

Characterization of Fiber Dust Resulting from Recycling of Carbon Fiber-Reinforced Thermoplastics (CFRP) and Their Cell Toxicity

Lisa Tölle¹, Christian Monsé², Nina Rosenkranz², Natalia Haibel³, Dirk Walter^{3,4}, Jürgen Bünger², Matthias Hopp¹, Götz A. Westphal²

¹Kunststofftechnik Paderborn, University Paderborn, Paderborn, Germany

²Institute for Prevention and Occupational Medicine of the German Social Accident Insurance, Institute of the Ruhr University Bochum (IPA), Deutsche Gesetzliche Unfallversicherung e.V. (DGUV), Bochum, Germany

³Laboratories of Chemistry and Physics, Institute of Occupational and Social Medicine, Justus-Liebig-University Giessen, Giessen, Germany

⁴Institute of Inorganic and Analytical Chemistry, Justus-Liebig-University Giessen, Giessen, Germany

Email: christian.monse@dguv.de, nina.rosenkranz@dguv.de, natalia.haibel@arbmed.med.uni-giessen.de, dirk.walter@arbmed.med.uni-giessen.de, juergen.buenger@dguv.de, lisa.toelle@ktp.uni-paderborn.de, goetz.westphal@dguv.de

How to cite this paper: Tölle, L., Monsé, C., Rosenkranz, N., Haibel, N., Walter, D., Bünger, J., Hopp, M. and Westphal, G.A. (2022) Characterization of Fiber Dust Resulting from Recycling of Carbon Fiber-Reinforced Thermoplastics (CFRP) and Their Cell Toxicity. *Journal of Materials Science and Chemical Engineering*, 10, 1-16.

<https://doi.org/10.4236/msce.2022.107001>

Received: June 4, 2022

Accepted: July 11, 2022

Published: July 14, 2022

Copyright © 2022 by author(s) and Scientific Research Publishing Inc. This work is licensed under the Creative Commons Attribution International License (CC BY 4.0).

<http://creativecommons.org/licenses/by/4.0/>



Open Access

Abstract

Carbon fiber-reinforced thermoplastics (CFRP) have excellent specific strength and rigidity, which has made them a popular material for lightweight construction. The growing demand for fiber-reinforced plastics (FRP) leads to the problem of the sustainable handling of FRP at the end of their life cycle. The aim of the research project was to gain knowledge about the shredding of FRP concerning the optimal machine and process design of the shredding process and the possible formation of harmful, inhalable dust fractions and WHO fibers. Toxicity should be investigated at the cellular level. The investigated shredding parameters influence the amount and length of fiber dust produced, both when shredding with a cutting mill and when shredding with a single-shaft shredder. In all investigations, an increasing rotational speed leads to an increase in the fiber dust mass or the fiber concentration. The proportion of short, respirable fibers increases, but raising the speed does not lead to a further, significant shortening of the fibers. A reduction in feedstock size leads to a slightly reduced mass of fiber dust in the ground material. A reduction in the screen size also leads to an increase in fiber dust mass and concentration. There was no recognizable cytotoxicity in the relevant concentration range up to 500 µg/cm² and no significant induction of cell migration. This indicates minor flammable effects of the dust formed after inhalation. The biological data indicate that the WHO fibers produced by shredding

are only a minor health hazard. Formally, the detected carbon fiber (CF) fragments meet the fiber definition of the legislator. However, carbon fibers currently have no specific limit value.

Keywords

Carbon Fibers, Fiber Dust, Recycling, Toxicity

1. Introduction

Carbon fiber-reinforced thermoplastics (CFRP) are increasingly used. With the growing need for these materials, the problem of sustainable handling of residual materials arises. Material recycling systems may enable sustainable handling and use of this resource. Recycling is a legal requirement in the EU [1]: a manufacturer is obliged to prepare an exposure assessment for a product over the entire life cycle.

A sustainable recycling process is mechanical recycling [2] [3], where the whole composite material can be reused as regrinding and further processed into a recyclate. Therefore, the composite is shredded followed by a compounding or extrusion process and granulated into a short fiber-reinforced regrind. Several studies demonstrated the potential to reuse the recycled thermoplastic fiber-reinforced material [4] [5]. Recycling requires shredding to a suitable size by single or multi-shaft shredder to a particle size of <50 mm or by fast-rotating cutting or hammer mills to a particle size < 10 mm for further processing [6], e.g. in the injection molding or extrusion process. During the shredding of thermoplastic fiber-reinforced plastics inhalable dust including fibers in the micrometer range as well as splitters with WHO fiber characteristics can occur. The possible formation of WHO fibers requires a high level of occupational safety. Inhaled asbestos fibers for example are classified as human carcinogens [7] [8]. The toxicity of such fibers is largely determined by their inflammatory properties. Other bio-persistent, inflammatory fibers are also suspicious due to similarities in structure and biological effects [9] [10]. The same is feared for new fibrous materials. Fibers of different geometry and bio-persistence must therefore be assessed carefully regarding possible toxic effects. However, there are no toxicological studies on the dust and fibers that occur when shredding fiber-reinforced thermoplastics.

Toxicity in animal experiments is rather low, following inhalation [11] [12] [13] [14], or intratracheal injection [15]. However, the bio-persistence seems to be pronounced [11] and inhalation studies can underestimate toxicity since longer fibers cannot be inhaled by the rat. The migration of inflammatory cells into the lungs is a reliable toxicological endpoint for the toxicity of particles [16]. This can be measured by a so-called broncho-alveolar lavage (BAL, flushing of the bronchi and lungs), which is equally stressful for animals and humans. Moreover, such an experimental procedure is far too complex for extensive

comparative investigations. Accordingly, *in vitro* test that depicts the inflammatory effects of particles and fibers is needed. In this research project, we, therefore, applied the particle-induced cell migration test (PICMA) [17] [18]. This approach assumes that predominantly inflammatory fiber effects lead to malignant cell changes and cancer. In addition, cytotoxic effects were examined.

This research project aims to achieve knowledge on fiber dust formation during the shredding of fiber-reinforced thermoplastics. One focus is on occupational health and safety, and a second focus concentrates on optimizing the shredding process.

2. Material and Methods

2.1. Material

For these investigations, polycarbonate with carbon fiber-reinforced (PC-CF) organic sheets Tepexdynamite210fr-C200(x) 45% from Bond Laminates GmbH (Lanxess, Germany) have been used. The sheets have a thickness of 1 mm and are cut into squared pieces, whose size depends on the respective shredder used.

2.2. Shredding Process and Analysis

2.2.1. Shredding Process in the Cutting Mill and Analysis of the Fiber Length in the Ground Material

For the investigations in the cutting mill SM300 (Retsch GmbH, Germany) equipped with a 10 mm screen is used while the rotational speed, and the feedstock size are varied. The organic sheets are cut into pieces of $30 \times 30 \times 1$ mm or $15 \times 15 \times 1$ mm size to analyze the effect of different feedstock sizes. Samples of 50 g each are prepared with the respective size and are then cut with the cutting mill for 1 minute each. The rotational speed is varied from 800 rpm, 1500 rpm to 2500 rpm. The ground material is sieved for 20 minutes to receive the particle size distribution and to separate the fiber dust from the rest of the material. The matrix of the fraction 0.125 mm is ashed to retain the pure fibers. The length of the fibers is analyzed using the scanner Epson Perfection V750 Pro (EPSON Deutschland GmbH, Germany) and the software FiVer (SKZ-KFE gGmbH, Germany). The results are shown in the particle size distribution (Figure 1) and the fiber length distribution (Figure 2).

2.2.2. Shredding Process in the Single Screw Shredder and Analysis of the Airborne Particles

The investigations with the single screw shredder WSC 250-400 (Weima Maschinenbau GmbH, Germany) focus on the analysis of the airborne fiber dust. Therefore, a feedstock size of the organic sheets of $250 \times 250 \times 1$ mm is determined. The rotational speed is varied from 66 rpm to 120 rpm and the screen size from 10 mm to 20 mm. A Scanning Mobility Particle Sizer (SMPS, Modell 3080, TSI Inc., USA) counts and measures particles on a nanometer scale (9 - 414 nm) whereas an Aerodynamic Particle Sizer (APS, Modell 3321, TSI Inc., USA) counts and measures particles in micrometer scale (0.5 - 20 μ m). The in-

halable fiber fraction (E-dust) was collected on a cellulose nitrate filter by using a VC25-Sampling device (former, Ströhlein GmbH, Germany). The filter was weighed before and after the shredding process and the fiber mass was standardized by dividing the collected fiber mass through the feedstock mass and the filtered air.

2.3. Toxicological Analysis

2.3.1. Sample Preparation

The fiber suspensions were prepared as follows: 20 mg of the fibers were filled in 4 mL brown glass vials and sterilized for 4 hours at 220°C. The fibers were then transferred in 5 mL Eppendorf vials and suspended in 2 mL PBS by ultrasonication using a Bandelin Sonopuls HD4050 Sonifier at 30 watt for 4 minutes. The used ultrasonic tip had a diameter of 2 mm and cooling was done using a salt/ice bath. Preparation of the stock solution for the milled matrix components was done by suspending 10 mg/mL of the particles in PBS.

2.3.2. Cell Lines

NR8383 cells were purchased from ATCC via LGC Standards GmbH (Germany) and cultivated at 37°C, 100% humidity and 5% CO₂ in Ham's F12 + 15% FCS (fetal calf *serum*, PAN-Biotech GmbH, Germany), 2 mM L-glutamine, 100 µg/mL penicillin, and 100 U/mL streptomycin. Approximately 3×10^6 cells were seeded in 35 mL (175 cm²) medium each.

2.3.3. Differentiation of HL-60 Cell

HL-60 cells were purchased from DSMZ (Germany). For the investigation of the chemotaxis we used trans-retinal differentiated HL-60 cells (dHL-60): The HL-60 cells were cultivated in RPMI 1640 medium (PAN-Biotech GmbH, Germany), 10% FCS, 2 mM L-glutamine, 100 µg/mL penicillin, 100 U/mL streptomycin and 1 µM trans-retinal at 37°C, 100% humidity and 5% CO₂, for three days [19].

2.3.4. Cytotoxicity

Measurement of cytotoxicity was performed by using the AlamarBlue Assay (Invitrogen, Life Technologies Corporation, USA). The assay measures the energy charge of the cells using the redox indicator. Resazurin enters the live cells as a blue, non-fluorescent compound and gets reduced to red-colored fluorescing Resorufin by any enzyme activity that is involved in the generation of NADH or NADPH, such as NADPH and NADH dehydrogenases or diaphorase. 10,000 NR8383 rat macrophages in 100 µL full growth medium (Ham's F12 medium containing 15% FCS, 2 mM L-glutamine, 100 µg/mL penicillin, and 100 U/mL streptomycin) were seeded into the cavities of a 96-well cell fluorescence culture plate (Corning Incorporated, USA). The cells were incubated at 37°C, 100% humidity and 5% CO₂ for 24 hours. The fibers were suspended in 100 µL full growth medium to a concentration of 512 µg/cm², diluted and pipetted into the wells, followed by another incubation time of 24 hours. Then 22 µL of the cell

viability reagent was added to the plate and after another 2 hours of incubation measurement was done with a fluorescence plate reader at 560/590nm (SpectraMax M3, Molecular Devices, USA).

2.3.5. Fiber Challenge and Cell Migration Assay

NR8383 rat macrophages (3×10^6 cells/mL) were suspended with a vortex in 1 mL Ham's F12 medium containing 15% FCS, 2 mM L-glutamine, 100 µg/mL penicillin, and 100 U/mL streptomycin and culture medium was added to a final volume of 3 mL. The surface area of the cell culture flasks was 12.5 cm², hence 240,000 cells/cm² cell culture dishes were seeded. The fibers were suspended in 1 mL culture medium Ham's F12 containing 15% FCS, 2 mM L-glutamine, 100 µg/mL penicillin, and 100 U/mL streptomycin and added to the cell culture flasks. The assay can be performed in a smaller volume at constant surface volume ratio. A sample in which cells without particles were incubated served as negative control. The subsequent experiments were repeated up to concentrations which gave the maximum induction of chemotaxis. Incubation of the cells with the fibers and particles was performed at 37°C, 100% humidity and 5% CO₂ for 16 hours. Thereafter, the cells were transferred to 5 mL Eppendorf vials. Cells were removed by centrifugation with 400 g for 5 minutes and particles were removed by centrifugation with 15,000 g for 10 minutes at room temperature. The supernatants were immediately used for the migration tests.

2.3.6. Chemotaxis Assay

Cell migration was investigated according to Boyden [20], with the modifications described previously [17] [18]: 200,000 unchallenged dHL-60 cells were F12 or RPMI 1640, respectively without FCS and seeded in each plate well insert (THINCERT, 3 µm pore size, Greiner bio-one, Frickenhausen, Germany) that were placed in the cavities of 24 black well plates (Krystal, Duna Labortechnik, Asbach, Germany). 500 µL of the supernatants of the particle-challenged NR8383 cells were added to the lower chamber. Migration of dHL-60 cells across the membrane was performed at 37°C, 100% humidity and 5% CO₂ for 24 hours. For calibration, 0 to 100,000 HL-60 cells were seeded directly into four plate wells that were left without inserts.

Staining of migrated cells and of the cell calibration was performed with Calcein-AM for 60 minutes at 37°C, 5% CO₂ and 100% humidity by adding 500 µL Calcein-AM in the plate wells (>90% HPLC, Sigma-Aldrich, Steinheim, Germany). Calcein-AM was delivered as 4 mM solution in DMSO, stored in aliquots at -18°C and diluted to a final concentration of 4 µM in PBS.

The cell suspensions were removed from the plate wells and collected with 400 g for 5 minutes at room temperature. 850 µL of the supernatant were discarded while the cells were re-suspended in the remaining volume of 150 µL. In addition, the adherent cells at the outside of the inserts were detached by adding 500 µL trypsin/EDTA (ethylene diamine tetraacetic acid) (0.05%/0.02%, PAN-Biotech GmbH, Aidenbach, Germany) for 10 minutes at 37°C, 5% CO₂ and 100% hu-

midity. Subsequently, the inserts were removed from the plate wells. The 150 μL of the collected cells were added into the plate wells that contained the 500 μL of the trypsin/EDTA detached cells. The cell count was determined by fluorescence measurement at 490/520nm and calculated from the cell calibration (SpectraMax M3, Molecular Devices, USA).

2.3.7. Quantitative SEM Analysis of WHO Fiber Fragments

In order to determine the number of airborne fibers that meet the WHO convention in terms of particularly hazardous fibers, air samples have been collected by using a FAP fiber sampling device (GSA Messgerätebau GmbH, Germany) at a distance of approximately 35 cm from the feeding shaft of the shredder. A gold-coated polycarbonate filter with a diameter of 37 mm and a pore size of 0.8 μm has been used for these investigations. The air flow is 2.0 L/minute and the collection time is 15 minutes in each case; only for the tests with high rotational speed and small screen size has the sampling time been reduced to 10 minutes. The fibers on the gold-coated filters have then been counted by using a scanning electron microscope (SEM) S-2300 (Hitachi, Ltd., Tokyo, Japan) including an energy dispersive X-ray analysis (EDX) and the resulting fiber concentration has been calculated according to the guideline of the DGUV [21]. For this purpose, a Kevex Si (Li) detector system (eumeX Instrumentebau GmbH, Heidenrod, Germany) at 1000 \times magnification was used.

3. Results and Discussion

3.1. Effect of Shredding Parameter on Fiber Dust Formation

The following figures show the effect of different rotational speeds and feedstock sizes on the particle size distribution of the ground material (**Figure 1**) as well as on the fiber length distribution in the fiber dust after the ashing process (**Figure 2**). It can be seen, that most of the particles (at least 65%) have a particle size bigger than 1 mm. The relative frequency decreases with decreasing particle size but slightly increases again for the particles < 0.125 mm. For every parameter combination, the relative frequency is higher compared to the fraction > 0.125 mm. The fiber length distribution shows a small amount of fibers with a length < 30 μm , whereas the fibers with a length of 30 - 60 μm provide the highest frequency. With increasing fiber length, the fiber length distribution shows a decreasing trend.

No significant effect of the feedstock size on the particle size distribution of the ground material can be observed (**Figure 1**). In the fiber length distribution (**Figure 2**), a tendency of less short fibers (< 60 μm) can be seen when shredding the organic sheets with a smaller feedstock size. This is in accordance to the percentage of fibers in the short particle fraction in **Figure 3**. Here, shredding the pieces with a feedstock size of 30 \times 30 mm leads to a higher percentage of fiber mass, except for the high rotational speed of 2500 rpm. Material with a smaller feedstock size needs fewer cutting steps until it is small enough to leave the cutting chamber through the screen. This could explain the smaller amount of short

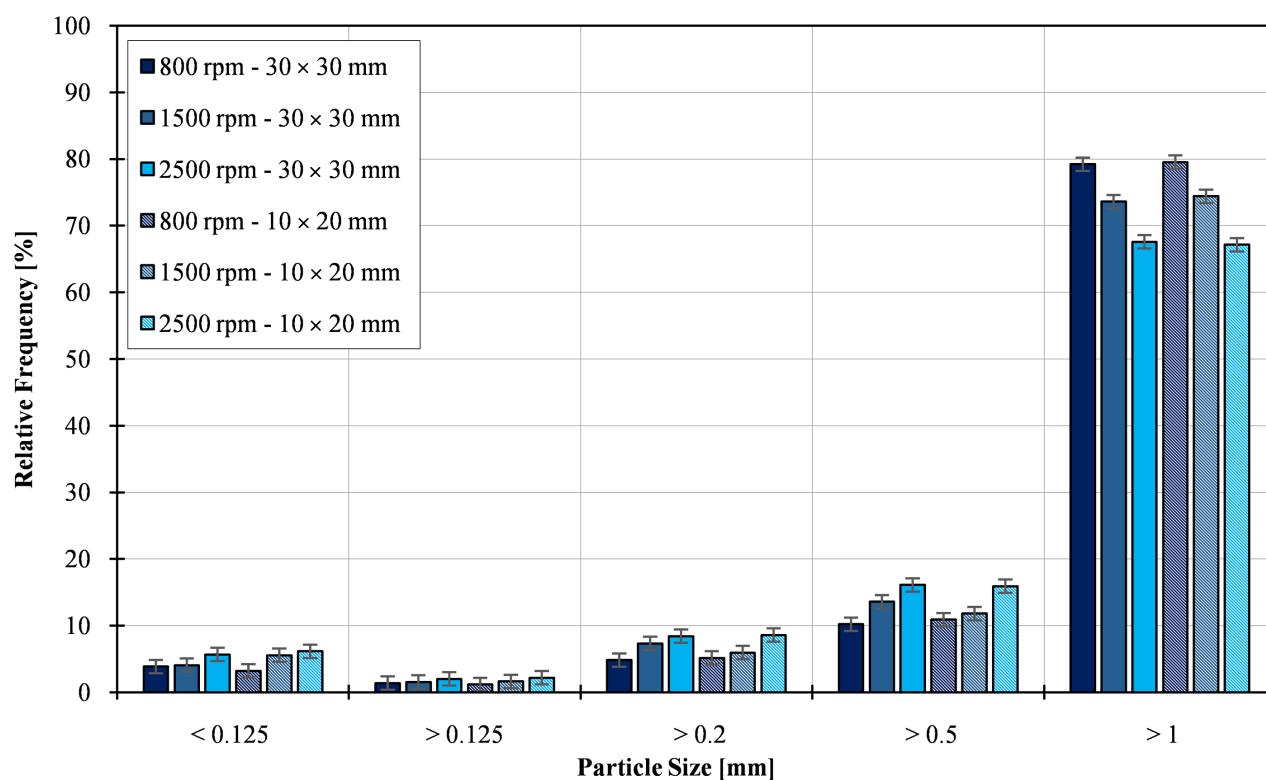


Figure 1. Particle size distribution of the ground material from shredding PC-CF organic sheets with different rotational speeds and feedstock sizes.

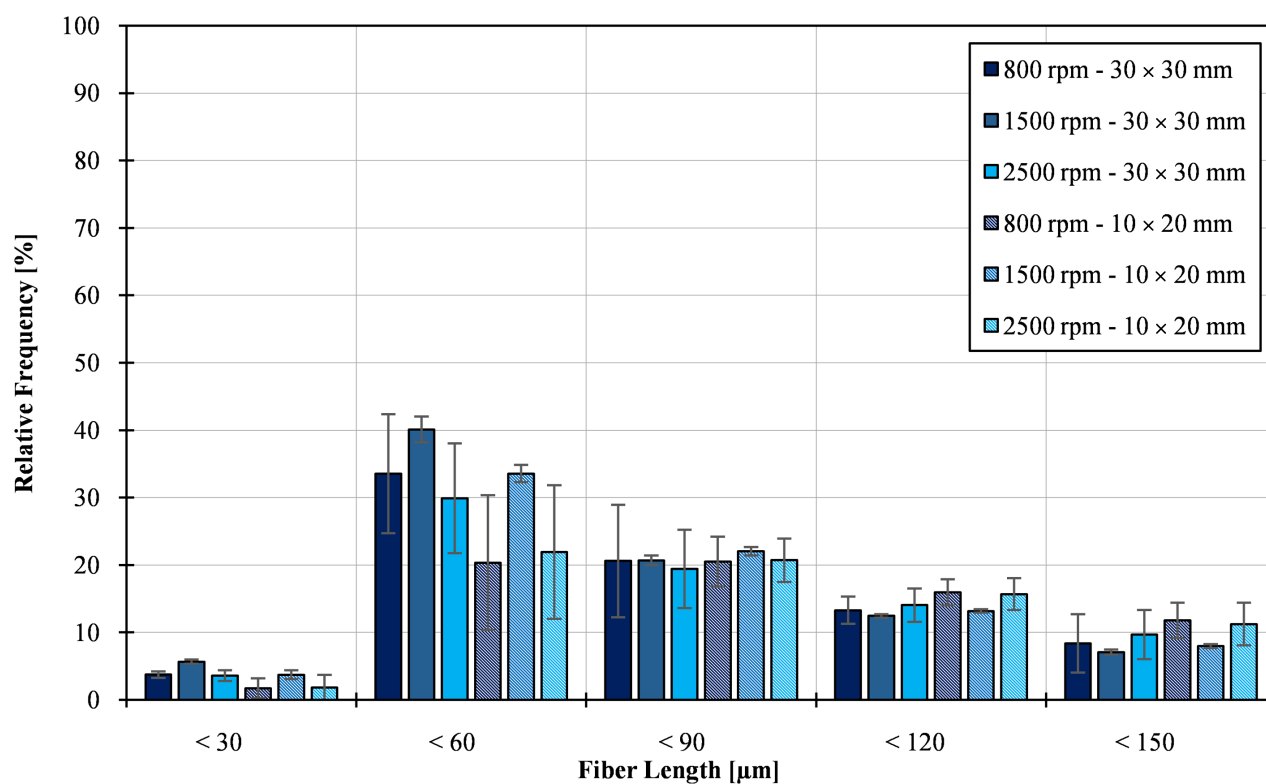


Figure 2. Fiber length distribution of the fiber dust of the ground material from shredding PC-CF organic sheets with different rotational speeds and feedstock sizes.

fibers while shredding material with a smaller feedstock size. On the other hand, a smaller feedstock size comes along with a higher number of pieces for the 50 g sample. Therefore, the particle-particle interaction in the cutting chamber is enhanced, which could be a reason why the favorable effect of the smaller feedstock size can just slightly be seen in the fiber length in **Figure 2**. In a similar investigation, Shuaib and Mativenga [22] also found no significant effect of the feedstock size on the fiber length and in [23] it was found that the effect of the feedstock size on the particle mass and the fiber length is material dependent.

Rising the rotational speed during shredding leads to a higher number of small particles (<0.125 mm) independent of the feedstock size (**Figure 1**). This can be seen in the other fractions as well. Polycarbonate is a thermoplastic material and therefore provides viscoelastic material properties. With increasing load speed, the capability to creep as well as the stress relaxation is reduced so that the material properties become more brittle [6]. A more brittle fracture behavior leads to uncontrolled crack propagation [24] and thus a broader particle distribution with a large amount of small particles [25]. In brittle fracture, the crack propagates through the material, so that uncontrolled fracture behavior is dominant. This is the reason for the increased quantity of fine particles with higher rotational speed. In **Figure 2**, no clear effect of the increased rotational speed on the fiber length distribution can be seen. Nevertheless, the percentage of fibers in the fraction < 0.125 mm increases with rising rotational speed (**Figure 3**), which is in accordance to the findings from the particle size distribution and is a result of the more brittle breaking behavior. This suggests that a higher rotational speed increases the number of fine particles in the ground material, but has no effect on the fiber length distribution in the dust. The absolute number of short fibers is enhanced, but the relative frequency is not significantly altered.

The investigations on shredding the PC-CF organic sheets with the single screw shredder enable the analysis of the airborne particle concentrations. The results are represented in **Table 1**, where the nanoparticle concentration was measured with the SMPS, the microscale particle concentration measured with the APS and the fiber dust mass, collected on the filter of the VC25-Sampling device, released during the shredding of PC-CF organic sheets on the WC 250-400 single-shaft shredder. On the nanometer scale, the mass-based particle concentration shows just a slight effect of an increased rotational speed with a big screen size. On the other hand, on the micrometer scale, an increased rotational speed leads to a significantly higher particle concentration for the investigations with a big screen size. Here again, the increased load speed promotes brittle material fracture and thus leads to uncontrolled breaking with a high number of small particles. This effect can be seen as well for the E-dust mass deposited on the filter.

The sample, which is shredded at high speed and with small screen size, shows increased particle concentrations in the nanometer scale compared to the shredding with a large screen size. Reducing the screen size leads to a smaller maximum

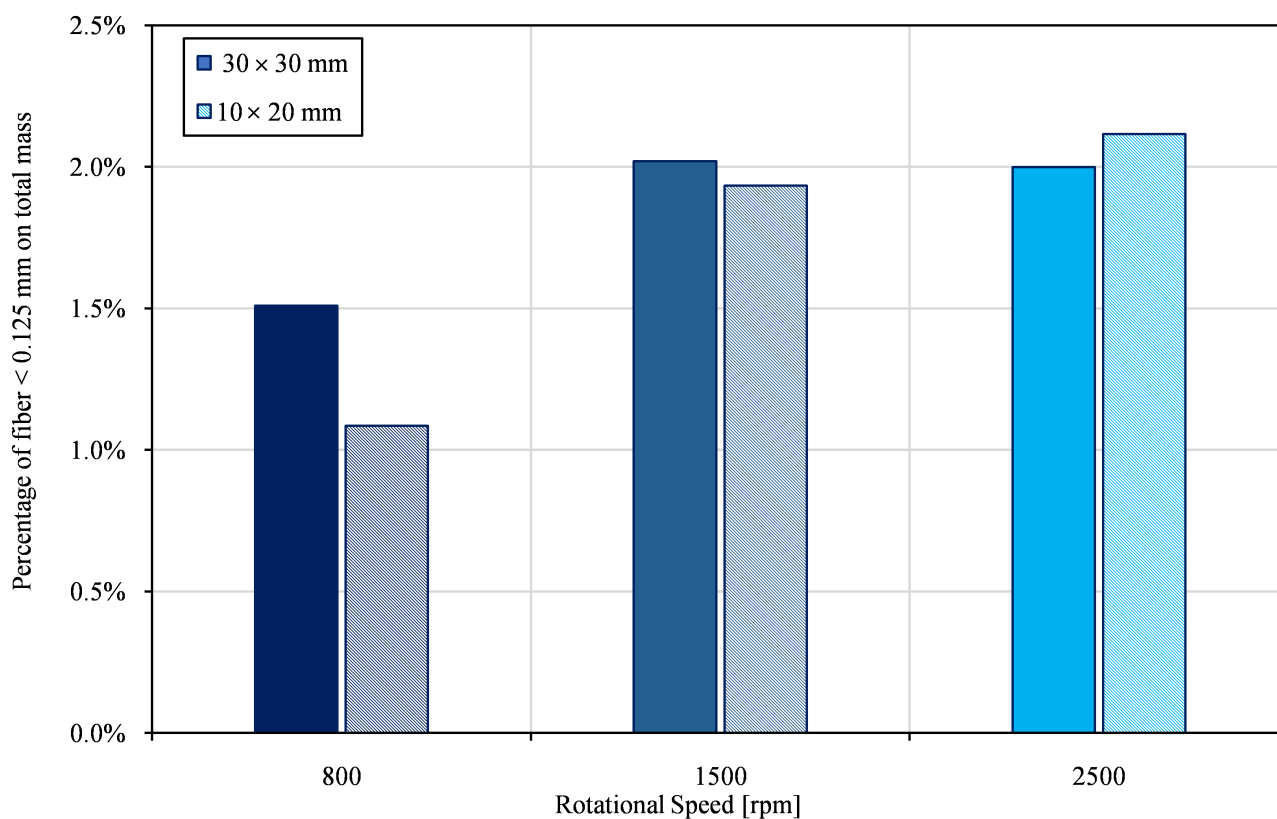


Figure 3. Percentage of fiber mass after ashing the particle fraction < 0.125 mm on the total ground material mass over the rotational speed while shredding. The two different feedstock sizes are presented.

Table 1. Particle mass concentrations according to SMPS, APS, and the fiber dust mass collected on the filter of the VC25-Sampling device. The absolute particle concentration is shown with the respective standard deviation.

	Low Rotational Speed & Big Screen Size	High Rotational Speed & Big Screen Size	High Rotational Speed & Small Screen Size
Nanoscale Particle Concentration [$\mu\text{g}/\text{m}^3$]	3.067 ± 0.61	3.54 ± 0.57	5.59 ± 4.3
Microscale Particle Concentration [mg/m^3]	0.76 ± 0.63	3.79 ± 7.36	0.93 ± 1.02
Inhalable E-dust mass [$\text{g}/\text{kg}^*\text{m}^3$]	0.0003	0.0004	0.0017

particle size of the ground material and to a prolonged cutting duration until the material is small enough to pass through the screen. In the micrometer scale, this can not be observed due to the high concentration of $3.79 \text{ mg}/\text{m}^3$ while shredding with high rotational speed and big screen size. The high standard deviation of this sample results from a high concentration of long fibers in the air which partly blocks the inlet of the APS. However, the decrease in screen size leads to a huge increase in the collected inhalable dust (E-dust) mass compared to the shredding with a larger screen mesh size. This high dust mass supports the observations during the shredding process that high dust masses are released due to the combined effects of the high rotational speed and the small screen size.

It can be concluded, that the investigations in the cutting mill as well as with

the single screw shredder show an increased fiber dust formation when shredding at high rotational speed as well as when reducing the screen size. The feedstock size does not have a significant effect on the fiber length distribution but on the released fiber mass.

3.2. Toxicological Investigations

3.2.1. Cytotoxicity

The AlamarBlue test was used to determine cytotoxic effects of the particles as an indicative measurement. The test measures the cells' energy charge as a measure of toxicity using the fluorescent dye and redox indicator resazurin. A decrease in fluorescence indicates cytotoxicity. Fluorescence is plotted against the concentration of particles added to the cells.

Initial investigations were carried out with the cutting mill (Figure 2) in order to determine the conditions for the large-scale test with the single-shaft shredder (Table 1). Increasing the rotational speed and feedstock size did not lead to any changes in the toxicity using the cutting mill (Figure 4).

Since cytotoxicity remains unchanged for all examined parameters and rotational speed, feedstock size and screen size were the main influencing factors concerning the particle quantity, these were varied for the large-scale investigation using the single-shaft shredder. Even under these conditions, the AlamarBlue test showed no increased cell toxicity, even at very high dust concentrations (Figure 5).

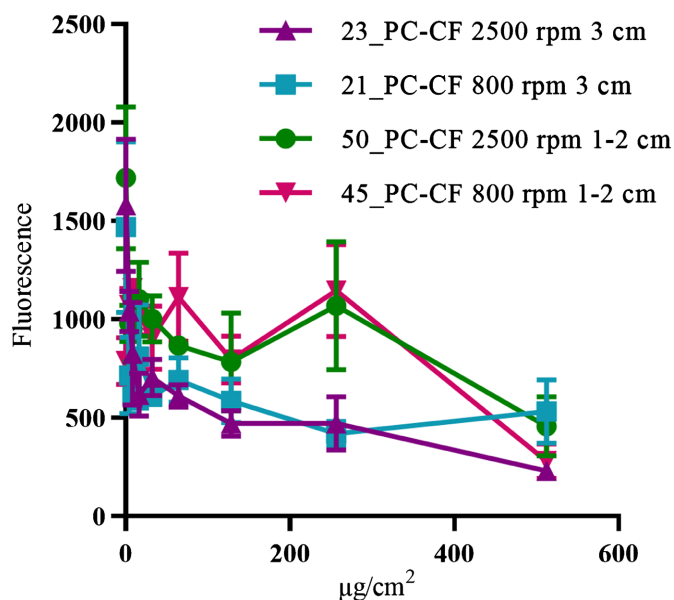


Figure 4. Cell toxicity determined with the AlamarBlue assay for ground PC-CF organic sheets that has been shredded at different rotational speeds and different feedstock size (1 - 2 cm or 3 cm) in the cutting mill. The results of three experiments carried out independently in terms of time ($N = 3$) are shown in each case. The fluorescence is plotted against the amount of CF particles ($\mu\text{g}/\text{cm}^2$) exposed to the macrophages. Cell toxicity can only be determined for the highest added quantity of $500 \mu\text{g}/\text{cm}^2$. As this is a very large amount, the observed toxicity is considered to be very low.

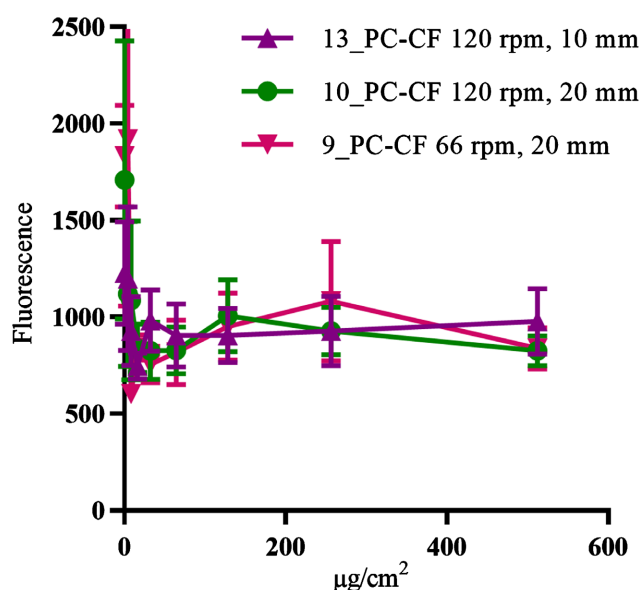


Figure 5. AlamarBlue assay for airborne dust samples from ground PC-CF organic sheets that were shredded with the single-shaft shredder at different rotational speeds and screen size. The results of three experiments carried out independently in terms of time ($N = 3$) are shown in each case. The fluorescence is plotted against the amount of CF particles ($\mu\text{g}/\text{cm}^2$) exposed to the macrophages. No cell toxicity is evident up to and including the highest level tested.

3.2.2. Investigation of the Induced Cell Migration by Particles Obtained from the Large-Scale Test with the Single-Shaft Shredder

Rotational speed, feedstock size and screen size were the main influencing factors concerning the particle quantity and the AlamarBlue did not show any significant cytotoxic effects, regardless of how they were obtained. Particle induced cell migration was therefore investigated for particles that were obtained for different speed and feedstock size in the large-scale test with the single-shaft shredder (Table 1). Particle induced cell migration was similar for all examined parameters. The PICMA, therefore, shows no increased effects, even at very high dust concentrations as well as the AlamarBlue test (Figure 6).

Comparison with the historical control (Figure 6 and Figure 7) shows that the induction of cell migration by carbon fibers is very weak and comparable to that of inert particles such as barium sulfate. Consistent with the results from the cell toxicity test, the migration test provides no evidence that the inflammatory or toxic effects of carbon fibers are significantly influenced by the type of shredding.

3.2.3. Quantitative Analysis of WHO Fibers Resulting from Shredding PC-CF with a Single-Shaft Shredder

High fiber concentrations have been counted for all shredding parameters, as well as a high proportion of WHO fibers (Table 2). Figure 8 presents an example of the collected fibers on the gold-coated core pore filter after shredding with different parameters.

EDX analysis of the collected fibers detected only the element carbon in addition to the sputtering material gold (Figure 9).

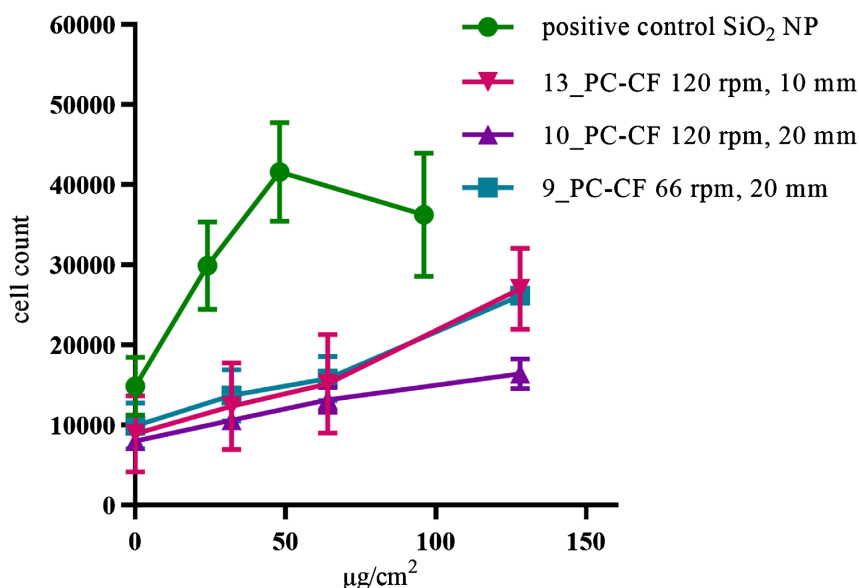


Figure 6. PICMA for airborne dust samples of ground PC-CF organic sheets that were shredded at different rotational speeds and different screen sizes. The positive control silica nanoparticles (SiO_2 NP) ensure comparability with the historical controls.

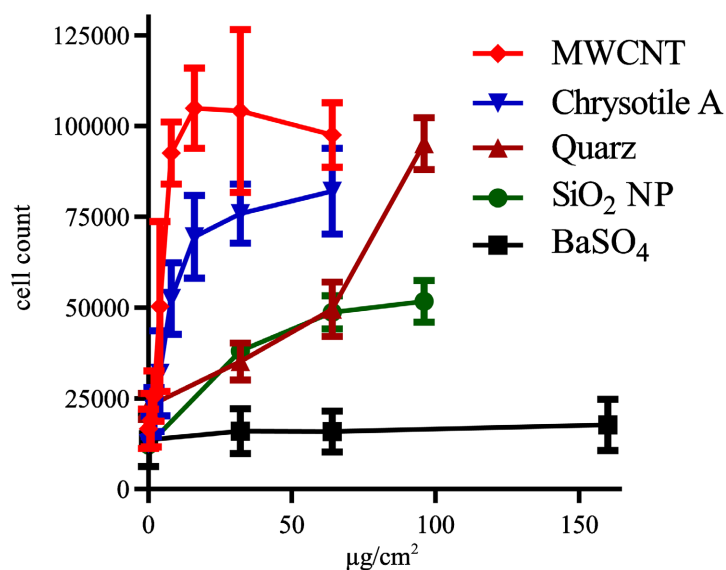


Figure 7. Historical results for the PICMA for particles of different inflammatory effects. Very effective: Multiwalled Carbon Nanotubes (MWCNT) and asbestos (Chrysotile A). Quartz and silica have an intermediate effect (the latter is always included as a positive control to ensure comparability). Inert: barium sulfate (BaSO_4). The results of three experiments carried out independently of one another in terms of time ($N = 3$) are shown. The number of migrated dhL-60 cells (cell count) is plotted against the amount of particles ($\mu\text{g}/\text{cm}^2$) to which the macrophages were exposed.

Table 2. Total fiber concentration and WHO-fiber Concentration of the airborne fiber dust formed during shredding with different parameters.

	Low Rotational Speed & Big Screen Size	High Rotational Speed & Big Screen Size	High Rotational Speed & Small Screen Size
Total Fiber Concentration [F/m^3]	7,100,000	7,500,000	15,500,000
WHO Fiber Concentration [F/m^3]	240,000	225,000	240,000

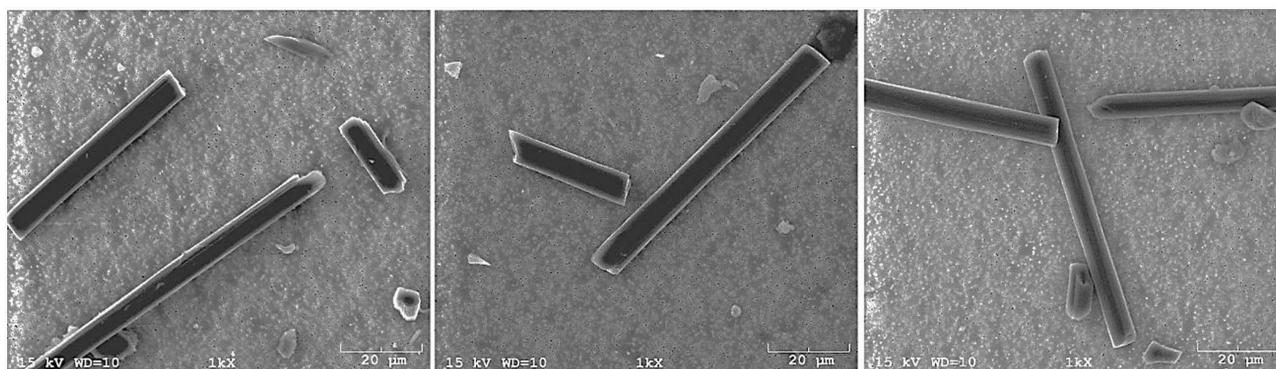


Figure 8. SEM images of detected fibers after shredding with different parameters. Low rotational speed & Big screen size (left), High rotational speed & Big screen size (middle), High rotational speed & Small screen size (right). Magnification 1000×.

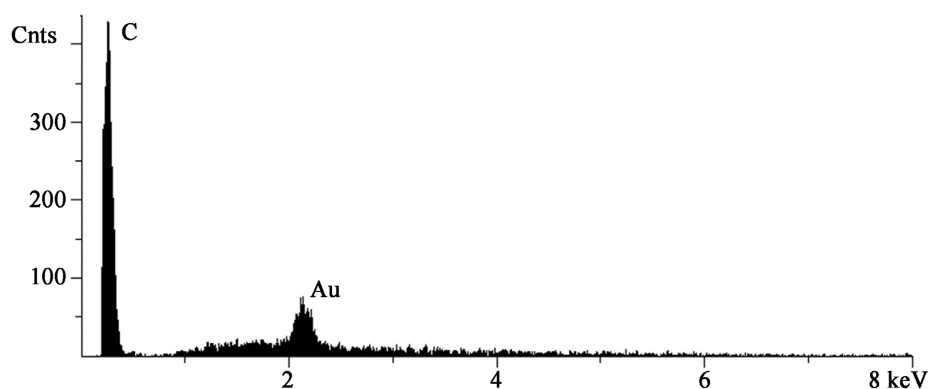


Figure 9. Representative EDX-spectrum of carbon fibers after shredding.

The results support the findings from the investigations in the micro- and nanoscale range that higher dust concentrations are produced during shredding at high rotational speed. The additional reduction in screen size significantly increases the amount of fibers. Furthermore, it is shown that the number of WHO fibers is not affected by the shredding parameters. This is in accordance with the results from the toxicological studies (Figures 4-6). Despite the different total fiber concentrations, no influence of the shredding parameters on toxicity can be detected here, since the potentially toxic WHO fiber fragments do not differ significantly in number.

Formally, the detected carbon fiber fragments meet the fiber definition of the legislator. However, carbon fibers currently have no specific limit value. Prevention is based on the provisions of TRGS 521 [26]. Thus, a protective measure concept takes effect that is based on the decision concentrations of 50,000 and 250,000 F/m³ (no limit values). In the future, the adaptation to this protection concept should be based on the actual health risk from carbon fiber dust.

4. Conclusions

Irrespective of the shredding parameters, fiber fragments of different dimensions are produced, but they are of low toxicity and have very little impact on cell migration. This indicates minor inflammable effects of the dust formed after inha-

lation. This is in accordance with the results of the animal experiments that report rather low toxicity. As a result, the optimization of the shredding processes should mainly focus on the amount of particles and their dimensions. The biological data indicate as well that the WHO fibers resulting from the shredding are only a minor health hazard. Most of these fibers are probably too thick to have any appreciable toxicity. Slight differences in the induction of cell migration are visible, but to an extent that it is not important to take them into account for the risk assessment. However, a pronounced bio-persistence of inhaled CFRP fragments must be assumed. Therefore, exposure to CFRP dust should be minimized in terms of preventive health protection.

In summary, it can be stated that during shredding fiber-reinforced thermoplastics, inhalable fiber fragments can arise, which probably do not pose any health risks that go well beyond that of granular dust. The proportion of these fibers can be reduced by a low speed or a large screen size. Especially when cutting carbon-fiber-reinforced thermoplastics and materials with a brittle matrix, special safety precautions should be taken, even if the dust produced shows only low cell-toxic effects in the tests.

Acknowledgements

We would like to thank the German Federation of Industrial Research Associations "Otto von Guericke" e.V. (AiF) for the financial support of the research project, which is funded by the Federal Ministry for Economic Affairs and Climate Action (BMWK).

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

References

- [1] European Agency for Safety and Health at Work: Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH). REACH, vom Regulation (EC) No. 1907/2006.
- [2] Howarth, J., Mareddy, S.S. and Mativenga, P.T. (2014) Energy Intensity and Environmental Analysis of Mechanical Recycling of Carbon Fibre Composite. *Journal of Cleaner Production*, **81**, 46-50. <https://doi.org/10.1016/j.jclepro.2014.06.023>
- [3] Li, X., Bai, R. and McKechnie, J. (2016) Environmental and Financial Performance of Mechanical Recycling of Carbon Fibre Reinforced Polymers and Comparison with Conventional Disposal Routes. *Journal of Cleaner Production*, **127**, 451-460. <https://doi.org/10.1016/j.jclepro.2016.03.139>
- [4] Li, H. and Englund, K. (2017) Recycling of Carbon Fiber-Reinforced Thermoplastic Composite Wastes from the Aerospace Industry. *Journal of Composite Materials*, **51**, 1265-1273. <https://doi.org/10.1177/0021998316671796>
- [5] Seidel, S. (2021) Organoblech-Verschnitte recyceln. Faserverstärkte Thermoplaste mechanisch recyceln und in die Produktion zurückführen. *Kunststoffe*, **1**, 25-27.
- [6] Schönert, K. and Schubert, G. (2002) Comminution Equipment for Non-Brittle

Waste and Scrap. *Aufbereitungs Technik*, **9**, 6-23.

- [7] IARC. International Agency for Research on Cancer (2002) Man-Made Vitreous Fibres. Unter Mitarbeit von WHO. World Health Organization. 81. Aufl. IARC Press, Lyon.
- [8] IARC. International Agency for Research on Cancer (2012) A Review of Human Carcinogens. Part C: Arsenic, Metals, Fibres and Dusts. 100C. WHO Press, Lyon.
- [9] Donaldson, K., Murphy, F.A., Duffin, R. and Poland, C.A. (2010) Asbestos, Carbon Nanotubes and the Pleural Mesothelium: A Review of the Hypothesis Regarding the Role of Long Fibre Retention in the Parietal Pleura, Inflammation and Mesothelioma. *Particle and Fibre Toxicology*, **7**, Article No. 5.
<https://doi.org/10.1186/1743-8977-7-5>
- [10] Schinwald, A. and Donaldson, K. (2012) Use of Back-Scatter Electron Signals to Visualise Cell/Nanowires Interactions *in Vitro* and *in Vivo*; Frustrated Phagocytosis of Long Fibres in Macrophages and Compartmentalisation in Mesothelial Cells *in Vivo*. *Particle and Fibre Toxicology*, **9**, Article No. 34.
<https://doi.org/10.1186/1743-8977-9-34>
- [11] Boatman, E.S., Covert, D., Kalman, D., Luchtel, D. and Omenn, G.S. (1988) Physical, Morphological, and Chemical Studies of Dusts Derived from the Machining of Composite-Epoxy Materials. *Environmental Research*, **45**, 242-255.
[https://doi.org/10.1016/S0013-9351\(88\)80050-1](https://doi.org/10.1016/S0013-9351(88)80050-1)
- [12] Holt, P.F. and Horne, M. (1978) Dust from Carbon Fibre. *Environmental Research*, **17**, 276-283. [https://doi.org/10.1016/0013-9351\(78\)90030-0](https://doi.org/10.1016/0013-9351(78)90030-0)
- [13] Owen, P.E., Glaister, J.R., Ballantyne, B. and Clary, J.J. (1986) Subchronic Inhalation Toxicology of Carbon Fibers. *Journal of Occupational Medicine. Official Publication of the Industrial Medical Association*, **28**, 373-376.
- [14] Warheit, D.B., Hansen, J.F., Carakostas, M.C. and Hartsky, M.A. (1994) Acute Inhalation Toxicity Studies in Rats with a Respirable-Sized Experimental Carbon Fibre: Pulmonary Biochemical and Cellular Effects. *The Annals of Occupational Hygiene*, **38**, 769-776.
- [15] Zhang, Z., Wang, X., Lin, L., Xing, S., Wu, Y., Li, Y., *et al.* (2001) The Effects of Carbon Fibre and Carbon Fibre Composite Dusts on Bronchoalveolar Lavage Component of Rats. *Journal of Occupational Health*, **43**, 75-79.
<https://doi.org/10.1539/joh.43.75>
- [16] Landsiedel, R., Ma-Hock, L., Hofmann, T., Wiemann, M., Strauss, V., Treumann, S., *et al.* (2014) Application of Short-Term Inhalation Studies to Assess the Inhalation Toxicity of Nanomaterials. *Particle and Fibre Toxicology*, **11**, Article No. 16.
<https://doi.org/10.1186/1743-8977-11-16>
- [17] Westphal, G.A., Rosenkranz, N., Brik, A., Weber, D., Föhring, I., Monsé, C., *et al.* (2019) Multi-Walled Carbon Nanotubes Induce Stronger Migration of Inflammatory Cells *in Vitro* than Asbestos or Granular Particles but a Similar Pattern of Inflammatory Mediators. *Toxicology in Vitro*, **58**, 215-223.
<https://doi.org/10.1016/j.tiv.2019.03.036>
- [18] Westphal, G.A., Schremmer, I., Rostek, A., Loza, K., Rosenkranz, N., Brüning, T., *et al.* (2015) Particle-Induced Cell Migration Assay (PICMA): A New *in Vitro* Assay for Inflammatory Particle Effects Based on Permanent Cell Lines. *Toxicology in Vitro*, **29**, 997-1005. <https://doi.org/10.1016/j.tiv.2015.04.005>
- [19] Breitman, T.R., Selonick, S.E. and Collins, S.J. (1980) Induction of Differentiation of the Human Promyelocytic Leukemia Cell Line (HL-60) by Retinoic Acid. *Proceedings of the National Academy of Sciences of the United States of America*, **77**, 2936-2940.

- <https://doi.org/10.1073/pnas.77.5.2936>
- [20] Boyden, S. (1962) The Chemotactic Effect of Mixtures of Antibody and Antigen on Polymorphonuclear Leucocytes. *The Journal of Experimental Medicine*, **115**, 453-466. <https://doi.org/10.1084/jem.115.3.453>
- [21] DGUV (2014) DGUV-Information: Bearbeitung von CFK Materialien. Orientierungshilfe für Schutzmaßnahmen. Hg. v. Fachbereich Holz und Metall der DGUV.
- [22] Shuaib, N.A. and Mativenga, P.T. (2016) Effect of Process Parameters on Mechanical Recycling of Glass Fibre Thermoset Composites. *Procedia CIRP*, **48**, 134-139. <https://doi.org/10.1016/j.procir.2016.03.206>
- [23] Tölle, L. and Hopp, M. (2022) Influence of Process Parameters on the Formation of Inhalable Fiber Dust during Shredding for Mechanical Recycling of Fiber-Reinforced Organo Sheets. *Journal of Applied Polymer Science*, **139**, e52646. <https://doi.org/10.1002/app.52646>
- [24] Ebert, R., Krampitz, T. and Lieberwirth, H. (2015) Grundlagenuntersuchungen zur Zerkleinerung nicht-spröder Leichtbauwerkstoffe mittels Scher-Reißbeanspruchung. *Recycling und Rohstoffe*, **8**, 563-576.
- [25] Wolters, L., Von Marvick, L., Regel, K., Lackne, V. and Schäfer, B. (1997) Kunststoff-Recycling. Grundlagen Verfahren-Praxisbeispiele. Hanser, München.
- [26] TRGS 521: Technische Regeln für Gefahrstoffe-Faserstäube.