowest emission rates of 2,4 mg/s were found during welding with massive wire under argon/CO_2 mixture, independent of type of alloy (low- or high-alloyed).

When the particle size distribution in the latter process is considered, it is found that the aerodynamic mass median diameter is between 0,13 and 0,45 µm. The number median of the mobility distribution is between 120 and 156 nm. The agglomerate emission rate of the welding fume particles is between 3,25 1011 for the coated sample material and 1,691012 for the flux-cored wire welding with self-shielding metal powder filler wire.

The mean primary particle number per agglomerate varies according to the used filler metal and shielding gas between 69 and 353. The calculated primary particle emission rates (1/sec) are between 3,8 10^{13} for high-alloyed filler wire and 3,9 10^{14} for non-alloy metal powder wire.

Metal inert gas welding

With this process, different aluminium and aluminium alloys and a nickel base alloy were welded. Ar and Ar/He mixtures served as shielding gases. The emission rates were measured for aluminium-silicon alloys and for pure aluminium alloys. As expected, the highest values were found for aluminium-magnesium alloys. Welding with an Ar/He mixture as shielding gas instead of pure argon leads to lower emission rates for aluminium-silicon materials.

With values between 0,32 and 0,42 µm,the mean aerodynamic mass median diameter AMMD is roughly equal for all tests and comparable to manual metal arc welding. The number median of the mobility distribution is between 60 for the nickel base material and 208 for the aluminium-magnesium material. The agglomerate emission rate (1/sec) is of the same order of magnitude for all materials and varies between 2,24 10^{11} for aluminium-silicon and 5,24 10^{11} for aluminium-magnesium materials.

The tests show great differences between the different filler metals in the tests as to the mean primary particle number per agglomerate. The values vary between 86 for welding with aluminium-silicon material under Ar/He mixture and 1535 for the same filler metal under argon as shielding gas. Thus the influence of the shielding gas on the primary particle number per agglomerate, i.e. on the formation of agglomerates, becomes evident.

For the nickel base filler metal used, a low primary particle number per agglomerate (116) was measured.

The calculated primary particle emission rates (1/sec) are in accordance:
the values are between 2,4 10^{13} for aluminium-silicon material under Ar/He and 7,3 10^{14} for aluminium-magnesium material under argon as shielding gas. For the nickel base material, the calculated primary particle emission rate is in the same order of magnitude as for aluminium-silicon, i.e. 4,5 10^{13}.

Tungsten inert gas welding

In this process, the emission rates are very low independently from the filler metals chosen and do not exceed 0,038 mg/s. The number median of the mobility distribution is also very low with values between 23 and 63 nm. The agglomerate emission rage (1/sec) is in the same order of magnitude for all materials and varies between 3,69 10^{11} for high-alloy chromium-nickel filler metal and 6,14 10^{11} for copper as filler metal. The calculated primary particle emission rates (1/sec) are between 6,2 10^{12} and 5,0 10^{13}.

Laser welding

The mass emission rates are in the same order of magnitude as for MIG welding, i.e. between 2,6 mg/s for AlSi1MgMn and 8 mg/s for hot zinc-dipped steel, although no filler metal was used. The zinc coating used as shop primer leads to a significant increase of emissions.

The mean aerodynamic median diameter varies between 0,1 and 0,25 µm. The number median of the mobility distribution has about the same magnitude for all tests at 152 to 181 nm. The agglomerate emission rate (1/sec) varies between 5,9 10^{11} for zinc-coated steel and 2,1 10^{12} for AlSi1MgMn. The calculated primary particle emission rates (1/sec) are partly in the same order of magnitude as for MIG welding, at 3,5 10^{14}, and partly are higher by an order of magnitude, at 1,5 10^{15}.

Laser hybrid welding

Here, the mass emission rates are higher than for laser welding, as expected, since on one hand a filler metal is used and on the other hand a combined exposure by two processes has to be anticipated. For this reason, values up to 18,5 mg/s occur for zinc-coated steel. As opposed to that, emission values which are much lower at 4,3 mg/s were measured for AlMg or AlSi1 alloys. The mean aerodynamic median diameter is the same for all tests at about 0,4 µm.
The number median of the mobility distribution is 97 nm as lowest value for AlSi materials and reaches 184 for zinc-coated steel. The calculated primary emission rates (1/sec) are in the same order of magnitude as for MIG welding and laser welding at between 1,6 10^{14} for zinc-coated steel and 2,8 10^{15} for low-alloyed steel.

Resistance spot welding/resistance spot welding with adhesive bonding

The mass emission rates are very low for all 6 tests and are far below 1 mg/s, being 0,05 mg/s at maximum. Here as well the emission values are higher for organically coated sheets as for zinc-coated sheets and the latter are higher than for non zinc-coated sheets, as expected. Generally the emission rates for welding are higher than for adhesion.
The agglomerate emission rates (1/sec) for this process are also low with values between 4,5 10^{9} for unalloyed sheet and 2,1 10^{11} for the organically coated sheet. The calculated primary particle emission rates (1/sec) are comparable to TIG welding at between 1,7 10^{11} and 3,9 10^{13} for organically coated materials.

Thermal cutting on water bath or with underfloor extraction

The measured mass emission values for plasma cutting of stainless steel and for thermal cutting of construction steel are very low at 0,66 mg/s at maximum - due to the use of a water bath below the workpiece and an extraction above the workpiece. The mean aerodynamic median diameter is lower for plasma cutting of stainless steel and laser cutting of construction steel (0,12 µm) than for thermal cutting of constructional steel and for laser cutting of stainless steel (0,35 µm). Due to the protective measures used, the agglomerate emission rates (1/sec) are lower than for equal processes without protection measures, at 9 10^{10} for laser cutting of stainless steel and are 1 10^{12} at maximum for plasma cutting of stainless steel. The calculated primary particle emission rates (1/sec) are comparable to MIG soldering and brazing and vary between 1,8 10^{13} and 1,8 10^{14}.

Soldering and brazing

The mass emission rates are lower for brazing than for soldering and reach 1 mg/s at maximum. The metals on the basis of tin and silver used contain in addition more than 20% each of copper and zinc for brazing. In the small fume quantities of 0,5 mg/s the metals mentioned above are hardly detectable.
The mean aerodynamic median diameter is slightly lower for brazing (0,34 µm) than for soldering (0,56 µm). The mobility distribution shows median values of between 39 and 65 nm.
The agglomerate emission rate (1/sec) shows differences between brazing with values of 8,5 10^{11} to 9,1 10^{11} and soldering with higher values in the range of 3,2 10^{12} to 4,3 10^{12}. The calculated primary particle emission rates (1/sec) are about 1 10^{13}.

Metal inert gas soldering and brazing

3 different filler metals on the basis of copper were chosen and each of them was welded with two different shielding gases: Ar/CO_2 and Ar/O_2 mixtures.
The parent metal used was zinc coated in all 6 tests.
The emission rates generated are higher than for soldering and brazing. They are between 1,9 mg/s for soldering with CuSi3 and 4,9 mg/s for soldering with CUAl8, both tests under Ar/O_2 mixture. The chemical composition of the welding fumes in all 6 tests reveals two components as prevailing: copper from the filler metal and zinc from the parent metal coating.

The mean aerodynamic median diameter varies between 0,17 and 0,33 µm. The mobility distribution shows similarly high median values: 129 to 164 nm.
The agglomerate emission rate (1/sec) shows slight variations of 2,9 10^{11} for CuSi3 under Ar/CO_2 mixture to 4,31 10^{11} for AlBzNi2 under Ar/O_2 mixture.

The lowest mean primary particle number per agglomerate of 149 is reached during soldering with the filler metal CuAl8 under Ar/O_2, the highest number of 735 is reached when using Ar/CO_2 with the same filler metal. At present, a recommendation for a targeted strategy cannot be given, since further investigations on the matter are required.

As to the calculated primary particle emission rate, the difference in the 6 tests is significant and varies between 5,2 10^{13} for CuSi3 materials and 2,4 10 for CuAl8 materials.

Electron beam welding in atmosphere

The mass emission rates are in most cases very high at about 22 to 32 mg/s. The mean aerodynamic median diameter varies between 0,17 and 0,28 µm. The mobility distribution shows similarly high median values: 124 to 237 nm. The agglomerate emission rate (1/sec) shows hardly any variations with 2,65 10^{12} to 3,65 10^{12}.
The lowest mean primary particle number per agglomerate of 43 is found for the zinc-coated parent metal. The magnesium parent metal has the highest mean primary particle number per agglomerate of 704 besides the highest mass emission rate. The calculated primary particle emission rate (1/sec) varies between 1,6 10^{14} for the zinc-coated parent metal and 1,9 10^{15} for the magnesium parent metal.

Summary

Mass emission and chemical composition

In the welding processes, where no filler metal was used, the parent metal is the main influencing factor for the emissions.
In the processes with filler metal, the filler metals are represented in the chemical composition of the welding fumes. The parent metals, however, only show little influence on the emission rate and the chemical composition.
At the same time, a relation to the welding parameters chosen can be seen:
In proportion to the increasing melting capacity, the emission rate increases as well; the tests also show that the emissions increase with increasing welding speed. This gives rise to the conclusion that the weld pool volume produced has an influence on the emissions.
In addition, a significant influence of the shielding gas is obvious.

Size distribution, primary particle emission rate and specific surface

Coagulation is one of the most important mechanisms influencing the size distribution of aerosols. In general, it leads to the formation of larger units and thus to a modification of the size distribution.
The shifting of the mean particle diameter and the modification of the number concentration even for relatively short aging periods may be significant.

The size distributions found in the investigations are mainly determined by the aging period of the measured dusts as to mass and agglomerate number and only depend on initial parameters to a smaller degree.

With increasing mass emission (mg/s), the primary particle emission rate also increases. This correlation is confirmed by a correlation coefficient r of +0,41 and indicates that with increasing mass concentration, e.g. at the workplace, generally an increasing exposure by the primary particle concentration has to be anticipated.

For answering the question concerning the relation between total particle surface and mass, a detour using the primary particle emission rate and the primary particle diameter is necessary. Due to the strong scatter of the values, reliable data can only be achieved by statistical methods.

Statistical evaluation shows that the majority of the examined processes, which rather belong to the conventional processes, are grouped relatively close to a straight line with a gradient of $2{,}3 \cdot 10^{13}$ P_p/mg. Only the three processes below are significantly different from the others. The electron beam welding is still relatively close to the line mentioned above with $4 \cdot 10^{13}$ P_p/mg. The resistance spot welding/resistance spot welding with adhesive bonding is about two orders of magnitude below that value with $4 \cdot 10^{11}$ P_p/mg. This is probably mainly due to the fact that in this welding process besides the typical agglomerates from the evaporated metals and metal oxides also other materials are released which rather form compact particles and thus contribute significantly less to the small primary particles. The laser welding processes on the other hand are roughly one order of magnitude above the straight line with $1 \cdot 10^{14}$ P_p/mg.

The information from the primary particle emission rates and the mass emission rates may be used for an evaluation of the specific surface of the welding dusts.
The mean value of the specific surface for all processes is 307 m^2/g. This value corresponds to the specific surface of a rust particle (density 5 g/cm^3) with a geometric diameter of 7 nm or a sphere with the density of 2 g/cm^3 and a diameter of 18 nm and is thus in a range, which can also be anticipated for the primary particles of the welding fume.

Primary particles and surface emission rate are principally correlated with the mass emission rate (mean primary particle emission rate about $5 \cdot 10^{13}$/mg). For some welding processes, this correlation is, however, only given with large deviation. As expected, welding processes, where other easily volatile substances may be released besides metal and metal oxide compounds, do not show this correlation by far (e.g resistance welding).

From the data available, a surface emission rate can also be estimated. The specific surface of the welding fumes can then be derived from the ratio of the surface emission rate and the mass emission rate. The mean specific surface for all examined processes was determined to be 307 m^2/g. The order of magnitude of this value corresponds to the specific surface that could be expected in view of the primary particle diameters.

The different welding processes also revealed, however, significant differences in the specific surface of the released welding fumes.
For conventional welding processes, the gradient of the straight line gives a mean value of 58 m^2/g. Laser welding processes deviate up from this value with 250 and 460 m^2/g (laser hybrid welding). Electron beam welding is between the above processes with 270 m^2/g. For the reasons already mentioned, resistance spot welding deviates significantly down with 0,4 m^2/g.

The results show that the processes used are suitable for an in-situ determination of the specific surface of welding fumes. Due to the variety of the welding processes investigated and the large number of the TEM images required per sample, multiple determinations for a further validation of the data were not possible in the framework of this project. Therefore, additional tests limited to the random determination of the primary particle and mass emission rate and which allow for multiple examinations are considered to make sense. In parallel to these tests, measurements with devices for aerosol surface area measurement, which are commercially available by now, shall be carried out. In case of concurring results, the already available and the future measuring results can be used to easily enlarge the data basis on the specific surface of welding fume.

Reasoning

The characterisation of ultrafine particles and the identification of the most important mass-related properties like mass emission rates, particle size distribution related to the geometric diameters, were carried out for 11 groups of processes. For each of these groups of processes, different filler metals/parent metals were used in each process.

On the whole, about 95 samples were evaluated as mean values from a total of about 350 samples.

Mass emission rates

When comparing all process/material combinations, the following becomes obvious:

- the highest mass emission rates are generated during **MIG welding of AlMg alloys** (31,96 mg/s), during **electron beam welding in atmosphere** (31,7 mg/s) and during **MAG welding with filler wire without shielding gas, with self shielding filler wire** (30,4 mg/s)
- **the above three process/material combinations with emission rates of more than 25 mg/s cause a very high hazard as the specified air limit values are expected to be exceeded several fold.**
- also the process/material combinations **MAG welding with filler wire** under 82 % Ar and 18 % CO_2 with an emission rate of 24,8 mg/s, **MIG welding of AlMg3 alloys** under Ar with emission rates of about 24 mg/s belong to the group of high emission processes. **The use of these process/material combinations includes high hazards as here as well significant exceeding of the limit values may occur, if no relevant ventilation measures are realised at the workplace.**
- **MIG welding of nickel** with emission rates of 1,45 mg/s **and laser hybrid welding with AlSi alloys** with emission rates of 1,2 mg/s **cause a medium hazard,** i.e. the expected concentrations at the workplace in the breathing zone of the welder are in the range of the limit values or slightly below.
- among all processes used, the following processes are confirmed to be **low-hazard processes as related to mass emission rates: TIG welding** for all materials used with emission rates of 0,038 mg/s at maximum, **soldering and brazing** with emission rates of 1 mg/s at maximum, **resistance spot welding/resistance spot welding with adhesive bonding** with emission rates of 0,050 mg/s at maximum and **thermal cutting on water bath** or with very effective extraction with emission rates of 0,66 mg/s at maximum.

Mean primary particle number per agglomerate

Together with the agglomeration rate, the mean primary particle number per agglomerate is used to determine the primary particle emission rate.

Here as well, the composition depends on the aging of the collected agglomerates. Per welding process, an average of 60 TEM images were evaluated and according to agglomerate size (generally more than 300 particles/agglomerate) several images per agglomerate were necessary.

A very high mean primary particle number per agglomerate (about 2600) was measured for laser welding without filler metal and for laser hybrid welding. Not all of the tests carried out during the a.m. processes yielded such a high agglomeration rate. On the contrary: the mean primary particle number per agglomerate during laser

hybrid welding with AlSi12 and for zinced parent metal was determined to be between 200 and 510.
During some tests with TIG welding values of more than 1000 for the mean primary particle number per agglomerate were found. Generally, most values are between 500 and 750.

Agglomerate emission rates and primary particle emission rates

When comparing all processes investigated as to **agglomerate emission rates** it is found that the values determined have similar tendencies in most tests and do seldom exceed the order 10^{12}. On the group level, the agglomerate emission rate is higher for electron beam welding in atmosphere and for MAG welding and much lower for MIG and TIG welding. The **primary particle emission rates** vary very strongly within the process groups and between them. Low values were calculated for soldering and brazing and TIG welding, relatively high values for MAG, MIG, laser MIG hybrid welding and electron beam welding.

Conclusion

As opposed to other characteristics (mass size distribution, agglomerate number emission rates, mean primary particle number per agglomerate), the primary particle emission rates are not subject to aging and are thus transferable at e.g. workplaces.

The primary particle size distribution, from which the primary particle median diameters are determined, is influenced by coagulation. For a given volume concentration, the number concentration decreases with time, the mean particle diameter increases analogously.
For the sampling of welding fumes, this means that for comparative measurements, identical sampling times and points shall be observed.

The number median of the mobility distribution (nm) also depends very strongly on time and increases with time.
From this it results that the variation of the mean particle diameter and the modification of the number concentration may be very significant even for relatively short aging times.

The size distributions found in the tests are determined mainly by the aging period of the measured fumes as far as mass and number of agglomerates are concerned and the initial parameters are less important.

For most of the investigated processes, the generated mass emission rates exceed 2 mg/s and even reach 32 mg/s. Thus, these processes are high-emission processes and shall only be used at the workplace with suitable ventilation measures preventing that the current limit values in the breathing zone of the welders are exceeded and excluding hazards by exposure to welding fume.

Literature source:
Pohlmann, G., and Holzinger, K., Comparative studies on the characterisation of ultrafine particles during welding and allied processes, Final Report, 2008, Hrsg. Vereinigung der Metall-Berufsgenossenschaften