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## ABSTRACT

Sustainable and microbiologically secure foodstuff production lines are of increasing scientific interest and are in the focus of recent research programs. Additionally, they are of great importance for the production industry due to the prevention of food-borne illnesses caused by pathogens such as *Salmonella* sp., *Listeria monocytogenes*, or *Escherichia coli*. These pathogens are responsible for production losses, loss of customer acceptance, and severe food-borne illnesses. A pathogenic threat is frequently combated with sanitizing steps of the production lines. For conveyor band cleaning, this study compares the cleaning abilities of nitric acid ($HNO_3$) and plasma treated water (PTW), which have been sprayed via a commercially available nozzle on two different polymeric surfaces (polysiloxane and polyurethane). Additionally, the cleaning agents $HNO_3$ and PTW have been characterized through their pH and their conductivity. These findings have been underpinned by experiments that focus on a possible influence of nozzle abrasion, such as brass and stainless-steel nanoparticles, on the antimicrobial potential of PTW and $HNO_3$. Adversely acting effects like an enhanced abrasion of conveyer band materials due to PTW or $HNO_3$ treatment have been checked by using light microscopic micrographs and topographic scans in high-resolution mode. Based on the presented results of the experiments, the suitability of an in-place sanitation step in foodstuff production lines has been demonstrated on a laboratory scale.



## I. INTRODUCTION

Producers of the European Union provide several prevention strategies to avoid and/or combat any contamination along the value chain of fresh food. Additionally, a risk reduction of a pathogenic contamination is ensured by following internationally established concepts such as good agricultural practice, good manufacturing practice, and the HACCP concept (hazard analysis and critical control points). However, various factors during the production processes of fresh food become sources of contamination along the entire value chain. Therefore, the production and processing of fresh food places high hygienic demands on production facilities. At the same time, since the production of food requires an exceedingly high amount of water, the conservation of natural resources like water must be considered. Moreover, the cleaning of the production environment, i.e., the production premises and the equipment, consumes a majority of the water resources.[1,2] Therefore, a need for more sustainable cleaning procedures becomes obvious in the face of costly and scarce resources.

Microbial contamination is found along the entire food value chain, particularly necessitating focused efforts on the internal process hygiene, as possible undetected cross contamination poses a potential hazard. Besides raw material, food-contacting surfaces primarily contain sources of severe contamination. Thus, food production frequently embraces wet chemical sterilization and disinfection processes to ensure the safety of the produce, which consequently also includes the consumer. High requirements pose a challenge to materials that are exposed to food. For example, conveyor belts must be produced from corrosion-resistant, non-toxic, and easy to clean and disinfecting materials.[3] They must be non-adsorbent, because this characteristic ideally prevents transfer of odors, colors, and flavors, and any other impurities. Additionally, high requirements are also placed on cleaning agents when they are exposed to the food itself.[4] Certainly, the cost-intensive and resource-consuming cleaning and disinfection agents lower the profit margin of the producer through production downtime during the often long cleaning procedures. Furthermore, since highly contaminated wastewater may lead to an ecological problem, a high throughput of disinfectants confronts the producer with the need for building more storage facilities and setting up a distinct effluent management system. Some disinfectants might also harm the material surfaces of the conveyor belts and as a result promote the formation of undetected biofilms through altered surface properties.

Cleaning procedures are an interplay of the cleaning target, the constructive avoidance of inaccessible areas, possible systemic parameters such as pressure or temperature, and the access time for the entire cleaning step.[5] All steps of such a procedure bear the risk of cross contamination or leftover residuals of pathogens. For instance, conveyor belts are usually not dismantled for cleaning, which supports a possible risk of microbial adhesion and biofilm formation. Of course, the right choice of a sanitizer or a disinfectant complements the setup of a cleaning procedure, and distinct cleaning demands can be fulfilled by a specific composition of sanitizers. For instance, the adhesion of pathogens can be prevented by choosing efficient and sustainable agents with antimicrobial effects.[6,7] The various possibilities for the sustainable use of disinfectants additively challenge producers. Nevertheless, in food production, most conventional or new disinfection methods for conveyor belts have so far been limited. They are often either insufficient for cleaning and disinfection or they are environmental unfriendly in nature.[5,8] Usually, conditions are created where producers often face the challenge of choosing between using methods that produce sufficiently high antimicrobial cleaning results and using sustainable alternatives. In this context, non-thermal atmospheric-pressure plasmas open a novel possibility for resource saving and sustainable processing, since the active components are formed by an easily controllable plasma input. Several studies have proved the efficiency of plasma applications for biological decontamination of surfaces.[9–11] Therefore, plasma technologies and PTW open new perspectives to supplement a potpourri of sustainable cleaning methods.

The aim of this presented work is to demonstrate the compatibility of plasma-treated water (PTW) with materials frequently used in industrial production. Concretely, the interplay of PTW and two different surfaces (a SI surface with a foamier appearance and an ultra-hydrophobic PU surface) has been screened in comparison with nitric acid, which is frequently used for surface decontamination purposes in the food industry, and water. Against the background of possible cross reactions of PTW with its surrounding materials, the anti-microbial efficacy of plasma treated water has been additionally tested. We hypothesize that no changes in the sanitizing performance are observable in comparison with those of established sanitizers. We discuss whether in a sustainable and environmentally friendly production line, non-thermal plasma technology may become a major player. It offers an optimal tool for "on demand" cleaning procedures and may complement or replace disinfectants and sanitizers that are still in use today. We aim to introduce PTW under the catchphrase "reduce and refine."

## II. METHODOLOGY

### A. Plasma source and PTW generation

PTW was generated via the PLexc$^2$ microwave-based plasma source (2.45 GHz). The PLexc$^2$ plasma source consists of a second stage, in contrast to the single stage PLexc,[9,12] which is a basic lab-scaled predecessor. The second stage of the source is a prerequisite to produce plasma-processed gaseous compounds for PTW production on a much higher scale to meet the demands of professional production lines. The source's operation principle and all process parameters are explicitly described in Refs. 13 and 14 and briefly summarized in Table I and Fig. 1. The plasma chemistry, which predominantly governs the antimicrobial activity of PTW, is based on reactive nitrogen and, to a lesser extent, reactive oxygen species (RONS), which in terms of the nitric part of the RONS primarily react to nitric acid in aqueous solutions. Nitric acid is identified as the major compound in terms of combating pathogens. Thus, conventional nitric acid was used for comparison. Nevertheless, the distinct chemical processes that underlie the antimicrobial activities are described in greater detail by authors in Refs. 15 and 16.

**TABLE I.** Technical source parameters for the experiments to generate plasma processed air (PPA) and the functionalization of distilled water to plasma treated water (PTW).

| | 1st stage (PLEXC) | 2nd stage (PLEXC$^2$) |
|---|---|---|
| Power (P) (kW) | 1.3 | 3.0 |
| Frequency (f) (GHz) | 2.45 | 2.45 |
| Volume flow rate (slm) | 12 | 60 |
| Gas | Air | Air |

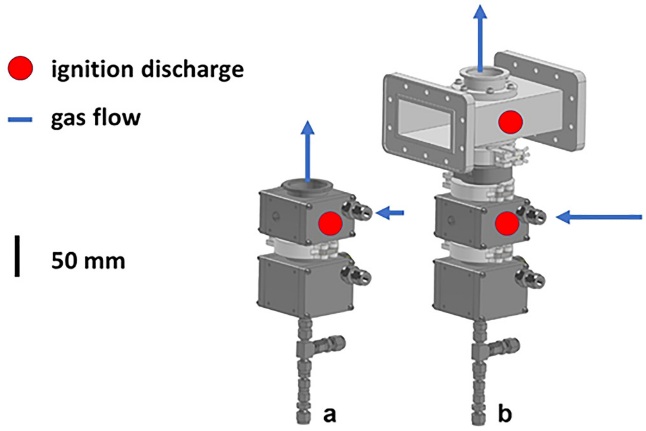


FIG. 1. Schematic diagram of the microwave plasma sources PLexc (a) and PLexc2 (b), the latter was used for the experiments described. The red dots highlight the structures, where the ignition discharges take place. The discharges are plasma torches that are driven by the gas flow. A power supply connector for a coax cable is located at the back of the sources. Technical specifications of both sources are summarized in Table I.

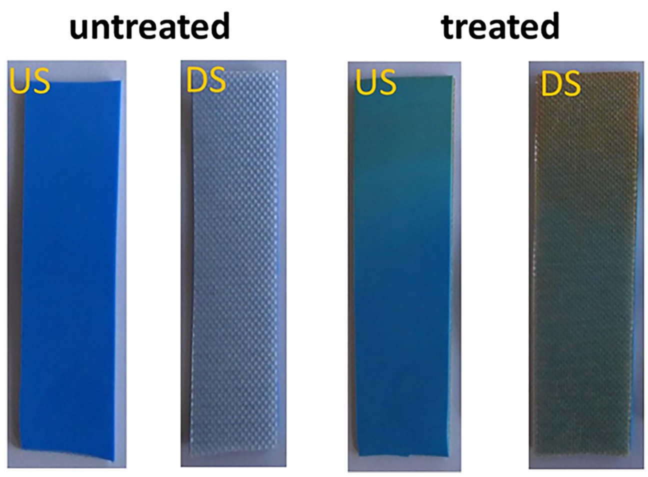


FIG. 2. A blue conveyor band made from PU. An adverse change in terms of color fastness appeared after a 7-day incubation period. The right-hand side shows a sample of a PTW-treated sample with its upside (US) and its downside (DS). Similarly, the left-hand side shows a PTW-untreated sample.

After the air flow is carried over the microwave discharge, it develops predominantly reactive nitrogen and oxygen species (RONS). Subsequently, the gaseous mixture of the reactive species is used to functionalize ordinary water (purified water for all lab scale PTW treatments). Therefore, the gas is simply passed into a tumbler (60 slm, rpm: 14), in which a PTW capacity of 1–10 l can be produced under stable conditions. The saturation time in the tumbler varies between 1 and 10 min, respectively. Since the RONS predominantly react to nitric and nitrous acid when fed into water,[17] stable production conditions are controlled via pH measurements. Consequently, through a defined incubation time in a pH-adjusted PTW solution (at an approx. pH of 1.5), reproducible conditions can be assured. In fact, the procedure, which brought the gaseous compounds in contact with water, is not as crucial as one might think. Definitely, a larger contact area between water and the gaseous compounds offers a larger reaction area for the entry of reactive compounds like RONS into the liquid phase. But contrarily, in previous experiments,[9] vastly different and specific methods, which vary in their contact areas, have been conducted with comparable results. Since the process has been repeatedly carried out up to a preset pH threshold (at an approx.: pH of 1.5), differences in the efficiency of various processes are not highly relevant, especially when they are not very pronounced.

### B. Conveyor belt materials and sample preparation

Generally, a conveyor belt adjoins materials that are capable of withstanding tractive forces and materials that build a produce specific surface, i.e., conveyor belts are made from composite materials. Our test series comprises belts that house tension-resistant polyester fabric downsides and surface materials, which vary in form and composition on their upper side (Fig. 2). Here, we tested silicone (SI, Shore hardness: A $30 \pm 3$) and polyurethane (PU, Shore hardness: $85.3 \pm 3$) surfaces (Vis GmbH, Treuen, Germany).

The SI surfaces possess a foamy appearance, which enables the belts for the transportation of sensible foodstuff like fresh cut lettuce or RTE apple produce. The PU belts offer a very rigid but ultra-hydrophobic surface with a self-cleaning ability. Samples used in the experiments were prepared as follows:

For optical microscopy and the mechanical stress tests, the samples were sized to a 50 mm length and approx. 10–13 mm width. Samples used in cleaning-performance experiments were sized in measurements of $20\,\text{mm} \times 20\,\text{mm}$. Samples used for microscopy were roughly cut to fit the sample holder of the used microscope. Accuracy was achieved for scanning-probe microscopy and light microscopy by a preset scan area for every sample of $100\,\mu m^2$ and a constant sample position, respectively (see also Sec. II D).

### C. Characterization of reference liquids and disinfectants

PTW was produced as described above (see Sec. II A). $HNO_3$ was diluted with purified water (double distilled water, Carl Roth GmbH, Karlsruhe Germany) from 65% nitric acid (Carl Roth GmbH, Karlsruhe, Germany) to a 2% nitric acid solution. The reference liquids tap water or purified water (double distilled water, Carl Roth GmbH, Karlsruhe, Germany) and the test disinfectant $HNO_3$ (2%) and PTW were chemically characterized by performing measurements of the pH value and the conductivity. Values for water hardness (in terms of carbon oxide concentration) are found in the literature.[18,19] A set of five independent measurements in a row was conducted with measurement gauges (Seven Compact Duo, Mettler Toledo, Gießen, Germany). Subsequently, belt samples were incubated in various experiments and treated as follows:

Samples used for belt-stress tests and surface characterization were submerged in the test liquids for up to 14 days (measuring points: 0d, 7d, and 14d). Samples used in the cleaning experiments were sprayed (30 s) via nozzles (producer: Lechler, Metzingen, Germany; series: 632; a water-jet angle of 45°; stainless-steel 1.4305; pressure: 200 kPa) in a spraying station (Fig. 3) with the test liquids with a volume flow rate of $0.63\,\mathrm{l\,min^{-1}}$. For the simulation of production residues, minced meat (pork) and apple slices (disk shaped, diameter. 5 mm, height: 3 mm), which introduce proteins, fatty components, and sugars into the experiment, were used for sample incubation in a refrigerator for 24 h at 5 °C. Subsequently, coarse soiling was roughly removed and the samples stored at 5 °C in a refrigerator for another 24 h. The effectiveness of the cleaning process was tested by microscopy.

### D. Material incompatibilities of the belt material and surface-cleaning tests

Since both the belt materials and the spraying nozzles are directly in contact with PTW, which houses many oxygenic compounds, tests to determine how PTW adversely influences such surfaces or components are particularly of great interest.[20,21] The physical and chemical properties of the surfaces were checked after $HNO_3$, PTW, and tap water treatments. Information about possible adverse alterations of the surfaces were gained optically by using optical micrographs and photographs and rheologically by performing a mechanical stress test. Additionally, the physico-chemical properties of the surfaces were obtained by performing atomic force microscopy (AFM) and contact-angle measurements. Also, possible mechanical abrasion from the nozzles might appear during its application when it is sprayed via a nozzle on the produce. Consequently, material incompatibilities of PTW might appear as a changed antimicrobial power of the sanitizers. Possible variations in sanitizing are validated by microbiological experiments.

#### 1. Hardness analysis

The tests included an untreated control in comparison with tap water, distilled water, PTW, and $HNO_3$. The samples were examined with the Texture Analyzer TAXT+ (WINOPAL Forschungsbedarf GmbH, Elze, Germany). A set ($n = 5$) of specimens were examined for five different test groups (control, tap water, distilled water, PTW, and nitric acid) under a quasi-static force transmission using a blade-like knife in a three point bending test. Before the test, the specimens were stored for 0 days (reference), 7 days, and 14 days. For the measurements, the recording started at a set point of 0.049 N. After reaching the required set point, the knife attachment was moved downward for 6.5 mm at a constant test speed of 1 mm/s. Meanwhile, the force was recorded at 500 measuring points per second (PPS). This experimental approach was carried out independently three times.

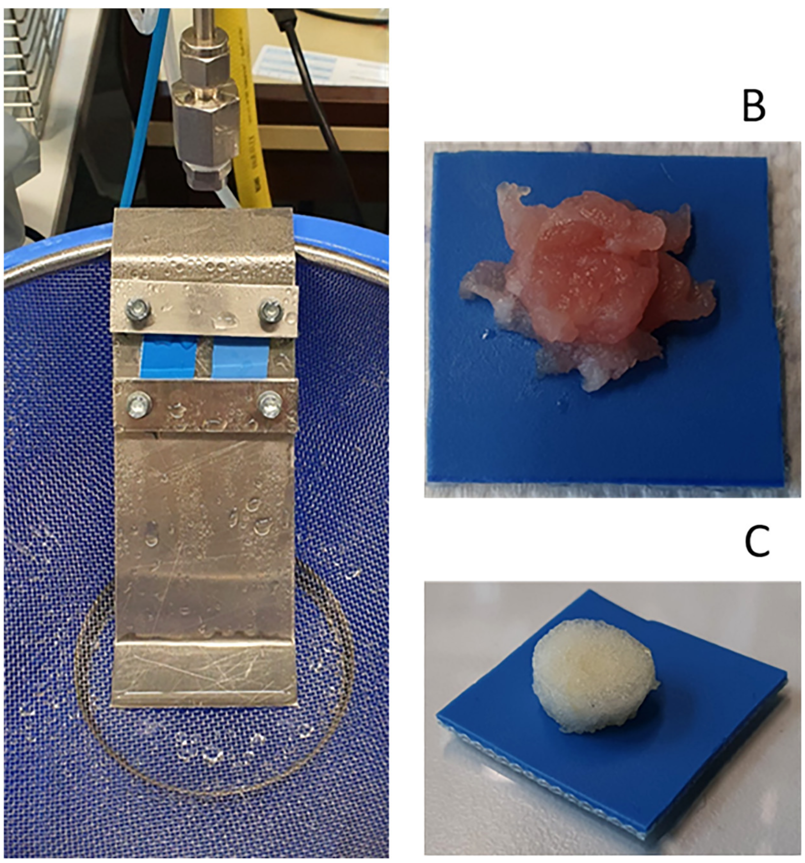


**FIG. 3.** Experimental setup for cleaning performance experiments. (a) Liquid spraying on the conveyor belt samples with a 45° angle. (b) Minced meat sample on conveyor belt. (c) Fresh apple pulp sample on the conveyor belt.

### 2. Light microscopy

The optical micrographs were recorded by using a digital microscope (KEYNCE, VHX-1000 series, Keyence Germany, Neu-Isenburg, Germany). All pictures were taken with a 50-fold magnification. The samples were fixed on a holder and measured under constant optical conditions. All samples were evaluated at five different points (the measuring point for each sample was N = 5).

### 3. Contact angle measurements

The surface energies of the belt materials were obtained by performing contact angle measurements. The contact angles of different liquid drops were measured using a commercial gauge (OCA 30, OEG, dataphysics, Fielderstadt, Germany). Deionized water ($H_2O$, surface tension: 71.99 mN/m,[22] ethylene glycol (EG, surface tension: 48.2 mN/m,[22] and diiodomethane ($CH_2I_2$, surface tension: 50.8 mN/m[22]) were used as measuring liquids. The liquids were held at room temperature and all further calculations assumed standard conditions for all samples. The free surface energies ($\sigma_{tot}$) were calculated by the Owens, Wendt, Rabel, and Kaelble (OWRK) method.[23] The method yielded a polar ($\sigma_p$) and disperse ($\sigma_d$) fraction of $\sigma_{tot}$, which behaved additively and summed up to $\sigma_{tot}$ ($\sigma_p + \sigma_d = \sigma_{tot}$). A commercial software (SCA 20, OCA software, dataphysics, Fielderstadt, Germany) was used to calculate all values. Here, the mean of five contact angles at five different spots was measured. Assuming Gaussian distribution of the values, the arithmetic mean and the standard deviation were calculated.

Due to their random asperity, which was determined in AFM measurements (see Sec. II D 4), the projected areas embraced by AFM scans do not depict the effective area of every scanned sample. The effective area is equivalent to the surface area in contact with the surrounding media, which governs the free surface energy. As a consequence, the measured free surface energy needs to be corrected by introducing a coefficient that accounts for roughness:

$$\cos(A1) = r^{*}\cos(A2), \tag{1}$$

$$r = \frac{S}{(L_X * L_y)}, \tag{1.a}$$

where $A1$ is the true contact angle, $A2$ the measured contact angle, and $r$ a correction factor (Si: 1.13 and PU: 1.01), with $S$ being the effective area of the sample (SI: $113.14\,\mu m^2 \pm 4.32$ nm and PU: $100.75\,\mu m^2 \pm 1.24$ nm) and $L_X$ and $L_Y$ being the length of each side of the sample in $x$ and $y$ directions ($L_x = L_y = 10\,\mu m$).

### 4. AFM topographies and surface properties

A sample of the SI- and PU surfaces was placed on object slides that were fixed on the sample holder of the atomic force microscope (AFM, NanoWizzard 3 JPK BioAFM, Bruker, Berlin, Germany). Subsequently, the samples were scanned in an intermittent-contact mode (set point: 0.5 V), with beam-shaped, silicon probes (Sicon, AppNano, Mountain View, CA, USA) without any coating, a nominal spring constant of $42\,N\,m^{-1}$, and a pyramidal-shaped tip (nominal aspect ratio: 7) used as cantilevers. The cantilevers have a nominal frequency at 300 kHz. Statistical quantities like roughness (Ra) were calculated from SPM micrographs that were measured from every produce-transporting surface of the conveyer belt. Therefore, the freeware software Gwyddion was used. For the determination of Ra, RMS, and the projected areas, five micrographs at different points of the same sample were taken. Again, assuming Gaussian distribution, an arithmetic mean and the standard deviation were estimated.

### 5. Antimicrobial efficacy and material incompatibilities of PTW with the spraying nozzles

Stainless-steel or brass tubes ($2600 \pm 20\,mm^2$) were PTW-incubated, reflecting a material spectrum frequently used for nozzle fabrication. The experimental setup comprised a negative control (0.85% NaCl solution, pH 5.4) and a positive control of pure PTW solution (pH 1.4), which were compared with the incubated PTW. The tubes were incubated in PTW for 2, 24, and 48 h and stored at 5 °C. The antimicrobial effectiveness of the solutions was tested on *Pseudomonas fluorescens* DSM 50090/ATCC 13525 (Leibniz Institute DSMZ—German Collection of Microorganisms and Cell Cultures GmbH, Braunschweig, Germany) suspensions with an OD of 0.11 (UV-3100 PC spectrophotometer, VWR International, Leuven, Belgium). Therefore, 0.5 ml bacterial suspension was mixed with 1.5 ml NaCl or PTW solution. To ensure an even distribution, the samples were shaken over the exposure times of 1, 3, and 5 min at 80 rpm. The antimicrobial effect was negated by pipetting a 3 ml tryptic soy broth (Carl Roth GmbH + Co. KG, Karlsruhe, Germany). The colony forming units were determined with a proliferation assay. The dilution was done with a maximum recovery diluent (MRD) in decimal steps. Tryptic soy agar (Carl Roth GmbH + Co. KG, Karlsruhe, Germany) was used as a solid nutrient. Colony forming unit (CFU) incubation was carried out at 30 °C for 18 h. The colonies formed were counted manually and the reduction factor was calculated in relation to NaCl.[14] A total of three experimental replicates were examined, whereas each experimental replicate contained repetitions of n = 3.

## E. Statistics

Assuming Gaussian distribution, all values represent the arithmetic means of the grouped values flanked by their standard deviations. For a statistical evaluation of a single experimental setup, the grouped values were compared with their references based on ANOVA. In the case of their pH, purified water and tap water were compared based on a two sided t-test. For both tests, the significance levels were chosen as $\alpha = 0.05$.

# III. RESULTS AND DISCUSSION

## A. Disinfectant characterization

To constrain the physicochemical properties of PTW, measurements of conductivity, the concentration of calcium oxide (CaO, water hardness), and pH were carried out. These parameters ensure a stable quality of PTW after its production. Additionally, alterations in pH or conductivity may adversely influence the produce during its processing. For instance, meaty produces must be processed at a certain degree of conductivity to ensure no osmotic drift during processing, which might enhance pathogen

**TABLE II.** Physicochemical properties of the test liquids: tap water, purified water, and PTW.

| | Tap water | Purified water | $HNO_3$ (2%) | PTW |
|---|---|---|---|---|
| pH-value | 7.12 ± 0.12 | 7.34 ± 0.15 | 0.52 ± 0.04 | 1.4 ± 0.02 |
| Conductivity (mS/cm) | 0.86 ± 0.01 | 0.01 ± 0.02 | 152.18 ± 4.76 | 11.37 ± 0.11 |
| Water hardness (CaO/mmol/l) | >2.5 | <1 | <1 | <1 |

spoiling.[24] These basic measurements characterize all fluids that were used for the experiments (purified water, tap water, nitric acid, and PTW). All values are summarized in Table II.

Not surprisingly, purified water shows the lowest conductivity (0.01 ± 0.002 mS/cm) at a pH of 7.34 ± 0.15. The local tap water (>3.8 mmol/l CaO) possesses a conductivity of 0.86 ± 0.01 mS/cm at a pH of 7.12 ± 0.12. $HNO_3$ (2%) has the highest conductivity (152.18 ± 0.15 mS/cm) and the lowest pH (0.52 ± 0.04) of all measured solutions. PTW shows a conductivity of 11.37 ± 0.11 mS/cm at a pH of 1.4 ± 0.02. Since all solutions, except tap water, are diluted with purified water, they show a low concentration of CaO (<1 mmol/l). The grouped values of pH appear statistically different for PTW and $HNO_3$ ($p < \alpha$) when compared with either tap water or purified water. On the other hand, purified water and tap water show no significant difference in pH ($p > \alpha$). Conductivity reveals statistical differences of all values for all solutions ($p > \alpha$).

All values appear in the realm of the expected. The conductivity and the pH values of the two kinds of water are only at a slight variance with the theoretical pH of electrochemically neutral water, but, in the case of drinking water, slightly alkaline waters are frequently observed.[18] The difference in the conductivities of $HNO_3$ and PTW is reflected by the lower pH of $HNO_3$, which appears promising for the use of PTW in distinct food production lines, since PTW's antimicrobial activity appears as high as that of $HNO_3$ by a reduced concentration of nitrate. However, all sanitizers may appear convenient in various cleaning scenarios. For instance, it may be hygienically important for sanitizers to be exchanged periodically. Rose *et al.*[10] report of *Salmonella* persistence in French broiler-chicken houses after frequent cleaning and disinfection with comparable sanitizers. Consequently, microorganisms develop resistances against sanitizers, which are frequently used within production lines.[11] PTW production is a well-known process, and it has been used to produce nitric acid in former times. But the procedure did not appear powerful enough to produce strong nitric acids, which were demanded by the chemical industry for fertilizer production or for functionalization reactions.[25] The moderate acidification of PTW during its production process appears to be an advantage in food production since solutions are sufficiently processed to gain antimicrobial activity on a demanded level (≥3 $\log_{10}$-cycle). No more highly concentrated and potentially hazardous acids must be stored anymore, and sufficient sanitizers can be produced on demand. The less acidosis and conductive PTW relieves the wastewater draw-off, decreases the cost for the safe storage of highly concentrated and hazardous sanitizer components, and relieves the production lines of potentially hazardous chemicals like highly concentrated nitric acid. Against this background, PTW extends the range of sanitizers, which are feasible for a concrete scenario. Nevertheless, the most striking advantage stems from an easy procedure to produce PTW, which can easily be installed in every existing production line. Since no storage for additional additives or concentrated acids is required anymore, a major threat for the possible occurrence of hazardous incidents is dramatically lowered. Last but not least, due to a lower pollution of waste waters, PTW might become a major player in sustainable food production lines, which is supported by a lower concentration of nitrogenous compounds compared with $HNO_3$.

## B. Antimicrobial efficacy and possible material incompatibilities

Therefore, the antibacterial effect of PTW might be adversely or positively affected by possible antimicrobial components, which were from the nozzle materials. The nozzles, which were applied in our experiments, possess metal or brass parts. The literature frequently reports the antimicrobial property of copper, which is a main component of brass. Consequently, due to an underlying antimicrobial effect of small brass or metal particles, particles in the sub-micrometer region might influence PTW's antimicrobial potential and additionally lead to an enhanced PTW ability in combating microorganisms.

Therefore, a possible increased antimicrobial effect due to an incubation of brass or stainless-steel pipes in PTW should be excluded by a comparison with bare PTW and a NaCl solution (0.85%) as a reference. Figure 4 summarizes the results of the experiments. Globally, we measured three distinct time periods for storage [storage times (ST) of 0 h, 24 h, and 48 h] with three different incubation times (IT) with *P. fluorescens* (1 min, 3 min, and 3 min).

A flat, not significant increase ($p > \alpha$) of the reduction factor (RF) of the *P. fluorescens* population depending on the incubation time was observed for pure PTW unless a ST of 24 h was exceeded (48 h: $p > \alpha$). In general, for ST > 48 h, the RF increases for a longer IT, an observation, which is additionally statistically significant for stainless steel when incubated in PTW ($p > \alpha$). The RF obtained in solutions contaminated by brass frequently suffers from relatively large standard deviations. Thus, no significance is observed, which is particularly striking for values with an ST of 2 h, a behavior, which was obvious throughout the experiment and at all recorded time points in all observed solutions. The *P. fluorescens* counts of bacterial solutions incubated in non-antimicrobial NaCl were taken as negative references. For the *P. fluorescens*-exposure times of 24 and 48 h, a decrease in the antimicrobial potential was observed in a descending order of pure PTW, brass, and stainless steel when both were incubated in PTW. The effect was not statistically significant ($p > \alpha$) after a storage time of 24 h but attained significance after an ST of 48 h. Additionally, slight differences could be seen at a 3 min exposure time PTW + brass on *P. fluorescens* and at 1 min

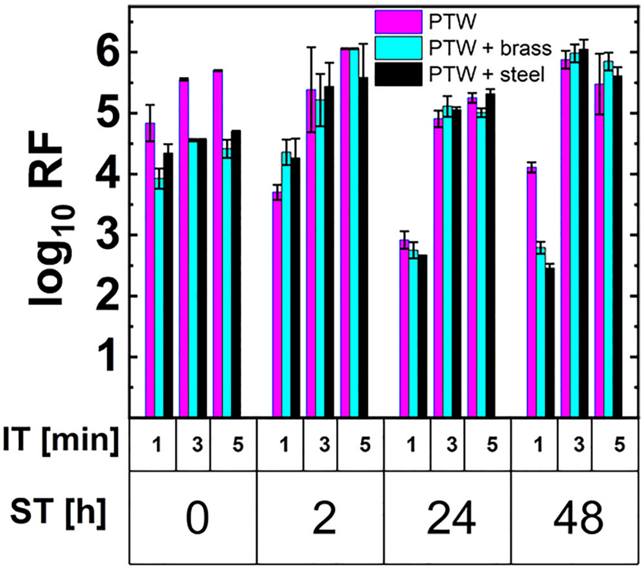


**FIG. 4.** Antimicrobial efficacy of PTW with and without brass and stainless-steel contact on *Pseudomonas fluorescens* (ATCC 13525) directly treated (0 h) with incubation times (ITs) of 1, 2, and 5 min or with storage time (ST) of metals in PTW (2, 24, and 48 h). The incubation of PTW with bacteria was 1, 3, and 5 min. The experiments were done with n = 3, and three independent experiments were conducted.

PTW + brass/PTW + stainless steel, respectively. Nevertheless, since data showed no obvious trend, no additional antimicrobial activity could be assigned to brass- or steel abrasions from the nozzle. Anyway, some data showed a better performance of pure PTW (e.g., 0 h, 3 min; 48 h, 1 min). Possible changes in pH were not found statistically meaningful ($p > \alpha$). The conductivity predominantly varied($p < \alpha$) for low conductive solutions steeped in purified water, a behavior that is not necessarily ascribed to an adverse influence of PTW, since solvation of ions from samples or the beaker wall may also lead to small but significant variations. But again, no obvious trend in data variation, which possibly reflects a concrete underlying process, was observed.

From our measurements, it is obvious that no significant changes ($p > \alpha$) in the antimicrobial potential of PTW were observed over an observation period of 48 h and IT > 3 min. Thus, the null hypothesis ($H_0: = a = \mu$) could not be rejected for those cases. Additionally, our data merely suggest that the inactivation performance decreases, especially when the ST exceeds 24 h. Here, a reduced antibacterial effect of PTW might possibly arise from longer contact with metal such as stainless steel or brass. In this regard, an experimental setup should be developed to support these findings, which also embrace deeper statistics like an equivalence testing. Additionally, the literature confirms our findings and an expected effect of an increased antimicrobial potential due to antimicrobial active metal particles was not observed.[26–29] To follow a logical argument, acidic solutions, particularly nitric acid, are famed for producing reactions on metallic surfaces.[30] Thus, a decreased effect might be traced back to a degradation of antibacterial active components in PTW with intensive metal contact,[31] a possible drawback that becomes meaningless for industrial applications, since the direct contact times of PTW with the nozzle material are solely in the range of seconds. Contrarily, it is known that in an acidic environment (PTW with pH 1.4), brass corrodes rapidly due to a dezincification process.[32] The dezincification of brass would mean that, on the one hand, it may lose valuable mechanical and physical properties, and on the other hand, antimicrobial active copper cations may be released into PTW.[33] As an alloy of copper (Cu) and zinc (Zn), brass can be oxidized by nitric acid ($HNO_3$, a possible reaction product in PTW) to copper nitrate [$Cu(NO_3)_2$], nitrogen dioxide ($NO_2$), and water,[34] which are in part antimicrobial components.[35] The experiments do not significantly support comparable mechanisms, which are described in the above cited literature. Nevertheless, the unpronounced variation of data in our experiments rather suggests variations within the error margins or outliers due to experimental uncertainties. In addition to the consistently high inactivation of *P. fluorescens* by PTW, these investigations have proven a sustained antibacterial effect of PTW over at least 48 h. As stated below, an enhanced antimicrobial potential may appear due to a better solvation of gases in cool liquids by PTW production. An enhanced activity of PTW due to a cooled storage in a glass vessel at 5 °C is also imaginable and an RNS stabilizing due to downregulated entropic processes also appears feasible. However, the nozzles show no adverse influence of PTW treatment. Overall, it can be concluded that brass or stainless-steel components installed in nozzles are apparently compatible and can be used for industrial PTW applications.

### C. Material compatibility of PTW

#### *1. Measurements of hardness*

Almost all components built into the decontamination unit underwent mechanical stress, which may lead to material fatigue and a decreased life span. Additionally, the materials also faced chemical stress due to the effect of nitrous compounds, and this stress is found in $HNO_3$ treatments and is less pronounced in PTW treatments.[36] Mechanically adverse effects of the treatments were monitored in measurements of the elasticity of the samples before and after a PTW-based or a nitric acid treatment, which were compared with a completely untreated reference and samples incubated in tap water and purified water that underwent the same experiment. The results of these investigations are shown in Fig. 5.

The data show no significant differences in comparing liquid-treated PU- or Si belt material over 7 ($p > \alpha$) and 14 days of incubation with PTW and $HNO_3$. The differences in observed pressure are due to the material itself, as Si is softer than PU. Slight differences may be detected between the untreated reference and the liquid-treated samples; however, for the PTW treatment, no difference in tap water or distilled water was detected. From these results, it could be concluded that a daily application of sprayed PTW for 13–14 min to clean the conveyor belts with a total duration of approx. 2 years (7 day test) and beyond (14 day test) should not significantly influence the elasticity compared with tap water or other liquids.

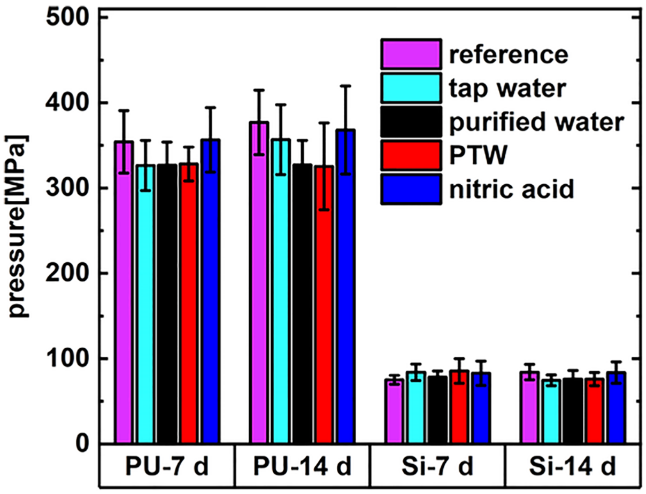


**FIG. 5.** Texture analyses of different conveyor belt materials incubated in different liquids over 7 and 14 days. All data points contain repeats of n = 5 with a preset force of 0.045 N. Before the experiments, the samples have been stored in tap water, purified water, PTW, and nitric acid for 0, 7, or 14 days. They have been compared with an untreated reference.

Generally speaking, harnesses for PU elastomers or SI polymers are measured with a durometer in terms of Shore hardness.[37] Since we used a texture analyzer for food stuffs for our measurements, the working principles of which are not fully covered by the requirements of national or international standards,[38–40] we display our results in terms of a pressure (MPa) that was needed to reach a distinct penetration depth. We ensured that the assumed basic conditions like a narrow temperature range (±2 K) were fulfilled. The promising results show a stable surface against any influences of the tested acidic solutions, which does, in the case of a PU surface, appear surprising. Frequently, PU surfaces show no resistance against diluted nitric acids,[41] whereas SI surfaces show a relatively pronounced resistance against acids.[42] However, acidic sanitizers are frequently used in food production lines,[43–45] and our experiments did not reveal a pronounced influence of sprayed off PU- or SI surfaces after a spraying period of minimum 13 min. Contrarily, the belt material withstood the treatments in the experiments and showed no mechanically adverse alterations. However, a possible adverse alteration of the mechanical properties of PU belts cannot be excluded after long periods of usage in the presence of nitric acids. The rheological behavior of the material is conserved within the framework of our methods and the rheological requirements and remains applicable in a production environment.

### *2. Wettability and surface energies*

The OWRK method divides surface energies (SEs) into a polar and an apolar part, which both contribute to the overall electro-chemical surface behavior. Therefore, we determined the contact angles of purified water, ethylene glycol, and diiodomethane. The experiments embrace PU and SI samples that were submerged in purified water, tap water, PTW, and nitric acid. The specimens were sampled after a day, 7 days, and 14 days. They were determined in contrast to a reference, which reflects the untreated surface. The materials of the top cover of the conveyor belts are made of PU and a SI compound. Parameters such as surface energies, roughnesses, and the hardness of the surface material were determined by conducting contact angle measurements, SEM micrographs, and elasticity measurements, respectively. These experiments are particularly of interest for a detailed characterization of a specimen's surface and its stability after its long-term use with relatively moderate acidic solutions such as $HNO_3$ or PTW.

PU has an overall surface energy of $41.72\ J/m^2 \pm 1.62\ J/m^2$, which is apparently the highest value observed on all references. The SE divides into a disperse part of $41.64\ J/m^2 \pm 1.56\ J/m^2$ and a polar part of $0.38\ J/m^2 \pm 0.38\ J/m^2$. A roughness of $31.52\ nm \pm 1.63\ nm$ was obtained. SI, the most rigid sample, has an SE of $14.99\ J/m^2 \pm 2.13\ J/m^2$ with a disperse part of $11.04\ J/m^2 \pm 8.71\ J/m^2$ and a polar part of $0.03\ J/m^2 \pm 0.07\ J/m^2$. AFM surface scans revealed a roughness of $103.2\ nm \pm 6.91\ nm$. The SE remained stable after a 24 h period when the samples were submerged into purified water and tap water. Samples having a PU-top cover on their carcasses gained their polar part of the SE, consequently lowering their dispersity. The PS-top cover's SE are conserved and show no statistically meaningful changes. Samples that were submerged into $HNO_3$ or PTW only marginally altered that picture. Indeed, a lower SE was observed for all top covers when they were incubated in $HNO_3$. The SE decreased on the PU and PS-top cover when they were incubated in PTW. Here, the alterations were so much into scope, but SE changes might appear significant ($p < 0.05$). However, the numbers that were obtained for the SE changes might appear statistically meaningful, but the macroscopic behavior of the surface changed only marginally. For instance, despite changes in the SE of the PU surface, it kept its ultra-hydrophobicity after a 24 h incubation time in solutions with relatively moderate (for more technical not biological systems) pHs. The above-described situation with an increase of the polar part of the SE by the submerged specimens was also observed for the aqueous acids $HNO_3$ and PTW.

The scenario that was observed after a 24-h incubation changed only marginally after 7-day and 14-day storage periods. Obviously, the SE also changed with statistical significance ($p < 0.05$) for various samples, when compared with that for a reference. For instance, the SI sample exposed an elevated polar fraction of their SE (polar fraction after PTW treatment: SI: $2.11\ mJ/m^2 \pm 1.26\ mJ/m^2$ (14d) vs $0.38\ mJ/m2 \pm 0.39\ mJ/m^2$ (24 h). Despite also meaningful SE changes ($p < \alpha$), PU also kept its hydrophobicity after such incubation times [the disperse part of PU after PTW treatment: $9.53\ mJ/m^2 \pm 1.45\ mJ/m^2$ (14d) vs $11.04\ mJ/m^2 \pm 1.63\ mJ/m^2$ (24 h)]. Repeatedly, SE variations were observed only in small margins and no change in their macroscopic behavior was observed [the biggest SE change after PTW treatment: PS $41.72\ mJ/m^2 \pm 1.62\ mJ/m^2$ (24 h) vs 37. $72\ mJ/m^2 \pm 1.18\ mJ/m^2$ (14d)]. The biggest SE change overall was observed for a reference, which was introduced into tap water. Roughness did not vary after incubation with those relatively moderate acidic pHs and appeared to be very stable on the nm scale. The roughness parameters remained remarkably stable and showed no significant changes

**TABLE III.** Summary of the surface energy measurements. The energies are listed with their polar and disperse proportions to the surface energy.

| 0d | SI | | | PU | | |
|---|---|---|---|---|---|---|
| | Reference | $HNO_3$ | PTW | Reference | $HNO_3$ | PTW |
| SE polar (mJ/m$^2$) | 0.04 ± 0.07 | 1.22 ± 2.65 | 0.39 ± 0.39 | 0.38 ± 0.38 | 0.73 ± 0.56 | 0.98 ± 0.33 |
| Roughness (nm) | 103 ± 8.71 | 101.2 ± 41.87 | 96.2 ± 13.48 | 31.52 ± 1.63 | 32.9 ± 7.75 | 32.94 ± 8.04 |
| 7d | SI | | | PU | | |
| | Reference | $HNO_3$ | PTW | Reference | $HNO_3$ | PTW |
| SE polar (mJ/m$^2$) | 0.04 ± 0.07 | 1.57 ± 1.38 | 0.57 ± 0.45 | 0.38 ± 0.38 | 1.33 ± 0.99 | 4.59 ± 0.54 |
| Roughness (nm) | 103 ± 8.71 | 101.2 ± 41.87 | 96.2 ± 13.48 | 31.52 ± 1.63 | 32.9 ± 7.75 | 32.94 ± 8.04 |
| 14d | SI | | | PU | | |
| | Reference | $HNO_3$ | PTW | Reference | $HNO_3$ | PTW |
| SE polar (mJ/m$^2$) | 0.04 ± 0.07 | 2.67 ± 1.25 | 2.21 ± 1.26 | 0.38 ± 0.38 | 0.97 ± 1.03 | 1.48 ± 1.41 |
| Roughness (nm) | 103 ± 8.71 | 103.45 ± 9.75 | 104.49 ± 9.3 | 31.52 ± 1.63 | 33.1 ± 8.45 | 34.66 ± 7.89 |

(7d-$HNO_3$: SI 101.2 nm ± 41.87 nm, PU 32.9 nm ± 7.75 nm; 7d-PTW: SI 96.2 nm ± 13.48, PU 32.94 nm ± 8.04 nm and 14d-$HNO_3$: SI 103.45 nm ± 9.75 nm; PU 33.12 nm ± 8.45 nm; SI 101.56 nm ± 13.56 nm; PU 31.97 nm ± 9.22 nm; 14d-PTW: SI 104.49 nm ± 9.34 nm; PU 34.66 nm ± 7.89 nm). All values are summarized in Table III.

In summary, all conveyor belt specimens keep their specifications for a secure application in a foodstuff production environment. A significant change in the polar fraction of the SE of SI samples in aqueous solutions is a consequence of a water uptake due to the top cover's spongy appearance rather than that of any adverse influence of any sanitizers ($HNO_3$ and PTW). This fact becomes obvious when changes are observed when solely purified water or tap water is used. Thus, the altered polarity of the specimens is production related, adjustable on customers' demands, and can be altered by manufacturers.[46,47] As mentioned above, the most pronounced changes in the overall SE are observed for a self-referential PS surface submerged into tap water. Additionally, it is evidence that SE changes are rather a cross reaction of the surfaces with the surrounding medium than a direct reaction to a PTW exposure. Roughness changes also sparsely appear in our test series. Certainly, abrasion happens during the use of the belts, which undoubtedly alters the surface roughness. But such abrasion processes are not implemented in our test series and they will gain importance only when the abrasion is increased after a $HNO_3$ or a PTW incubation. Such an experiment is out of scope of our present methods, and it is also highly unlikely to be performed due to an unchanged hardness of all specimens after treatments with acidic solutions. In summary, even after a 14 d incubation with $HNO_3$ and PTW, all belts show their elemental material properties in terms of their wettability, their roughness, or their hardness. Contrarily, our experiments do not cover all possibilities for an abrasion. For instance, components in contact with acidic solutions can alter in color when they are in contact with ambient air directly after their incubation. Here, it is most likely that the acids possess the ability to trigger or promote redox processes.[48] Experiments to test the stability of the substrates of the belts are also implemented but show no significant changes in the scope of the measuring area (data not provided). Thus, an adversely altered stability has not been assumed.

## D. Cleaning experiments

The cleaning experiments embraced a washing step with the pressurized sanitizers $H_2O$, $HNO_3$, and PTW. The experimental setup intended to simulate an industrial washing procedure employing a spray nozzle that sprays the sanitizer on the conveyor belt's surface at an angle of 45° (Fig. 3). The sanitizers were sprayed upon the surfaces with a pressure of 200 kPa. The surfaces of the belt specimens were incubated with minced meat or a defined disk-shaped apple slice, a procedure that directly soiled the specimen's surfaces.

Light-microscopic micrographs reveal a structured, faveolated appearance for the SI samples (Fig. 6), whereas the PU sample appears rather unstructured with a topographic of smooth and random waviness (Fig. 7). These conditions are reflected by their surface roughness, which have been recently used for surface characterization (Sec. III C 2).

The untreated references [Fig. 6(a)] that were microscoped directly after their extraction showed a relatively soiled surface for the SI reference. Randomly, longish, filament-like dust particles spread [Fig. 6(a), +] over the surface. In contrast, the PU reference widely revealed a dust-free surface [Fig. 7(a)]. Consequently, all sanitizers were used to rinse all sample surfaces until no further soiling was detected [Figs. 6(d), 6(g), and 6(j); Figs. 7(d), 7(g), and 7(j)]. Conclusively, all sanitizers left behind a clean surface after their application. Solely, the PTW step left behind particles with a much lower density [Fig. 6(j), +]. The PU surface revealed a brighter stripe-like structure after $HNO_3$ washing [Fig. 7(g), #]. In summary, both surfaces were dust free and showed no obvious soiling after $H_2O$-, $HNO_3$-, and PTW washing steps.

Since the cleaning abilities of $H_2O$, $HNO_3$, and PTW were to be compared, unwashed samples, incubated with minced meat or overlaid with a disk-shaped apple slice, served as a basic reference. Figures 6 and 7 summarize the results of a 24 h incubation time of both surfaces with both kinds of organic soiling, which reflects soiling that can be found in contemporary production environments. For surfaces incubated with minced meat, the SI surface [Fig. 6(b)] shows firmly dried up meaty residuals (*) and a brighter

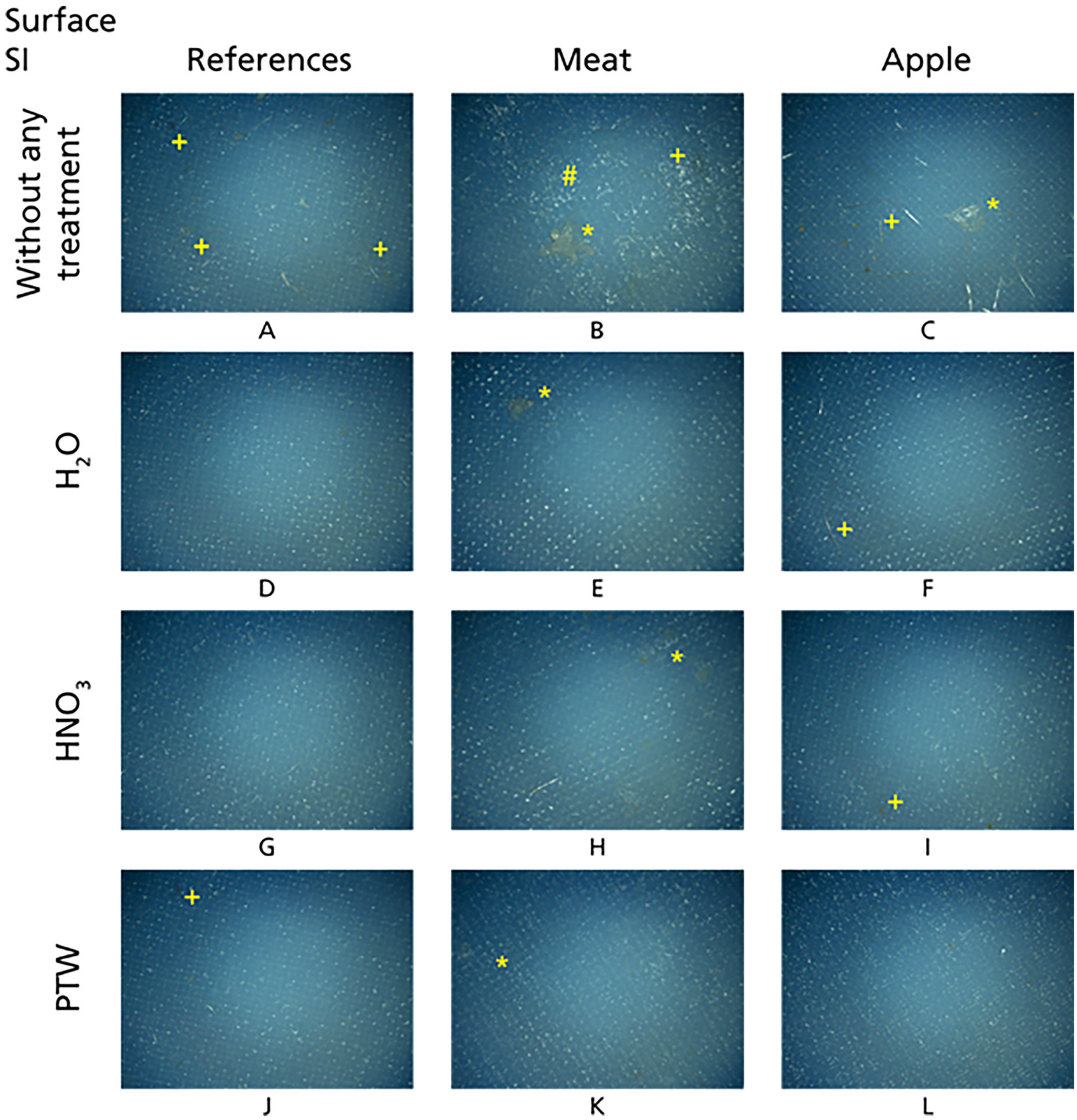


**FIG. 6.** Summary of pictures, which recorded for the cleaning experiments of SI surfaces. The upper line shows micrographs of surfaces without any treatment (bare reference, an uncleaned surface when incubated with meat, and an uncleaned surface when incubated with apple). The second row shows those surfaces when cleaned with water. The third row encompasses micrographs when cleaned with nitric acid, and the fourth row embraces micrographs of surfaces when cleaned with PTW.

structure with a moisture-like appearance (#). Additionally, dust-like soiling appears on that surface (+). The PU surface also reveals meaty residuals (*) and a structure with a moisture-like appearance [Fig. 7(b), #]. The PU references show no dusty soiling. The apple-contaminated references appear slightly different. Predominantly, the SI surfaces rather show rod-like structures like the ones observed on the bare reference [Fig. 6(c), +] but are thicker in appearance [Fig. 6(a), +]. An organic residue is also found

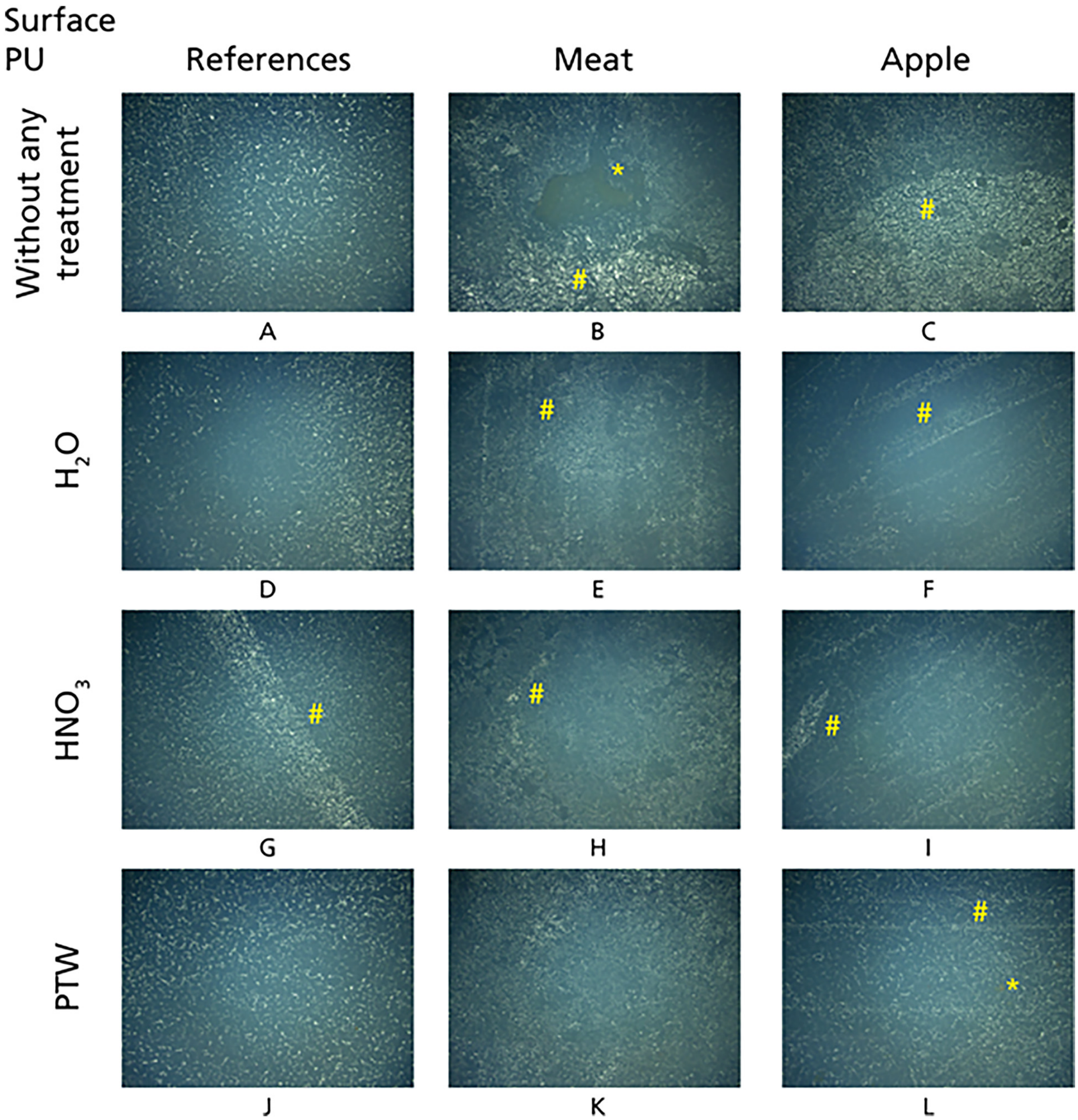


**FIG. 7.** Summary of pictures that were recorded for the cleaning experiments of PU surfaces. The upper line shows the micrographs of the surfaces without any treatment (bare reference, an uncleaned surface when incubated with meat, and an uncleaned surface when incubated with apple). The second row shows those surfaces cleaned with water. The third row encompasses micrographs when cleaned with nitric acid, and the fourth row embraces the micrographs of surfaces cleaned with PTW.

[Fig. 6(c), *]. The PU surface solely shows the residuals of a dried-out suspension [Fig. 7(c), #]. In summary, all sanitizers sufficiently clean the SI- and PU surfaces. The samples that are incubated with minced meat reveal contamination with organic, meaty residuals that are not very densely spread over the surface [Figs. 6(e), 6(h) and 6(k); *]. The density of the contamination that has been found on the SI surfaces after sanitation reveals an increasing order with respect to sanitizers such as PTW, $HNO_3$, and $H_2O$.

The surfaces of the PU samples appear predominantly without any contamination. A low abundance of structures of dried-out suspensions appears upon the surfaces [Figs. 7(e), 7(f), 7(h), 7(i), and 7(l); #]. The PTW cleaning step predominantly reveals surfaces with no further contamination such as organic residuals or dried-out remnants of a suspension [Figs. 7(k) and 7(l)]. The scenario, when samples are contaminated with an apple disk, resembles the one observed for a minced-meat incubation. After the cleaning step, the samples solely show the remaining contamination in an exceptionally low abundance. The SI surfaces reveal rod-like structures [Figs. 6(f) and 6(i), +], whereas the PU surfaces present only dried-out residuals [Figs. 7(f) and 7(i), #]. The PTW cleaning step leaves behind surfaces without any further contamination [Fig. 6(l)] for SI surfaces. The PTW cleaning step leaves behind a surface where additional contamination with an organic remnant is found [Figs. 7(l) and 7(c), *].

Certainly, all sanitizers leave behind a homogenous picture in terms of their cleaning abilities. As shown in Figs. 6 and 7, no spacious soiling is observed after a particular sanitizer treatment. Concretely, the experimental setup has an atomization nozzle that is mounted at a 45° angle on the samples. Due to the mounting angle, the main forces that remove the soiling from the surface are predominantly governed by shear stresses.[49] Additionally, since all sanitizers are based on an aqueous solution, the applied pressure at the outcome is widely the same for the experimental setup (200 kPa), and since the angle of the nozzle is not changed when performing the experiments, another result for cleaning has not been expected. All micrographs presented in that study show the worst cases of soiling, which have been found for all surfaces at each data point (i.e., PU surface, $HNO_3$ rinsed, meaty soiling). Predominantly, the untreated SI surfaces show rod-like structures with a higher density, which are interpreted as dust grains. The PU surfaces widely appear without any soiling, particularly in their references, a fact that is reflected by the low surface energies of the samples and the self-cleaning ability of an ultra-hydrophobic surface.[50] We rinsed the treated references with water, $HNO_3$, and PTW, which reflect the spectra of the tested sanitizers. Micrographs of the references show no soiling for the SI surfaces and traces of dried-out solutions, which have been interpreted as residuals of the sanitizers. These findings are most likely based on the brand new samples of the conveyor belts that were used throughout the experiments. This interpretation is also supported by the AFM micrographs and the roughness parameters, even though the surface energies that were determined for all surfaces are low.

Both samples, when incubated with minced meat, show dried-out accumulations of organic residuals that have been interpreted as dried-out minced meat. When meat-incubated SI surfaces are sanitized with aqueous solutions, our experiments reveal dried-out organic accumulates, as observed on the incubated reference. Certainly, the density of those accumulates is lower after the sanitation, but those residuals might host microorganisms and possibly support the proliferation of food-borne pathogens. These findings embrace all sanitizers used in our experiments. The density of the contamination, which has been found on the SI surfaces after sanitation, reveals an increasing order with respect to sanitizers such as PTW, $HNO_3$, and $H_2O$. PTW and weaker $HNO_3$ lower the adhesion forces of the meat residuals or change their structures, which, in consequence, favor an unbound state. But we are only hypothesizing based on our data. Nevertheless, since samples, when incubated in $HNO_3$ and PTW, do not significantly vary in their surface energies, altered adhesion forces due to a pronounced alteration of the surface energies might be discounted.[51] A structural change in the organic residuals might provide an explanation for this; also, structural changes occurring in proteins in the presence of acids have been described manifold.[52–54] But contrarily, $HNO_3$ and PTW are acidic solutions and fatty and protein residuals are rather removed with basic solutions.[55,56] In summary, the rinsing steps of the ultra-hydrophobic PU surfaces reveal pictures of remarkably clean surfaces, although isolated areas of dried-out sanitizers are observed, a scenario that appears to be valid for all tested sanitizers. The ultra-hydrophobic surfaces of the PU samples predominantly underlie the interactions with aqueous sanitizers, and a low wettability of the surfaces prevents a spreading of the sanitizers on the surface, which also complicate possible decontaminating effects.[50]

A comparable interpretation appears valid for both surfaces that are contaminated with an apple slice. After contamination, they show a relatively dense soiling, which is interpreted as multiple clusters of organic residuals of the incubation object. Both surfaces, with SI surfaces being more pronounced, show organic residuals in predominantly two appearances. On the one hand, the SI surfaces reveal clustered organic remains, which have been interpreted as small portions of pulpy apple tissue. On the other hand, rod like structures, which are thicker in appearance as the dust grains on the bare reference, are also frequently observed on the SI surfaces. These structures have been interpreted as cell-wall residuals of the fruit skin. The rinsing step leaves behind clean surfaces, particularly after a PTW treatment. SI samples that have been rinsed with $HNO_3$ or water sparsely show residuals of cell-wall material. The SI surface seems more suitable for an apple production line than for animal produce, which is also reflected by the softer surface that is more protective to more susceptible produce such as apples or leafy greens.[57] PU samples, when overlaid with an apple slice, leave behind a predominantly dried-out suspension on their incubated surfaces, which have been observed for water and $HNO_3$ samples. Pulpy residuals are only sparsely observed on PU surfaces after PTW rinsing.

A reduction of conventional cleaning agents and a refinement of existing processes may also positively influence the economic viability of production processes. Certainly, in a direct comparison (excluding side costs such as storage or wastewater disposal), PTW appears as a worse choice economically than $HNO_3$. Undoubtedly, PTW production is an energy consuming exercise (approx. 72 kW h/m$^3$, which is the overall consumption of the process, embracing not only an energy coupling into PTW but also energy to run equipment like cooling stages in the plasma generator), which may be economically viable only when greater amounts of PTW are produced for usage. However, the cost intensive production process may be cushioned or may be overcome by the production of the right amount of PTW at the right time. The need to keep reactive and potentially hazardous ingredients of cleaning agents in stock is totally eliminated or a refined wastewater-disposal management may also appear advantageous and more environmentally friendly. Environmentally friendly production processes may also increase consumers' acceptability of a produce or generate interest for producers. Generally, plasma processes are often referred to as "chemistry from the socket," which reflects the eliminated need for a more elaborate infrastructure as they can be installed almost everywhere. A PTW cleaning step consumes electricity, water, and ordinary air, which all have to be compressed. However, since PTW (and $HNO_3$) as an aqueous sanitizer offers a wide range of applications, concrete cost calculations for conveyor band cleaning are extremely inaccurate and vary due to unequal energy costs.

However, in-place cleaning procedures should not only be thought of in terms of sanitizers. A cleaning procedure is an interplay between the surfaces that must be cleaned, production variables such as temperature or moisture, the desired grade of decontamination, the time that an equipment/surface is accessible for a cleaning step, and sanitizers.[55] Against this background, PTW might widen the spectra of useful sanitizers in the area of food production. For instance, our experiments reveal an enhanced effectivity of PTW under cooled conditions (although sparse data that have not been provided), which can be interpreted as an enhanced uptake of plasma activated gas due to Henry's Law.[58] However, a correlation between Henry's law and an enhanced effectivity of PTW has not been proven yet, which reflects the need for a higher research effort to gain a deeper insight into distinct chemical processes that happen from the point at which there is gas uptake and that predominantly govern PTW chemistry and its effectivity against pathogens. Basic research has already been done by various authors such as in Refs.15, 16, and 59. However, there are still unanswered questions.

## IV. CONCLUSION

As it is reflected in our data, PTW may be advantageous when protein and fatty residuals must be removed from SI surfaces. This is apparently still valid for PU surfaces, where PTW sanitizes as sufficiently as water or $HNO_3$. SI surfaces even appear less contaminated after a PTW rinsing step than SI surfaces that have been rinsed with water or $HNO_3$. On the other hand, it slightly appears less effective when fruit pulp must be removed from PU surfaces. Nevertheless, for PU surfaces, mainly its ultra-hydrophobicity and the aqueous nature of the sanitizers govern the density of soiling, but as was observed for $HNO_3$, PTW did not adversely influence the desired properties of the conveyor belt surfaces. Additionally, PTW shows a decontamination efficacy as high as that of $HNO_3$, but it is not affected by factors such as low pH. Conclusively, PTW sufficiently decontaminates plastic surfaces such as SI and PU surfaces, does not adversely change any physical or chemical properties of the surfaces, and produces good cleaning results. Thus, PTW offers an extensive spectrum of possible sanitizers for specialized cleaning demands in a food production line.

## AUTHORS' CONTRIBUTIONS

T.W. and U.S. contributed equally to this work. T.W. and U.S. were involved in conceptualization; T.W., U.S., and S.N. in methodology; T.W., U.S., H.W., T.M., and J.S. in investigation; T.W. and U.S. in writing—original draft preparation; S.N., O.S., and J.E. in writing—review and editing; J.E. in supervision; O.S., and J.E. in project administration; O.S., and J.E. in funding acquisition. All authors have read and agreed to the published version of the manuscript.

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## DATA AVAILABILITY

If not listed in the article, the data that support the findings of the study are available from the corresponding author.

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