Emission profile of butan-2-one oxime from commercially available neutral silicone sealant

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ABSTRACT

The paper presents results of research on emission profile of butan-2-one oxime (methyl ethyl ketoxime, MEKO) released from two types of commercially available neutral silicone sealants: silicone type 'S' and silicone type 'T'. To determine the emissions from silicone samples, the system consists of a stationary emission chamber, and the thermal desorption-gas chromatography-mass spectrometry system was applied. First, the research aimed to describe the emission profile of butan-2-one oxime considering different methods of application of silicone caulk: separate strips, merged strips, and dotted. Silicone sealant was applied in these three different ways to produce varying surfaces. As a result, significant differences in the emission profile were observed for the different methods of silicone application. The greatest difference in the emission rate was found to be between those of the dot shape and those of the separate strips; the emission rate was approximately five times smaller for dot shape. Moreover, the studies focused on quantitative emissions of butan-2-one oxime over a set time period. The research results indicate that, in the emission process of butan-2-oxime from a neutral silicone sealant, there are three main stages of emission. The first stage is dynamic emission, occurring during the first few days after application. The second stage is declining emission, and the third stage is stable residual emission. Between the first and third stages, the amounts of emitted oxime differ significantly. The difference in the emissions of butan-2-one oxime between the first and last stages of emission ranged from factors of 100–1000.

1. Introduction

Recently, as an increasing number of people spend most of their time indoors [1,2], modern housing construction strongly emphasises energy management and the regulation of indoor conditions, such as temperature, humidity, and air-exchange ratio. These indoor environmental factors significantly affect human health and the perception of indoor air quality. Indoor environmental factors may also significantly impact the dynamism of the emissions of chemical compounds (with respect to both type and amount) from building and constructing materials [3–6]. Despite the existence of common indoor solid materials (wood, brick, plastic, tile) and coatings, such as paints, varnish, and wood laminates, adhesives such as glue, resins, and water-vapor curing materials like silicone sealants are also employed in enclosed areas. In the case of silicone application during indoor activities, the temperature and humidity affect the curing rate. During the curing process of silicone materials, several harmful chemical compounds are emitted such as acetic acid, propan-2-one oxime, butan-2-one oxime, or cyclohexylamine. The mentioned sealants are an important issue because they

are commonly used to increase the thermal isolation of buildings and to waterproof household elements. Improving the sealing around windows and doors decreases the heat loss through thermal bridge and stabilises temperature fluctuations in the building. Furthermore, improvements in sealing technology limit the diffusion of water vapor inside and outside of buildings. Sealing the installations (water pipes, ventilation, air conditioning, bathroom accessories) and spaces inside the building is necessary to protect the rest of the building against flooding and penetration of water vapor. This forces the introduction of fast-curing silicones in housing construction.

The most commonly applied sealants for waterproofing are silicones, such as liquid foils, silicone adhesives for tiles, and silicone caulk. One popular type of silicone caulk sealant available on the market is room-temperature-vulcanisation (RTV) silicone sealant. RTV silicone [7] is often used because it does not require special skills for application and is characterised by its non-time-consuming application procedure. RTV silicones cure via condensation, with water vapor contained in the air under the room temperature. During the condensation cross-linking reaction of RTV silicones, several chemical

compounds [8] may be emitted directly to the gaseous phase, thus affecting the quality of indoor air [9,10]. The humidity of the building, as well as the concentration of chemical compounds, such as volatile organic compounds (VOCs), in the indoor environment, may be directly associated with the well-being of its residents [11]. The interaction between VOCs and the appropriate humidity level may cause the formation in indoor air of oxygenated VOCs from water-soluble organic gases (WSOGs). Silicone sealants are mainly used in enclosed spaces that are exposed to continuous or cyclical contact with water (e.g., kitchen, bathroom, garage). The accumulated humidity may create an aqueous micro-environment in which WSOGs are soluble and undergo further oxidation reactions [12,13]. RTV silicones can be divided in three groups depending on their pH: (i) acidic, (ii) alkaline, and (iii) neutral. In the acidic group, the most commonly emitted compounds are characterised by acetic properties, which may be the cause of the corrosion of steel or ceramic elements and are characterised by a specific irritating acetic smell. The use of acidic silicone as sealants in steel installations may cause steel corrosion, thereby unsealing connections and failure of the joints. Moreover, another issue is the destruction of decorative coatings on fittings, via the corrosion of ceramic tiles and decorative elements. The next group of RTV silicones are defined as alkaline silicones. This group is characterised by side-group amines in the main chain of the siloxane polymer. Amines and aminoxy silicones are largely used in the automotive industry and have specific outdoor applications, like sealants for high-voltage stations. Alkaline silicones are characterised by a specific ammonia or amine odour. Amines and aminoxy compounds may be the cause of the corrosion of polymer materials and can be harmful to humans.

The last group of RTV silicones are neutral silicones, in which the 'neutral' emitted compound can be one of four groups of compounds: alkoxy, enoxy, ester, or oxime. The most common commercially available silicones are oximes or alcohols. Silicones with a neutral curing system are a common odourless alternative for acidic or amino silicones. 'Neutral' silicones are less corrosive than acidic ones, and less harmful to humans than amino silicones. According to available safety data sheets for neutral silicones, they contain or may emit a few chemical compounds such as butan-2-ol, propan-2-one oxime, and butan-2-one oxime. Butan-2-one oxime is mostly used as an anti-skinning agent in the paint industry and as a reagent for the production of cross-linking agents, e.g., *syn*-O,O′-di(butan-2-one oxime)dimethoxysilane, used in neutral silicone. Butan-2-one oxime has been the subject of many studies [14,15], owing to the possible cancerogenic properties of the compound. The substance-evaluation report [16] of butan-2-one oxime (methyl ethyl ketone oxime, MEKO) presents several studies [15,17] on the non-human harmful properties of this oxime. The results of these studies show that exposure to butanone-2-oxime may cause dizziness, respiratory problems, and possible formation of liver tumours.

The main aim of this study was to quantitatively measure the emission of butan-2-one oxime from neutral silicone samples. To do so, it was necessary to synthesise, under laboratory conditions, the butane-2-oxamine oxime standard. Studying the emission profile over time aids in understanding and describing the stage of emission. The second part of the study was focused on investigating whether the ratio of the surface area to the mass of the sample affects the first stage of dynamic emission as well as the last stage of residual emission.

2. Materials and methods

2.1. Chemicals, reagents, and instruments

In this study, the following chemical reagents were used: pyridine (99.9%, POCH, Poland), ethanol (99.9%, POCH, Poland), butan-2-one (technical grade, 98%, Sobieski, Poland), hydroxylamine hydrochloride (ACS reagent, 98.0%, POCH, Poland), and ethyl acetate (99.5%, POCH, Poland). The following analytical devices and tools were employed: stationary environmental test chambers (Markes International Micro-

Chamber/Thermal Extractor M-CTE250, UK), a thermal desorber (Unity v.2, Markes International Ltd., Pontyclun, UK), stainless-steel tubes filled with Tenax TA (MARKES) (average mass of sorption medium = 200 mg, USA), a gas chromatograph (Agilent Technologies 6890), a mass spectrometer (5873 Network Mass Selective Detector, Agilent Technologies), and a 60 m × 0.25 mm × 1 μm capillary column (Agilent 122-5563, J&W DB-5MS, USA).

2.2. Synthesis of the butane-2-oxamine oxime standard

It was necessary to synthesise a butan-2-one oxime standard. First, 2.2 mL of pyridine (99.9%, POCH, Poland) was dissolved in 100 mL of ethanol (99.9%, POCH, Poland). Then, 2.1 mL of butan-2-one (technical grade, 98%, Sobieski, Poland) and 2 mL of hydroxylamine hydrochloride (ACS reagent, 98.0%, POCH, Poland) were dissolved in ethanol. A mixture of pyridine, 2-butanone, and hydroxylamine hydrochloride in ethanol was placed in a round-bottom flask, combined with reflux condenser, and heated for 12 h. Next, the post-reaction mixture was mixed with ethyl acetate (99.5%, POCH, Poland) in a glass separatory funnel to transfer the oxime into the organic phase. Then, sodium sulphate was added to the organic-phase oxime to remove residual water. The final stage of the reaction was to remove the solvent by evaporation under reduced pressure. The reaction yield of described synthesis was 72%. The general reaction scheme of the synthesis of the butan-2-one oxime standard is shown in Fig. 1.

2.3. Preparation of silicone sealant sample

Two commercially available RTV silicone sealants with a neutral cross-linking system were used to study the emission profiles. The first silicone was labelled as silicone 'S' and the second as silicone 'T'. Silicone 'S' contained approximately 1% of butan-2-ol as solvent, according to the datasheet. Silicone 'T' used *syn*-O,O′-di(butan-2-one oxime)dimethoxysilane as the curing agent. Two series of samples were prepared. The first series was used to test the impact of sample geometry on the emission profile. The second series was used to investigate changes in emission profile over time.

The mass of the prepared samples was approximately 650 ± 50 mg of silicone sealant; the samples were placed in aluminium foil containers. The first set of samples was prepared with three different application techniques. Two samples were prepared for each application technique. The schematic models of silicone surface coverings are shown in Fig. 2. For the first technique (i), silicone was applied in parallel continuous strips without merging the surfaces of strips. For the second technique (ii), silicone was applied in parallel continuous strips, merging the surfaces between strips. In the third technique (iii); silicone was applied in dots. The three techniques described are different in terms of their area-to-mass ratios; the first should have the largest area, the third should have smallest due to its geometry, and the second should have an area-to-mass ratio between that of the first and third techniques.

The second set of samples was prepared to study the emission profile of butan-2-oxime. Two commercially available silicone sealants with the neutral curing system were used to prepare the samples. The samples were prepared using the first application technique and placed into disposable aluminium foil containers; the mass of a single sample was approx. 600 ± 50 mg. After application and between
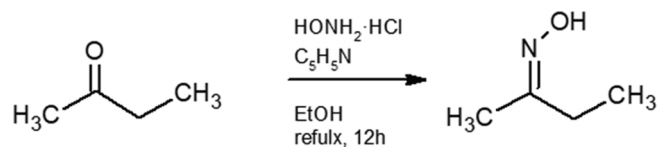

Fig. 1. Simplified reaction scheme for the synthesis of butan-2-one oxime from butan-2-one.
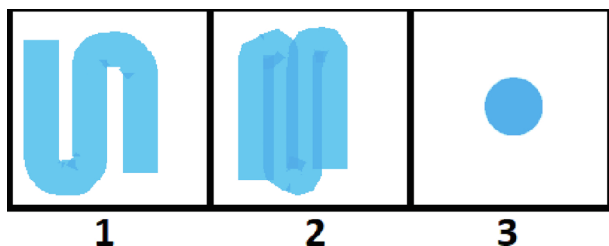
Fig. 2. Overview of three sample application techniques: (1) largest surface-to-weight ratio; (2) medium surface-to-weight ratio; (3) smallest surface-to-weight ratio.

measurements, the samples were placed and seasoned in polyethylene plastic containers. The plastic containers were open for 1 h before the sample was placed in an environmental chamber for measurement. The samples were prepared in pairs, from silicone 'S' and silicone 'T'; four pairs of samples were prepared in 24-h intervals.

2.4. Analytical equipment parameters and working conditions

To analyse the emission of butan-2-one oxime from silicone sealant samples, the following procedure was applied:

1) The first stage of the analysis was sampling the gaseous phase. The container containing the silicone sample was opened 1 h before insertion into the emission chamber. In the meantime, the external standard of ethylbenzene was introduced directly on the sorption bead. The amount of ethylbenzene introduced inside the tube was 866 ng, following the procedure and dedicated analytical equipment described in detail in previous papers [18,19]. Then, the silicone sample was placed inside the chamber for 5 min to stabilise the temperature; afterward, the stainless steel tube with Tenax TA was placed in the outlet. The analytes were collected for 5 min. During the conditioning and sampling process, the chamber's working conditions were a temperature of 30 °C and an inert gas-flow rate (dry nitrogen) of 45 mL·min^{-1}.

2) The second step of the applied procedure was two steps of thermal desorption of the analytes collected and retained on the employed sorption tubes. The tubes were placed in a thermal desorber oven to perform desorption/extraction of the analytes from the sorption medium to the gaseous phase at a temperate 280 °C for 10 min under a helium flow rate of 50 mL·min^{-1}. Liberated analytes were transferred to the multibed micro-trap filled cooled at 1 °C. After this, the analytes preconcentrated on the microtrap were rapidly thermally desorbed by heating up to 300 °C for 5 min. During this process, the analytes were liberated from the micro-trap sorption medium and transferred directly to the chromatographic column by helium gas at a flow rate of 1.5 mL·min^{-1}. The transfer line (TD-GC) temperature was 150 °C, and the analytes were introduced using the splitless and split modes: 1:1, 5:1, 11:1, 21:1, and 203:1. Following splits were set up due to the large emission rate in the first day and large decrease during next days of the study.

3) The last step was the separation, identification and determination of the analytes retained on the sorption medium. The applied GC–MS working parameters were as follows. (i) Oven temperature program: 60 °C for 0.5 min, then heating 15 °C·min^{-1} to 280 °C, which was maintained for 10 min. Then, the temperature was maintained at 300 °C for 10 min to flush possible siloxane origins from the silicone sealant. (ii) Parameters of the mass spectrometer: ion source temperature was 230 °C, the quadrupole mass analyser temperature was 150 °C, and the GC–MS transfer line temperature was 280 °C. Data were collected in the SCAN mass spectrometer working mode.

The most important steps of described analytical procedure and the general working parameters of employed analytical equipment were shown schematically in Fig. 4.

2.5. Quality assurance and quality control

Before taking new measurements, blank field tests of the used TD-GC–MS system were performed to ensure the correctness and reliability of the conducted research. Each stainless sorption tube was conditioned after analysis in a thermal desorber for 10 min at 250 °C under a constant gas flow rate in the conditioning mode.

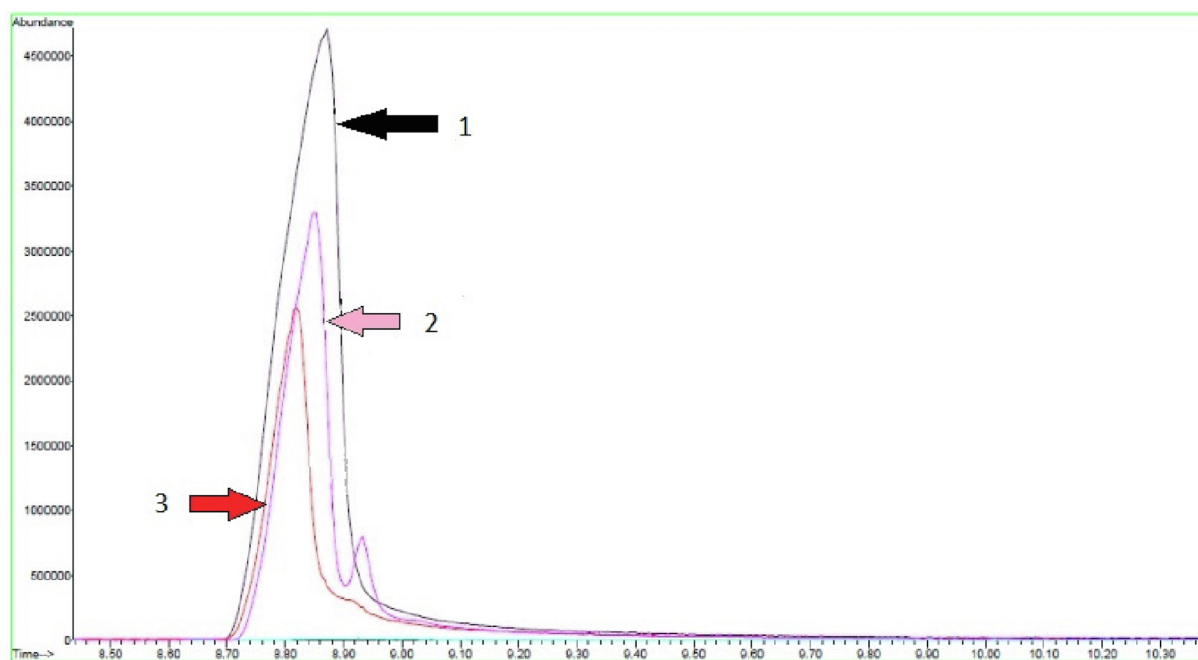An internal standard calibration was performed based on the

Fig. 3. Comparison of three chromatograms obtained for three different silicone caulk application techniques: (1) first technique without merging the surfaces of the strips; (2) second technique with merged strips; and (3) third technique applying silicone dots.
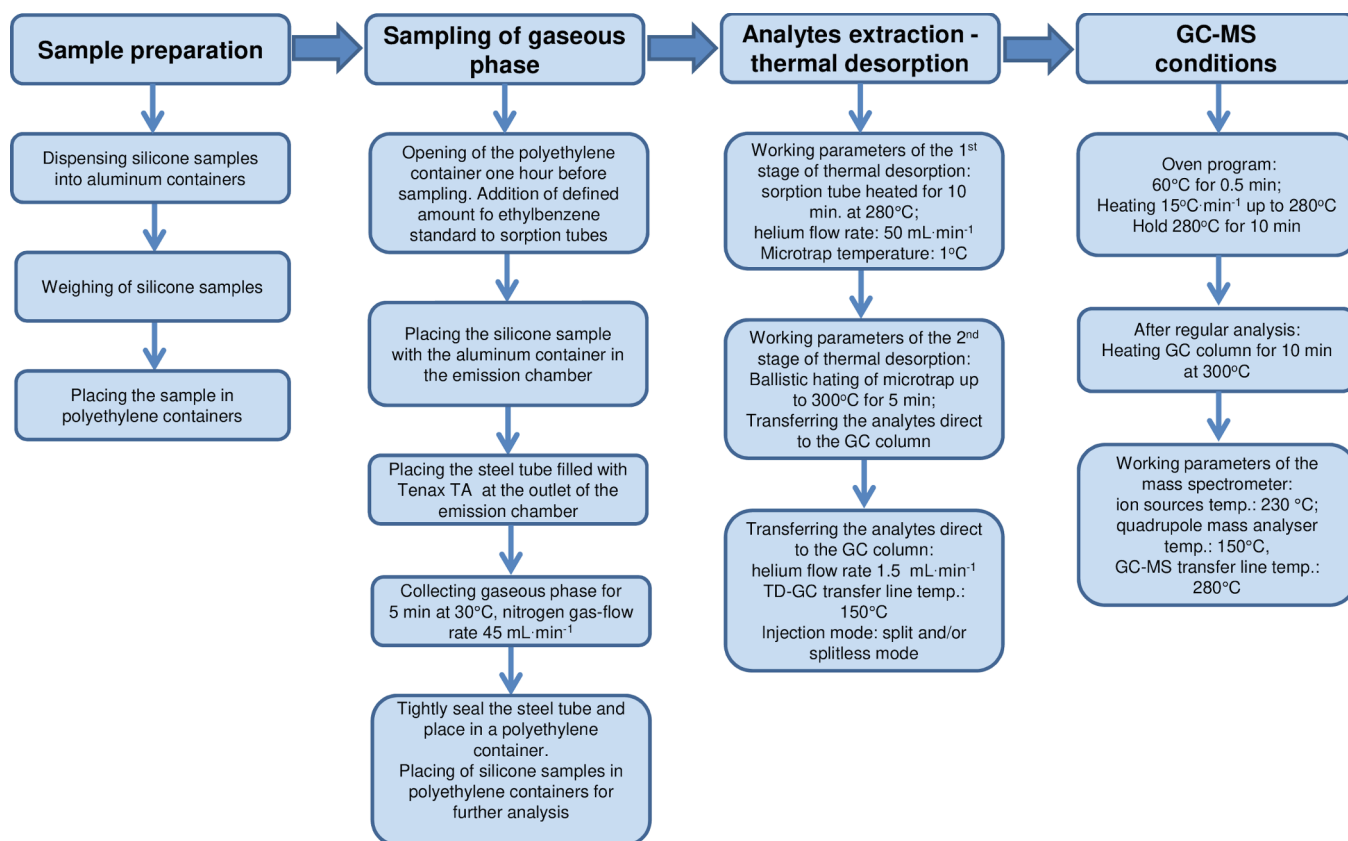
Fig. 4. The general scheme of applied analytical procedure to determine the emission of butan-2-one oxime from silicone sealant samples.

prepared butan-2-one oxime (78% pure) and ethylbenzene (POCH, Poland, 99.8%) dissolved in 1 mL of methanol (POCH, Poland, 99.8%). The external standard solution was prepared by adding 867 µg of ethylbenzene to 1 mL of methanol (POCH, Poland, 99.8%). Calibration solutions were prepared by adding external standard solution to samples, and adding an incremental amount of butan-2-one oxime. The concentrations of butan-2-one oxime in the calibration solutions were 1441, 2162, 2882, 3603, 4324, and 5045 $ng \cdot \mu L^{-1}$. The calibration solution was introduced via a 10 µL glass syringe directly into the sorption medium, Tenax TA, and flushed with inert gas at a flow rate of 40 $mL \cdot min^{-1}$. Each time, the prepared solution was introduced on the sorption medium. More detailed information about the calibration process, applied equipment and its working parameters are described elsewhere [18,19]. The determination coefficient (R^2) of the prepared calibration curve was 0.978.

3. Results and discussion

3.1. Dependence between surface and mass ratio

The main aim of this study was to estimate the volume of butan-2-oxime emitted during the first stage of emission and select a technique that provided the most stable and repeatable emission profile. Three series of silicone samples were made using different sample-dispensing techniques. The samples had approximately equal mass; the difference in area resulted from the different application techniques. The first technique should provide the largest area-to-mass ratio. The second technique included merging of the silicone strips, which should provide a smaller area-to-mass ratio than that of the first technique but larger than that of the third. The last technique produced a dotted sample; this should provide the smallest area-to-mass ratio among these three techniques. The dependence between the surface form with butan-2-oxime emissions was the subject of investigations performed by He

et al. (2019) [14]. Results were acquired for the three different application techniques and are compared in Fig. 3. For this comparison, the area of butan-2-oxime from chromatograms was used. A difference between the first and second techniques could clearly be observed; the second technique's samples emitted nearly three times less butan-2-oxime than those of the first. Samples applied using the third technique emitted five times less butan-2-oxime than samples applied by the first technique.

3.2. Emission profile of butan-2-one oxime

To study changes in emissions of butan-2-one oxime over time, two sets of silicone sealant were prepared. Silicone samples were prepared with pairs of silicone name 'S' and silicone name 'T'. Four pairs of silicone samples were prepared in 24 h intervals. Detailed information about the timeline of sampling is listed in Table 1.

On the first day after application, the pairs of samples were analysed three times: after 1, 2, and 3 h after application. The high frequency of measurements in the first few hours allowed the investigation of the first (dynamic) phase of emission. Then, samples were tested in 24 h intervals, to study long-term emissions.

The results of the emission study for silicon type 'S' and silicon type 'T' are shown in Table 2. The obtained results are presented as average emissions of butan-2-oxime from the silicone sample in $ng \cdot mg^{-1}$.

To describe the long-term emission profile, the percentage of emission (1) was introduced. The percentage of emission describes the decrease or increase in the emissions from the sample and consists of the actual emission from the sample divided by the highest noted emission from the same sample.

$$\frac{actual\ emission\ [ng \cdot mg^{-1}]}{highest\ noted\ emission\ from\ sample\ [ng \cdot mg^{-1}]} \cdot 100\% = percentage\ of\ emission\ [\%]$$

(1)

Table 1
General information about the schedule of sampling for silicone "S" and silicone "T".

Sample name	Weight [mg]	Split during second stage desorption								
		203:1			21:1		11:1		1:1	
		Time of sampling [h]								
Silicone type "S"										
PS1	628.1	1	2	3	24	48	72	96	N.M.	N.M.	168
PS2	523.2	1	2	3	24	48	72	N.M.	N.M.	144	168
PS3	464.1	1	2	3	24	48	N.M.	N.M.	120	144	168
PS4	561.3	1	2	3	N.M.	N.M.	72	96	120	144	168
Silicone type "T"										
PT1	670.4	1	2	3	24	48	72	N.M.	N.M.	144	168
PT2	697.1	1	2	3	24	48	N.M.	N.M.	120	144	168
PT3	544.4	1	2	3	24	N.M.	N.M.	96	120	144	168
PT4	586.8	1	2	3	N.M.	N.M.	72	96	120	144	168

N.M. – not measured.

Table 2
Results of analysis of emission of oxime from silicone type "S" and "T".

Time of sampling	Silicone caulk "S"		Silicone caulk "T"	
	Average mass of oxime per mass of sample [ng·mg^{-1}]	Percent of emission [%]	Average mass of oxime per mass of sample [ng·mg^{-1}]	Percent of emission [%]
1	230.48	100	175.70	82.94
2	171.49	74.41	161.68	76.32
3	143.62	62.31	154.54	72.96
24	3.40	1.48	211.83	100.00
48	2.07	0.90	171.92	81.16
72	2.03	0.88	163.46	77.16
96	2.06	0.89	41.51	19.60
120	0.66	0.28	40.01	18.89
144	0.71	0.31	38.71	18.27
168	0.63	0.27	36.10	17.04

Fig. 5 compares the butan-2-one oxime emissions from the 'S' silicone and 'T' silicone samples. It can observed in graph (1) of Fig. 5 that emissions from silicone 'S' were higher than from silicone 'T' in the first hour. The difference in the emissions between silicones was approximately 11%. Furthermore, the data in graph (1) of Fig. 5 shows that the maximum value for emissions for silicone type 'S' was observed 1 h from the beginning of the test; however, for silicone type 'T', the maximum emission was observed after 24 h. In studies described by He et al. (2019) [14], maximum emissions were observed 5 days after application, which may be attributed to the different application techniques and shapes of the samples. This indicates that the curing rate and maximum emissions are dependent on the application technique and shape of the samples.

The results, shown as a percentage of emission, indicate that the emissions from silicone type 'S' reached a maximum in the first hour and then decreased sharply to approximately 1% of the maximum value and then remained constant until the end of the study. In contrast, the emission profile for silicone type 'T' showed that maximum emissions were observed after 24 h from the beginning of the experiments; the emissions then decreased by 20% and finally remained constant for the next 24 h.

Considering the data presented in Fig. 5, the emission profile for silicone type 'S' can be described as follows. (i) In the first hour, the emission reached the maximum; (ii) there was a decrease of approximately 25% in the second hour and a slight decrease of 12% between the second and third hours; (iii) between the third and 24th hours, there was a large decrease from 62% to 1%; and (iv) after 24 h, the emissions remained quite stable, slightly decreasing from 1% to 0.27%, between the first and 7th day of study.

Considering the data presented in Fig. 5, the emission profile for silicone type 'T' can be described by the following stages: (i) in the first 3 h, the emissions decreased from 83% to 72%; (ii) between the third and 24th hour, the emissions reached the maximum value; (iii) 24 h after the maximum value was reached, the emissions decreased to 81% and then decreased over the next day by 5%; (iv) between the third and fourth day, a maximum decrease from 77% to 20% was observed; and (v) after the fourth day, the emissions remained constant with a slight decrease from 20% to 17%, 7 days from the start of the study.
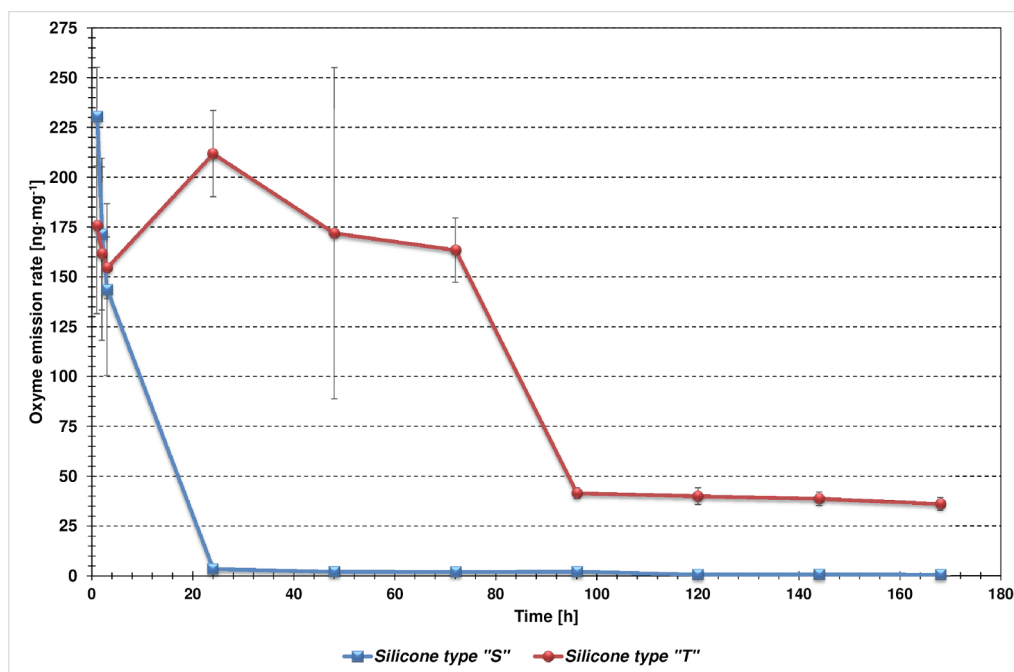

Fig. 5. Comparison of oxime quantity emitted from silicone 'S' and silicone 'T'.

4. Conclusions and future perspectives

The results of this study on the geometry of application and its impact on emissions from silicone gives a reason to conclude that the geometry of the silicone sample has a significant impact on the amount of emitted representative of VOCs and its emission profile. An important factor impacting the emissions is the surface area of the sample. The surface area in contact with air plays an important role in the curing process. The partially cured layer on the silicone surface in contact with the ambient air is called the skin; the time it takes for the skin to form is considered a technical parameter of the silicone sealant. Skin formation is important for the shape retention of the applied silicone; however, it also impacts the surface in contact with water molecules in the air. Skin is a diffusion barrier/layer that determines the ability of water particles to enter the silicone matrix, and thus the number of water particles that can diffuse into the matrix determines the time for silicone to cure. The amount of emitted VOCs originating from the curing agent is limited by diffusion out of the matrix. A study on three application techniques showed that the area-to-mass ratio influences the amount of emitted oxime. However, there were no data to describe the connection between these phenomena.

From this study on the emissions over time, it can be observed that the emissions of oxime from a neutral silicone sealant can be divided into three main stages:

- The first stage is dynamic emission. The results from this study and previous works show that this stage is characterised by extremely dynamic emissions before the maximum value is reached. The dynamics of butan-2-one oxime emissions result from the formation of the skin phase on the surface of the silicone and the entry of water molecules deep into the silicone mass.
- The second stage is a short period of decreasing emissions. It transitions between maximum and residual emissions.
- The third stage is residual emissions. These residual emissions can be the result of sorption and desorption processes in the silicone matrix and the residual presence of the curing agent.

The first stage of emission, owing to its dynamics and highest emission value, can be a risk to employees who work and stay in indoor areas containing silicone sealants or coatings. The third stage of emission may be a risk to residents. The various uses of silicone at home produce different geometries to test.

The relationship between the geometry of the sample and emission profile should be further studied. Long-term emissions from different dosage techniques should be considered. Furthermore, the two main variants of silicone sealant applications, large-area thin films similar to 'liquid' silicone coatings and straps that imitate joints, should be investigated.

In contrast, long-term emissions from silicone sealant samples should also be studied. Following the investigations carried out in this study, we observed that residual emissions occurred from the 3rd to 7th day after application.

As for the future perspectives of described preliminary research, there is a possibility to expand studies to the emission assessment stage depending on the changes of relative humidity (RH), as well as the temperature under real conditions in indoor environment. However, it should be highlighted that in the indoor environment (apartments, buildings, etc.) the RH values mainly oscillated between 40 up to 65%. The RH fluctuations in indoor air are not very dynamic. Lower values of RH might affect the oxime emission profile, mainly increasing the time of polymerization process (which needs the presence of water), as well as the slower process of creating so-called skin/film on the outside, increasing the oxime emission. The conditions of indoor environment such as temperature, RH and ventilation ratio are very specific and characterized by low variability.

CRediT authorship contribution statement

Krzysztof Klewicz: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Resources, Writing - original draft. **Mariusz Marć:** Writing - review & editing, Supervision. **Bożena Zabiegała:** Conceptualization, Formal analysis, Writing - review & editing, Supervision, Project administration.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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