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Mechanically induced dye-release from polyurea microcapsules in a rubbery adhesive

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Abstract

Especially in the building industry the demand for *in situ* monitoring concepts of adhesive joints persists. In a new approach, microcapsules filled with special dyes, are added into a rubbery adhesive. Certain stress levels within the adhesive lead to a breakage of the capsules, which release their liquid cores. This results in a detectable colour change and thus, enables non-destructive condition monitoring of the bond. Dye-filled polyurea microcapsules were produced by interfacial polymerization by varying synthesis parameters while their influence on capsule morphology and wall thickness was determined. The morphology and thermal properties of the microcapsules were characterized by scanning electron microscopy and dynamic differential calorimetry, respectively. Subsequently, the method of nanoindentation was used to study the deformation and fracture behaviour of the microcapsules. Finally, it was analysed to what extent the microcapsules break within a polymer matrix, e.g. by critical stress levels or deformations. This development is a new quality assurance method for glass façades.

Keywords: microcapsules, dye-release, fluorescence, nanoindentation

(Some figures may appear in colour only in the online journal)

1. Introduction

Adhesive joints for connecting glass panes within modern façade constructions are attracting growing interest due to new design options with high degree of transparency. However, there are still reservations on the durability of loadbearing joints designed as point or linear connections. The examination of their condition is often limited to visual inspections, where statements about the load history are restricted and small defects are not always being detectable. But for safety issues, a condition monitoring of adhesively bonded joints is of utmost importance and topic of recent research projects [1]. This article focuses on a condition monitoring method for damage detection in rubber adhesive bonds based on microcapsules.

Microcapsules have now found their way into almost all branches of industry. It is expected that the global microcapsule market will rise from $6.3 billion in 2018 to $11.8 billion by 2023 [2]. This is due to the versatility of microcapsules, which is also expressed in the adjustability of the capsule properties, e.g. its release behaviour. Some microcapsules are

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intended to permanently separate a substance from the environment (e.g. phase change materials [3–5]). In some cases it is important that an encapsulated substance is released over a period of time (e.g. controlled release in pharmaceuticals [6]). Many other applications use an external stimulus to initiate the release of the capsule content. For example, in the case of carbonless copy paper, the pressure applied by a pen initiates a capsule break in a coated sheet of paper. In this way, a special dye is locally released and creates the copy [7]. Microcapsules are also used to secure screw connection by coating the thread with microencapsulated adhesives. By screwing in, shear forces are exerted and lead to the release of the adhesive and finally its curing in the thread gap [8]. Furthermore, some self-healing materials use a mechanically induced release from microcapsules. In these, a healing agent is encapsulated and distributed within a matrix. When the matrix is damaged, the capsules open and the adhesive infiltrates the appearing crack. After hardening a regeneration of the mechanical properties may be possible. The initiation of an autonomous regeneration of the mechanical properties of plastics, adhesives, paint and cement systems [9–13] has been the subject of research and development for many years.

However, it is not only the repair of structures that is important, but also the condition monitoring [14–17]. The use of microencapsulated dyes as damage indicators in load bearing transparent adhesive bonds may be a promising approach to detect damage at an early stage.

While the approaches to self-healing and damage indication are already fundamentally described in the literature on rigid materials, the behaviour of microcapsules in elastomeric materials such as silicone adhesives has not yet been covered. This article therefore, describes how the morphology of microcapsules affects the possibility of a 'smart' adhesive for detection of critical conditions. A detection of damages in transparent joints is enabled by using micro-encapsulated dyes. When the adhesive joint is stressed critically, the embedded microcapsules rupture. The microcapsule breakage leads to a release of the core material, in this study a fluorescence dye, in the occurring crack. This release becomes visible under UV light. The used fluorescence dye exhibit aggregation-induced emission (AIE) properties. In aggregated or crystallized form, however, intramolecular motions within the AIE molecules are restricted. This promotes efficient photoluminescence in the solid state. In this case closed microcapsules are non-fluorescent. Upon capsule rupture and evaporation of the solvent, a fluorescence is visible [17].

Here, the mechanical properties of the microcapsules are of particular importance. On the one hand, the microcapsules must be sensitive enough to release their contents when a defined force is applied. On the other hand, they must be stable enough to be incorporated into highly viscous silicone adhesive systems without being destroyed, even under high shear forces. Microcapsules with such fine-tuned properties has been studied rarely yet. In the course of the investigations, the influence of the chemical composition on the capsule wall thickness and thus on the mechanical properties was examined.

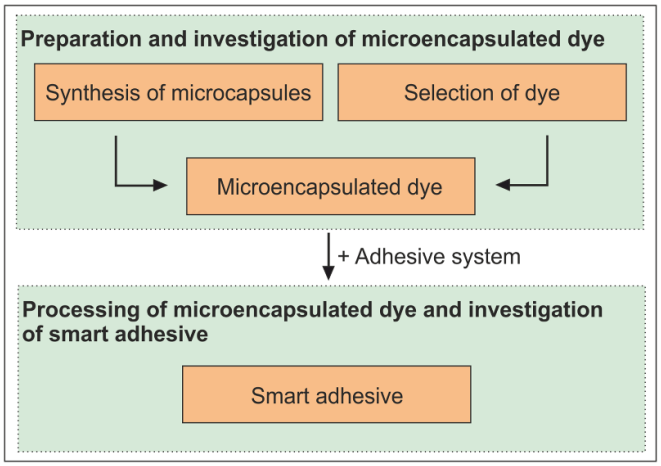

Figure 1. Methodical approach.

2. Methodology and experimental details

In figure 1 the methodical approach is constituted. In the first part of the study the selected dye is microencapsulated and the resulting system characterized. In the next step the microcapsules are added to the two-component silicone adhesive to obtain the smart adhesive. The mechanical properties and the influence of the microcapsules on the smart adhesive are characterized. Among others, a comparison of the mechanical testing results between microcapsules added adhesives and regular adhesives without microcapsules is made.

2.1. Dye-containing microcapsules

2.1.1. Synthesis of microcapsules. The preparation of core–shell microcapsules is done by interfacial polymerization. A polyurea as capsule wall material was selected for the microencapsulation of the damage detector. For the core solution the diisocyanate Desmodur L75 is dissolved in the hydrophobic solvent xylene with a fluorescence dye. For the first experiments the dye coumarin is used. This dye is cost-effective and visible via light microscopy. For further investigations the AIE dye tetraphenylethene (TPE) is applied, which leads to a much more perceptible fluorescence after release under UV light.

The core solution is emulsified in water with the addition of stabilisers. This was done with an Ultraturrax disperser from IKA Staufen, Germany. After emulsification, an aqueous solution of the trifunctional amine guanidine hydrochloride is added and distributed in the aqueous phase. With further stirring and an increase in temperature, both components react at the O/W interface to form polyurea, which shapes the capsule wall. A homogenous size distribution and uniformity of the capsules was obtained by fractional sieving with a sieve stack. Individual size fractions were obtained by using four different sieves. For further investigations the capsule fraction with 25–36 μm capsule diameter was used because with these size it is possible to detect a reliable visual effect of

Table 1. Wall thickness as a function of $NCO:NH_2$ ratio for different microcapsule batches with the coumarin dye.

Label	$NCO:NH_2$	Wall thickness (nm)	Temperature (°C)
CUM-MC 1	0.067	160	70
CUM-MC 2	0.044	135	
CUM-MC 3	0.035	100	
CUM-MC 4	0.027	60	
CUM-MC 5	0.018	30	
CUM-MC 6	0.009	—[a]	

[a] Wall thickness was not determined because the capsules were not stable to drying.

Table 2. Overview of microcapsules with the TPE dye used for further investigations.

Label	$NCO:NH_2$	Wall thickness (nm)	Temperature (°C)
TPE-MC 1	0.035	100[a]	RT
TPE-MC 2	0.067	160[a]	
TPE-MC 3	0.044	135[a]	

[a] Wall thickness was not determined because the capsules were not stable to drying.

the fluorescent dye caused by the mechanical changes in the elastomeric matrix and to obtain the physical properties of the matrix, compared to the pure matrix without microcapsules inside, as far as possible. Capsules of these fraction show a greater relative deformation before breakage compared to larger ones ($d > 36$ μm) [18]. Thus larger capsules would already rupture during the mixing process (2.2.1). In order to incorporate the aqueous suspension of the washed capsules into the silicone adhesive, the capsules were dried. To obtain free-flowing microcapsules and homogeneous redispersion in one of the adhesive components, protective colloids are added. The different capsule fractions used for the presented experiments are listed in tables 1 and 2.

2.1.2. Confocal laser scanning microscopy of microcapsules. First quality control measurements were performed with a Leica DMI 4000 B confocal laser scanning microscope (CLSM, Leica Microsystems GmbH, Wetzlar, Germany). The capsules were placed out of the dispersion and were measured in wet as well as in dried state with 405 nm excitation wavelength and 450–600 nm emission filter settings of the fluorescent mode and also with the transmission mode. The CLSM measurements were used to evaluate the approximate size distribution, the homogenous dye filling and the drying stability directly after the synthesis. Later they were used to perform long-term stability experiments.

2.1.3. Scanning electron microscopy (SEM) of microcapsules. The SEM images were recorded on NEON40 and Ultra SEM (Carl Zeiss Microscopy GmbH, Oberkochen, Germany). The microcapsules were placed out of the dispersion on the sample holder and dried. After drying, the samples were coated with a 3 nm thick platinum layer. Finally, the capsules were cut using a scalpel.

2.1.4. Dynamic differential calorimetry (DSC) of microcapsules. The thermal characterization was performed with DSC using the device DSC1 (Mettler Toledo, Gießen, Germany). An aluminium crucible was filled with a small amount of dried microcapsules and heated from room temperature to 300 °C under nitrogen atmosphere with a heating rate of 10 K min^{-1}.

2.1.5. Nanoindentation of microcapsules. The indentation experiments were carried out with an MFP-3D AFM from Asylum Research (Santa Barbara, CA, USA) coupled with an inverted microscope Observer Z1 from Zeiss (Jena, Germany). Instead of a conventional cantilever, a nanoindenter (spring constant of 578.494 N m^{-1}) with a sapphire sphere (diameter 800 μm) was used. The inverted optical microscope was used for the positioning of the immobilised microcapsule and to determine the size before the measurement.

For the indentation experiments, a sufficient number of loosely distributed microcapsules were placed on a slide. The capsules must also be immobilised on the glass surface. For this purpose, a few drops of a polyethyleneimine solution (PEI, Mw 25.000 g mol^{-1}, 1 g l^{-1}, Sigma Aldrich, USA) were added to the slide and distributed homogeneously as a liquid film. After evaporation of the solvent, a thin polymer film remains on which a few drops of the capsule dispersion are added. After drying, excess and non-immobilised microcapsules were removed by distilled water and an argon stream. For the actual measurement, an exposed microcapsule is selected and positioned right below the centre of the probe. The deformation of the microcapsule is recorded as force–deformation curve. The sensitivity of the force-sensor (inVols) was measured before and after the experiment.

2.2. 'Smart adhesive'

2.2.1. Processing of microencapsulated dye. For the preparation of the microcapsules containing adhesive a two-component room-curing silicone adhesive is used. Contrary to commercial systems for structural glass constructions, which are often opaque due to their fillers, this adhesive is translucent. The microcapsules were added at a concentration of 2.5 and 5 vol.-% respectively to component A of the silicon adhesive followed by stirring for 10 min. For this purpose a vacuum tumbling mixer (SpeedMixer® DAC 700.2 VAC-P, Fa. Hauschild) is applied to achieve homogeneous distribution of microcapsules within the silicon and to remove the entrapped air bubbles. Then, a stoichiometric amount of component B (1:10 by weight) was added followed by stirring for 5 min. It is particularly critical in the processing of microcapsules that they fail at defined stress and strain conditions in the adhesive, but at the same time can withstand the processing process

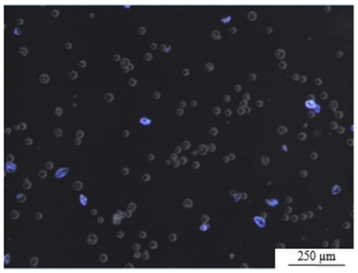

Figure 2. Superposition of the light and fluorescence channel of the adhesive system with fractioned microcapsules after processing. Grey: intact microcapsules, blue: damaged microcapsules which fluoresce under UV-light.

with low damage. To confirm that the microcapsules remain intact during the mixing and application process and are suitable for further investigations, a piston dosing pump was used and the adhesive/microcapsule dispersion was applied with a pressure of 100 bar. Figure 2 shows the superposition of the microscope images (total capsules) with the fluorescence signal (damaged capsules). It can be seen that the highest proportion of the microcapsules remain intact after processing.

2.2.2. Thin-film tensile testing. To evaluate the behaviour of the microcapsules beyond a (locally acting) indentation test, a new method was developed, which allows the deformation behaviour of the microcapsules to be studied under a strain load. For this purpose, the microcapsules are embedded in a model elastomer (polyurethane). The resulting thin elastomer film is then stretched by a self-constructed stretching device, see figure 4. By coupling this with light microscopy, the deformation of the microcapsules can be observed at different elastomer elongations. Figure 3 shows the basic methodology in a sheme.

2.2.3. Mechanical characterization of the 'smart adhesive'. For the mechanical tests the steel substrates were mechanically polished thoroughly by corundum. Then the surface was cleaned with fibreless tissue paper soaked in isopropanol. The bondings were performed with a two-component silicon resin unfilled and filled with the synthesized microcapsules. The adhesive behaviour was studied by carrying out tensile tests according to DIN ISO 37 with a strain rate of 0.001 1 s^{-1}. All tests were carried out at room temperature. The results reported are an average of at least five measurements with standard deviation.

To observe the deformation of the microcapsules *in situ*, a device was built which allows bonded glass point holders to be stretched under the microscope (figure 5). A point holder is used to fix the glazing to a substructure. The point holders used in this project consist of a stainless steel plate with a thread. The stainless steel plate is bonded to a glass substrate with an adhesive layer thickness of 6 mm. Then the point holder is screwed down at the threaded rod of the device, then it is stressed by the towing device.

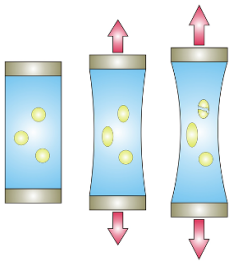

Figure 3. Scheme of the deformation behaviour of the microcapsules during elongation.

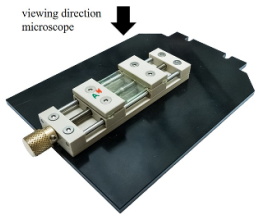

Figure 4. Stretching device for the installation in a light microscope (self-constructed).

3. Results and discussion

3.1. Capsule morphology especially with regard to wall thickness

After a number of different capsule syntheses, the isocyanate-amine ratio was varied in order to specifically influence the wall thickness. Figure 6 shows SEM pictures of a broken capsule which make it possible to measure the wall thickness.

The values determined for the wall thicknesses are shown in table 1. The wall thicknesses show a direct dependence on the isocyanate-amine ratio used. This means that the wall thickness and thus the mechanical properties of the microcapsules can be directly influenced during synthesis. Due to preparative difficulties and only a small number of measurable microcapsules, the values obtained here are only to be considered as approximations. The microcapsules of batch CUM-MC 6 were very thin-walled, so that they collapsed during drying and thus could not be measured. The listed wall thicknesses were measured at least five times, then the average value is calculated.

During the production of the microcapsules, it was found that the increased temperature during synthesis often leads to incompletely filled or even dented capsules. This is probably due to the high vapour pressure of the xylene at high temperatures during the wall forming process and the low vapour pressure after cooling down to room temperature afterwards. Therefore, microcapsules with a selected NCO:NH_2 ratio were produced at room temperature in the next step.

3.2. Release behaviour

As the DSC diagram demonstrate dense capsules with good thermal stability can be obtained by interfacial polymerization. No significant weight loss occurs between room temperature and 180 °C (figure 7). Only when the temperature

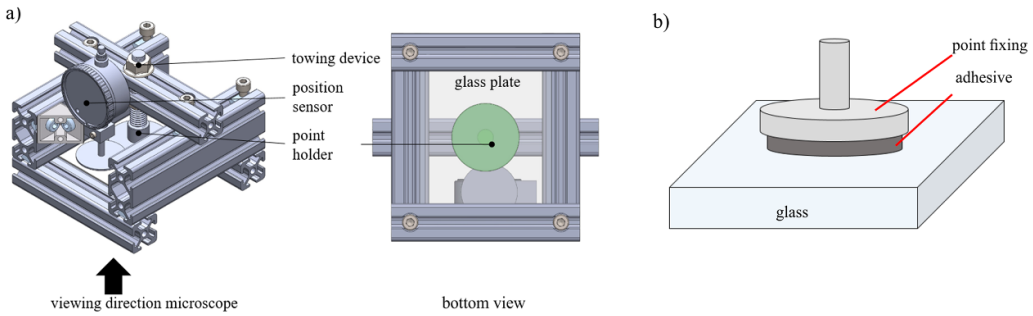

Figure 5. (a) Self-constructed device for stressing adhesively bonded point holder under microscopic observation and (b) bonded point fixing.

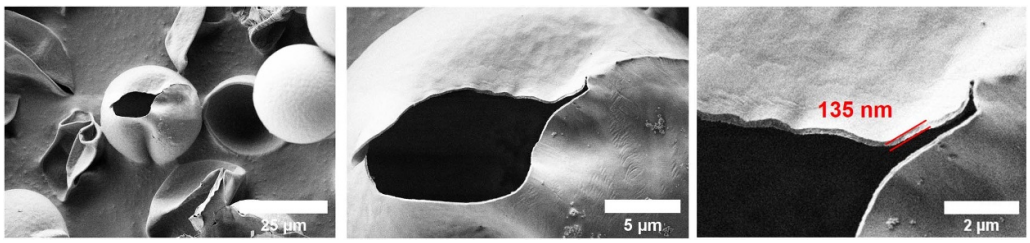

Figure 6. SEM images of broken microcapsule for estimation of the wall thickness.

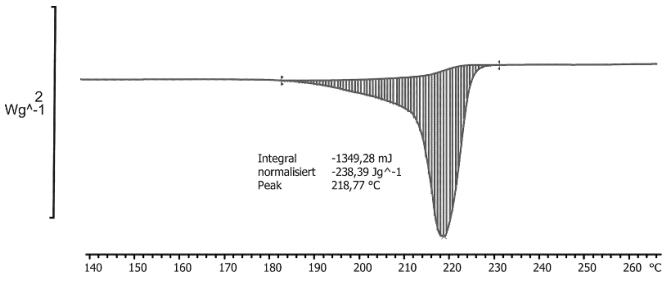

Figure 7. Thermal analysis of microcapsules TPE-MC 2 by DSC.

exceeds 180 °C, volatile constituents (e.g. residues adhering to the capsule surface) begin to evaporate. The endothermic peak at 218 °C (release temperature) indicate that the core solvent evaporates. The masses were determined before and after the measurement and were correlated to each other. A mass decrease occurs which additionally provides information about the microcapsule payload. Microcapsule payload is defined as the weight ratio of core material to the overall capsule weight.

Furthermore, it was determined if the ratio of isocyanate to amine has an influence on the release behaviour. The results match with the thickness of capsule walls identified by SEM (figure 8). Capsules with a high release temperature have thicker capsule walls. Thus, by variation of the ratio of isocyanate to amine the release behaviour of the microcapsules can be adjusted and so it can be adapted to the failure behaviour of the adhesive.

3.3. Deformation and breakage behaviour

3.3.1. Capsule behaviour during nanoindentation. For a sensitive damage detection the deformation and breakage behaviour of the microcapsules play a major role. The micromechanical behaviour of the microcapsules was tested using nanoindentation.

Three force–deformation curves were selected as examples for the three different mechanical fractions, shown in figure 9. The black curve has the steepest increase in the force–deformation diagram and the capsule shell is very stiff. The blue curve shows a softer capsule than the black one and the red curve a bursting capsule. This bursting is visible through the integrated optical microscope. It was also found that uniform bursting requires bulging of the capsule wall. The capsules in figure 9 have most likely obtained their structure through shrinkage processes. Bulging capsules could be well obtained by proper solvent and by synthesis at room temperature. With further increase of the applied force on the capsule, the plastic regime is reached until critical deformation and capsule failure or dye leakage, cf figure 9. By combination with an inverted optical microscope a selected capsule can be observed before and after nanoindentation (figure 9(b)).

Figure 10 shows the force–deformation curves of selected microcapsules of batch CUM-MC 1–5. The 20–30 capsules were measured per fraction, and a mean curve was shown in each case. In addition, the wall thicknesses determined via SEM and the respective isocyanate-amine ratio of the corresponding batch is presented. With decreasing isocyanate content, the force required for deformation also decreases, because the capsules have a thin wall thickness.

Thus, by changing the ratio of isocyanate to amine, it was possible to adjust the wall thickness and also the mechanical strength. When using microcapsules with a small wall thickness, even low forces are sufficient to break the capsules.

As described in 2.1, the microcapsules synthesised at 70 °C are dented and not bulging, which is caused by a loss of

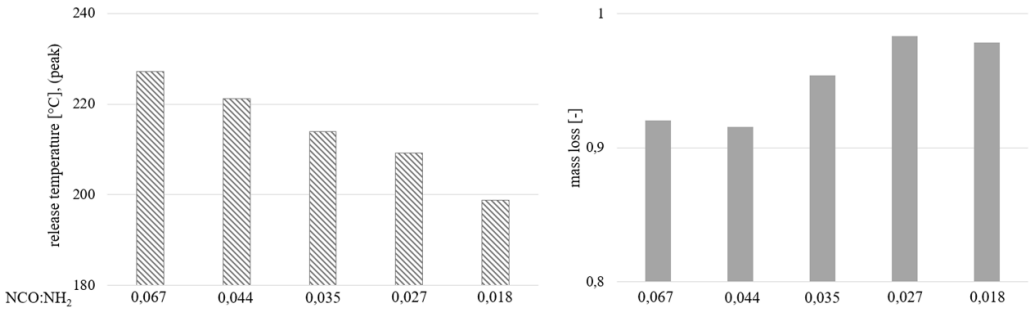

Figure 8. Influence of the ratio of isocyanate to amine on the release behaviour of the microcapsules.

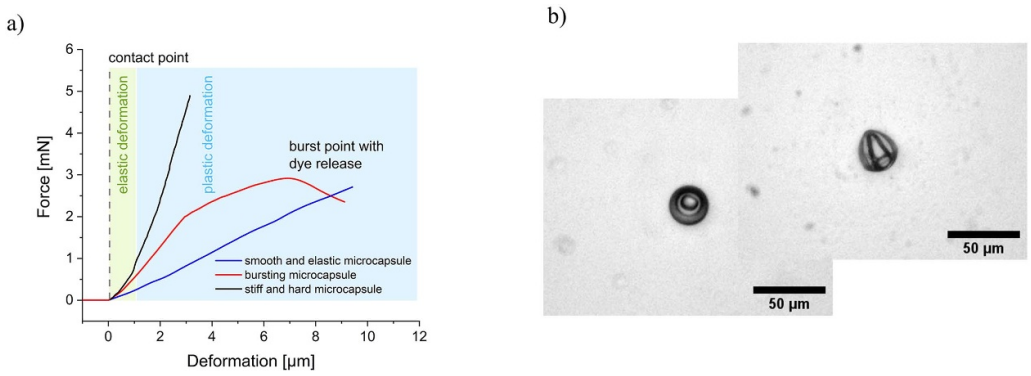

Figure 9. (a) Force–deformation-curves of different microcapsules, (b) pictures of one capsule (TPE-MC 2) before and after nanoindentation.

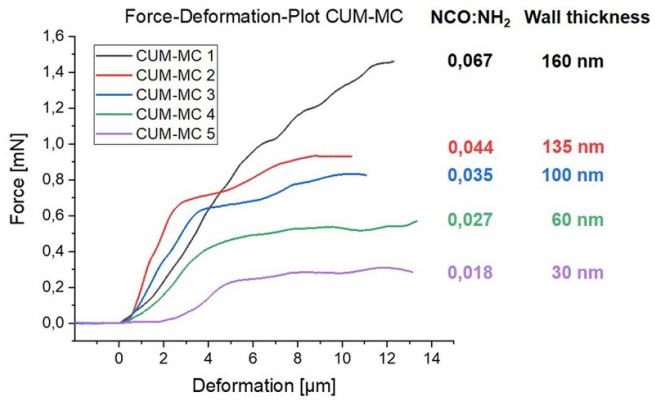

Figure 10. Force–deformation curve of different microcapsule charges with the wall thickness determined by SEM.

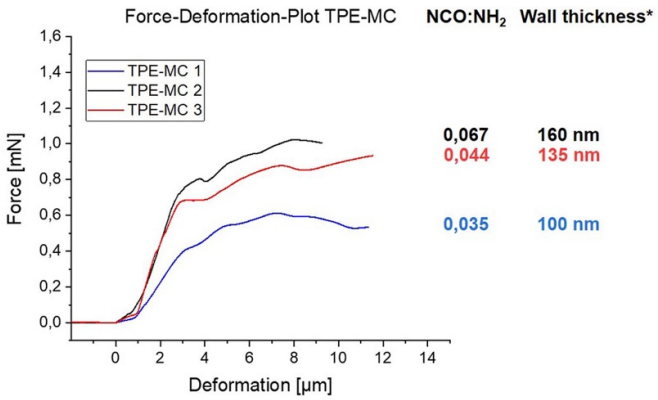

Figure 11. Force–deformation curve of different microcapsule charges.

volume during the cooling process. To avoid this, the synthesis of the microcapsules was repeated at room temperature. Figure 11 shows the curves of the microcapsules with different NCO:NH_2 ratios (0.0667, 0.044 and 0.035), which were produced at room temperature. The examinations show that the wall thickness of the microcapsules decreased with decreasing NCO:NH_2 ratios. When added to the adhesive, these capsules break during mixing, so only the microcapsules with a NCO:NH_2 ratio >0.044 were used for further investigations.

In principle, the curves are very similar, with identical force progression and deformation. But during the individual measurements of TPE-MC1–3, the bursting and release of the dye could be perceived visually much better than during the measurements with the microcapsules synthesised at 70 °C. This is also shown by the curve progression of TPE-MC 1 and 3. After an increase of the curves a falling is detectable. In contrast, the microcapsules that are not filled to the bulging point are reminiscent of being squeezed out.

3.3.2. Capsule behaviour in thin elastomeric films. The method was first used to investigate delamination phenomena in more detail. It was observed that capsule-matrix delamination phenomena did not occur below 50% elastomer elongation in any case. These occur at 75%–89% elastomer elongation at the earliest. What this looks like in practice is

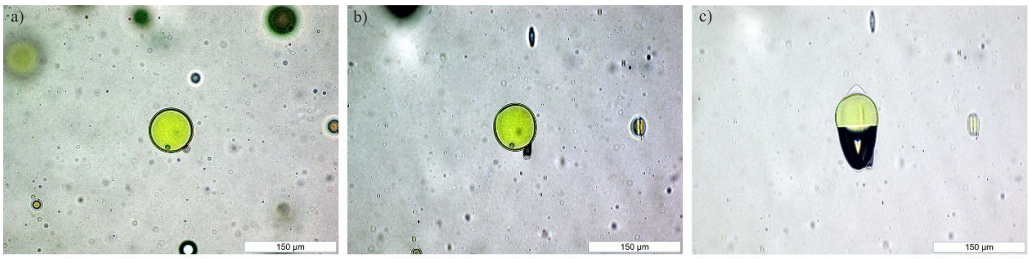

Figure 12. Example of capsule elongation in (a) initial condition, (b) at 100% and (c) 150% Cauchy strain of the surrounding elastomeric matrix.

Figure 13. Example of capsule bursting in (a) initial state, (b) at 100% and (c) over 100% Cauchy strain of the surrounding elastomeric matrix.

illustrated in the following pictures, using the example of a stiff, hardly stretchable capsule filled with the coumarin dye (figure 12). That is why there is no visual difference between the size of the capsules in the left and middle picture.

With this method it was also possible to determine the deformation behaviour of the microcapsules under different elastomer strains. Depending on the mechanical properties of the capsule shell, different strain rates of the model elastomer are necessary to achieve a deformation of the microcapsules. In addition, cracking of the capsule shell was observed in some microcapsules when a critical strain was exceeded. This was followed by a clear dye leakage and discolouration of the elastomer in the capsule environment. Figure 13 shows examples of different elongations of the model adhesive and the microcapsules with the resulting dye leakage when the capsule shell tears.

Within the stretching tests, it could be confirmed that the mechanical properties of the microcapsules vary to some extent. Depending on the elastomer used, the measurement setup allows a clear differentiation between hard, brittle and very soft elastic microcapsules according to the subsequent application properties. The wrinkling observed during nanoindentation could also be confirmed by the stretching tests. By measuring the change in length and the necking of the microcapsules as a function of elastomer elongation, the elongation behaviour of the microcapsules in a thin model substrate can also be described quantitatively.

Figure 14 shows the elongation of the microcapsules as a function of elastomer elongation for microcapsules with a different ratio of NCO:NH_2. As already described, these microcapsules differ in isocyanate content (table 1). The resulting different wall thicknesses have a direct effect on the deformation behaviour of the microcapsules. With the same force application, a higher wall thickness shows a significantly lower degree of deformation. The results

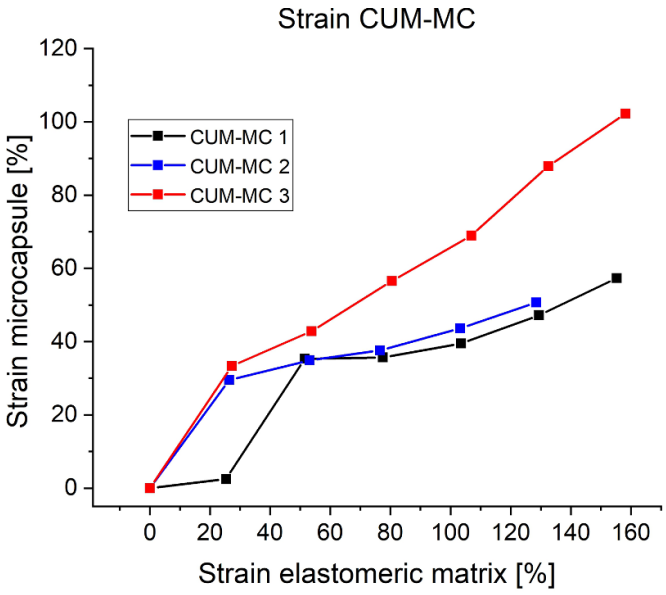

Figure 14. Elongation behaviour of microcapsules with a different ratio of NCO:NH_2.

obtained during the elongation tests also confirm the results of the nanoindentation.

3.3.3. Influence of microcapsules on the mechanical properties of the adhesive. To further examine the formulated adhesives, especially to ensure that the mechanical properties did not changed during the formulation process, tensile tests with pure substance samples (following ISO 37–1 A) and shear tests (following DIN 14 869) were carried out. Figure 15 shows the stress–strain curves of the pure silicon adhesive as well as for the silicon adhesive

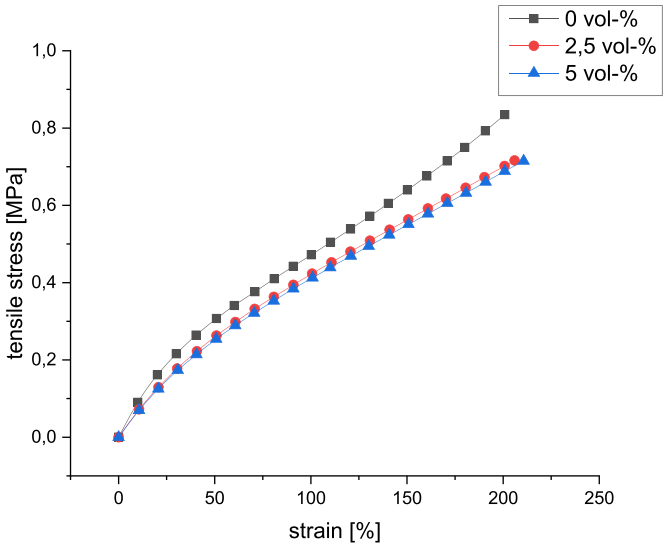

Figure 15. Results of the tensile tests of the unfilled adhesive (grey), the adhesive filled with 2.5 vol% capsules (red) and 5 vol% capsules (blue) of batch TPE-MC 3.

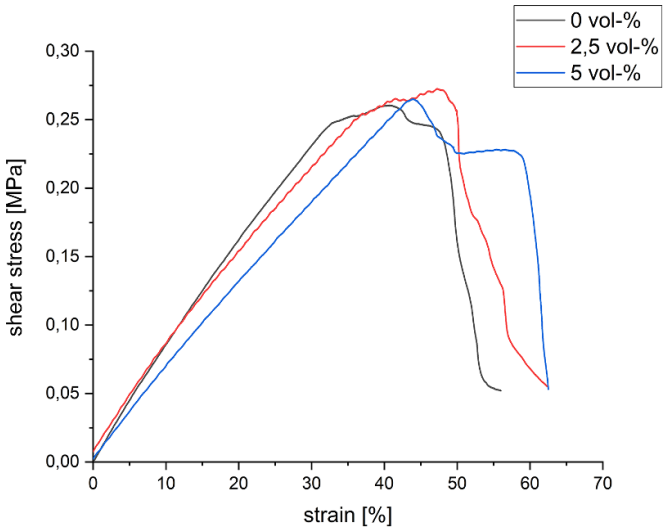

Figure 16. Results of the shear tests of the unfilled adhesive (grey), the adhesive filled with 2.5 vol% capsules (red) and 5 vol% capsules (blue) of batch TPE-MC 3.

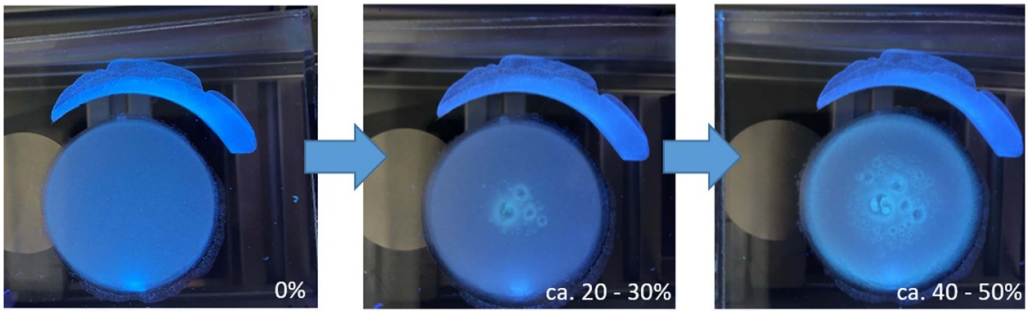

Figure 17. Point holder under tensile stress. The fluorescence increases with increasing tensile stress.

with different concentrations (2.5 vol%, 5 vol%) of encapsulated microcapsules. The tensile stress of the filled systems is reduced by 20% compared to the pure silicone, but the inelastic strain capacity and the toughness of the adhesive increased.

In figure 16 the results of the shear tests of the pure silicon adhesive as well as for the silicon adhesive with different concentrations (2.5 vol%, 5 vol%) of encapsulated microcapsules are presented. The silicon adhesive filled with 2.5 vol% capsules shows a higher stiffness than the unfilled

system. Furthermore, the elasticity of the adhesive filled with capsules is higher. A cohesive failure of all samples was detected.

3.3.4. Further investigations on the mechanical behaviour. To demonstrate the feasibility of the approach a point holder is fixed in a special device (figure 5). When the TPE-filled microcapsules in the adhesive bond of a point holder are irradiated under UV light (hand lamp, wavelength approx. 365 nm), the effect of the smart adhesive can be observed particularly well (figure 17). In the unstressed state, the specimen fluoresces only slightly. With increasing tensile stress, defects occur particularly inside the point holder, i.e. the area of highest stress. This leads to an increased fluorescence. Fluorescence enhancement can be attributed to local detachment and defect formation known as 'voiding' [19]. At the highest tensile stresses fluorescence also increases in the edge region before failure. In this area the deformations are highest, so that the cause of this fluorescence can possibly attributed to a strain-induced release of the fluorescent dye.

4. Conclusion

The presented studies show that the development of an intelligent adhesive for visual damage detection is a major challenge. All components must meet different requirements and have been extensively characterized. Homogeneous microcapsule fractions with defined stiffness and adjustable release behaviour could be produced, which are free-flowing, storage-stable and can be uniformly resuspended in an adhesive component. A suitable dye was found that is released within the adhesive in case of a microcapsule damage and becomes visible when illuminated with UV-light.

The preliminary tests on the influence of microcapsules on the quasi-static properties of the adhesive show a reduced tensile strength. However, the Young's modulus of the adhesive composite is not influenced. According to current knowledge, the sensory adhesive developed is suitable for use in structural glass engineering and exhibits reproducible strength and adhesion properties on glass and stainless steel. By varying the synthesis parameters, it is possible to adapt the required strength to the boundary conditions resulting from the use in glass facade bonding.

The study has impressively shown that a non-destructive method can be developed to visualize overstressing and damage to structural bonded joints, but there is still a great need for further investigations. In glass façade construction the shear stress is only transferred by the bond, when there are no additional mechanical advices. So shear loading is an important point that should be examined in the future. Furthermore, it is planned to investigate the area of strain rate (and also temperature) dependence within the adhesive more closely in prospective work.

Extensive additional investigations are still required with regard to manufacturability and durability before the product is ready for the market.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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