Bundesinstitut für Risikobewertung

Method evaluation study on the determination of volatile compounds in silicone materials

Report on the Inter-laboratory comparison exercise NRL-DE-FCM-02/2020 of the German National Reference Laboratory (NRL) for Food Contact Materials

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1 Summary

The German National Reference Laboratory for Food Contact Materials (NRL-FCM) has organized a method evaluation study (MES) on the determination of volatile compounds in silicone materials (NRL-D-FCM-02/2020). The MES was open to Official Control Laboratories (OCLs) and NRLs.

The present MES aims to assess the analytical capabilities of OCLs and NRLs for the determination of volatile compounds in silicone materials, to evaluate a recently revised test procedure and to propose a measurement uncertainty.

Previous research indicated that parameters like sample conditioning, ventilation/air supply in the oven, weighing pan material, and sample handling may affect the reproducibility and repeatability of the test results. Participants were asked to determine the mass loss of three different silicone materials after tempering for 4 h at 200 °C following the method that was adapted according to the results of previous investigations.

In total, 17 laboratories from seven EU member states participated in this MES, including nine OCLs, seven NRLs, and one university. Sixteen of those reported results. One laboratory was not able to report results due to limitations related to COVID-19.

The assigned values x_{pt} and the standard deviation σ_{pt} were derived from the mean results as reported by the participants as robust average and relative standard deviation according to the Q/Hampel method.

Laboratory results were rated using z scores according to ISO 13528:2015. Almost all laboratories that followed the stipulated test method achieved satisfactory results.

Four of the initially received 16 data sets were excluded from the calculation of the statistical data since the respective participants deviated from the stipulated test method by performing the tests with ventilation turned on in the oven. Three of these four laboratories repeated the experiments without ventilation and without knowing the assigned values. Two additional data sets were excluded because the respective laboratories used nonconductive weighing pans. Thus, the statistical data were calculated based on 13 data sets. All of those achieved satisfactory z scores for samples A and C, whereas for sample B 12 results were satisfactory and one was questionable.

All of the four data sets excluded due to ventilation resulted in unsatisfactory z scores for samples B and C. For sample A, three results were rated satisfactory and one was rated questionable. The laboratories that used nonconductive weighing pans achieved satisfactory scores with the exception of sample C where one result was questionable.

The provided method led to comparable and reproducible results and showed robustness against minor disturbances. However, distinct deviations from the stipulated procedure led to flawed results and should be avoided. Based on the results of the current MES, an expanded measurement uncertainty of 25% is recommended for the provided method.

2 Introduction

The method evaluation study (MES) on the determination of volatile compounds in silicone materials was designed both as a study of an adapted test procedure and as a proficiency test. It was organized by the German National Reference Laboratory for Food Contact Materials (NRL-FCM) established within the Unit Product Analytics of the Department of Chemicals and Product Safety at the German Federal Institute for Risk Assessment (BfR).

FCM and other consumer products made of silicone may contain residual volatile oligomeric materials whose post-manufactural release may lead to changes in the properties of the material itself and may affect objects in contact with it [1]. Thus, a post-production treatment is mandatory to remove volatile components from FCM by means of tempering. Such tempered silicone-based consumer products must not release more than 0.5% (w/w) volatile organic compounds [2-4]. The stipulated test procedures are based on a conditioning step of the silicone material followed by tempering for 4 h at 200 °C. The amount of released volatile compounds is determined as mass loss during tempering [5-10].

Previous research indicated that the test conditions might be somewhat ambiguous, preventing sufficient reproducibility and comparability of the obtained results. The German NRL-FCM was asked to identify relevant test parameters and to revise and optimize the method based on previous results and own experiments. Possible crucial parameters that affect the test results are ventilation and air supply in the oven [11], the material of the used weighing pans, type and condition of the desiccant, sample pretreatment, sample handling and storage, as well as environmental conditions including the relative humidity in the weighing chamber.

This MES comprised the gravimetric determination of volatile compounds in three different silicone materials after tempering for 4 h at 200 °C, according to the test method revised by the German NRL-FCM considering the results of previous investigations.

This MES was open to Official Control Laboratories (OCLs) and National Reference Laboratories (NRLs). The 17 laboratories listed in table 1 are kindly acknowledged for their participation in the MES exercise.

Table 1: Participating laboratories

Organization	Country
Bayerisches Landesamt für Gesundheit und Lebensmittelsicherheit (LGL)	Germany
Chemisches und Veterinäruntersuchungsamt Münsterland-Emscher-Lippe (CVUA-MEL)	Germany
Chemisches und Veterinäruntersuchungsamt Ostwestfalen-Lippe (CVUA-OWL)	Germany
Chemisches und Veterinäruntersuchungsamt Stuttgart (CVUA Stuttgart)	Germany
Finish Customs Laboratory	Finland
German Federal Institute for Risk Assessment (BfR)	Germany
Landesamt für Verbraucherschutz Saarland (LAV Saarland)	Germany
Landesbetrieb Hessisches Landeslabor (LHL)	Germany
Landesuntersuchungsamt Rheinland-Pfalz	Germany
Landesuntersuchungsanstalt für das Gesundheits- und Veterinärwesen (LUA) Sachsen	Germany
National Institute of Public Health – Centre of Toxicology and Health Safety (SZU)	Czech Republic
National Laboratory of Health, Environment and Food	Slovenia
Niedersächsisches Amt für Verbraucherschutz und Lebensmittelsicherheit (LAVES)	Germany
Public Analyst's Laboratory Sir Patrick Duns Hospital	Ireland
Service Commun des Laboratoires Laboratoire de Bordeaux-Pessac	France
Technical University Dresden	Germany
Universidade Católica Portuguesa Edifício de Biotecnologia (CINATE)	Portugal

This report summarizes the results of the MES exercise.

3 Scope

As stated in Regulation (EU) 2017/625 [12] one of the core duties of NRLs is to organize inter-laboratory comparative testing or proficiency tests between OCLs. The present MES aims to assess the analytical capabilities of NRLs and OCLs for the determination of volatile compounds in silicone materials, to evaluate a recently revised test procedure and to propose an expanded measurement uncertainty. The participants were asked to condition the provided silicone materials for 1 h at 100 °C, followed by tempering the samples for 4 h at 200 °C and to determine the respective mass losses after this tempering procedure.

This MES is identified as NRL-D-FCM-02/2020.

4 Set up of the exercise

4.1 Time frame of the MES

The MES NRL-DE-FCM-02/2020 was announced on February 10, 2020. Registration was opened till February 21, 2020. Samples were sent to the participants on February 25, 2020, with the exception of LC-017, whose samples were sent on March 27, 2020 due to problems during the registration process. The original deadline was set to April 09, 2020 but was extended due to the COVID-19 lockdown. The last set of results was submitted on July 07, 2020.

4.2 Quality assurance

The German NRL-FCM is accredited according to ISO/IEC 17025 (certificate number: D-PL-18583-02). The reported results were evaluated following the relevant administrative and logistic procedures.

4.3 Confidentiality

The procedures used for the organization of this MES guarantee that the identity of the participants and the information provided by them is treated confidentially. The participants in this MES were assigned with a random unique laboratory code used throughout this report.

4.4 Distribution

Each participant received:

- 50 g each of cut and homogenized silicone elastomers (samples A, B, C)
- 3 stainless steel weighing pans of 8 cm diameter and 2.5 cm height
- NRL_DE_FCM_02_2020_Confirmation of receipt.pdf
- NRL_DE_FCM_02_2020_Instructions.pdf
- NRL_DE_FCM_02_2020_Questionnaire_Results.xlsx

4.5 Instructions to participants

NRL_DE_FCM_02_2020_Instructions.pdf provided detailed instructions to the participants (see 10.1 Instructions).

Participants were asked to check and report whether the test items were undamaged after transport using the NRL_DE_FCM_02_2020_Confirmation of receipt.pdf form.

Participants were asked to determine the relative mass losses of samples A, B, and C after tempering. Approximately 10 g of sample was to be weighed in, conditioned for 60±5 min at 100 °C, followed by at least 30 min cooling in a desiccator. After 240±5 min tempering at 200 °C, the sample was to be cooled at least 60 min in a desiccator. Weighing was to be performed promptly after the cooling periods. All experiments were to be carried out in triplicates, with neither ventilation nor air supply in the oven, and with $CaCl_2$ or silica gel as desiccants. The participants were asked to report the sample mass before and after tempering as well as the relative mass loss after tempering as individual results for each experiment.

Participants received an individual code to report their measurement results and to complete the related questionnaire. The questionnaire was used to gather additional information related to the conducted sample treatment (conditioning and tempering) and the final measurements and laboratories (see 12 Annex - Questionnaire).

The laboratory codes, the questionnaire and the results form were communicated to the participants via e-mail.

5 Test items

5.1 Preparation

Silicone baking molds (samples A and B) and silicone baking mats (sample C) were purchased in local stores in Berlin, Germany, and cut into pieces of approximately 1x2 cm with scissors (samples A and B) and a guillotine trimmer (sample C). For each sample the cut material was combined and thoroughly mixed to yield a bulk material. All samples were stored at room temperature in closed plastic (samples A and B) and cardboard containers (sample C), lined with aluminum foil.

For shipment, approximately 50 g of each sample was taken from the bulk material, weighed in and packed in aluminum foil and a plastic ziplock bag.

5.2 Homogeneity and stability

Investigations and evaluation for the homogeneity and the statistical treatment of data were performed by the German NRL-FCM. The homogeneity assessment of the cut silicone materials was performed with the bulk materials after sample preparation and before distribution to participants. Results were evaluated according to ISO 13528:2015 [13]. All samples were considered to be adequately homogeneous (see 10.2 Homogeneity test results). The stability of the test items was checked by the German NRL-FCM. Repeated tests were conducted with the homogenized bulk materials between August 22, 2019 and June 29, 2020. All samples were considered to be sufficiently stable (see 10.3 Stability tests).

6 Assigned values and standard uncertainties

6.1 Assigned values

No reference values were available for the mass losses of silicone materials. The assigned values x_{pt} for the respective relative mass losses [%] were derived from the results reported by the participants as a robust average (Q/Hampel method [14]; according to ISO 13528 [13] and DIN 38402-45:2014 [15]).

6.2 Standard uncertainties of the assigned values

Because the assigned values were derived as robust averages of the mean results reported by the participants the standard uncertainties $u(x_{pt})$ of the assigned values were estimated according to ISO 13528 (7.7.3) [13]:

$$u(x_{pt}) = 1.25\frac{s^*}{\sqrt{p}} \qquad \text{(Eq.1)}$$

where s^* is the robust standard deviation (Q/Hampel method [14]; according to ISO 13528 [13] and DIN 38402-45:2014 [15]) of the single results reported by the participants and p is the number of values used for the calculation.

In this model, where the assigned value x_{pt} and the robust standard deviation s^* are determined from participants results, the uncertainty $u(x_{pt})$ of the assigned values can be assumed to include the effects of uncertainty due to inhomogeneity, transport, and instability [13].

6.3 Standard deviations for proficiency assessment – preliminary evaluation

Relative standard deviations for proficiency assessment σ_{pt} were derived as robust averages of the mean results reported by the participants according to the Q/Hampel method [13].

An initial assessment of the submitted data revealed that the results from four laboratories (LC-012, LC-013, LC-015, LC-017), especially for samples B and C, were considerably higher than those from the other participants. Further inquiry and discussions with the respective laboratories revealed that they deviated from the stipulated test method by performing the test with ventilation turned on in the oven, either unintentionally, on purpose, or because ventilation could not be turned off. Those four sets of results were excluded from the calculation of the statistical data. Three laboratories repeated the experiment without ventilation and without knowing x_{pt}. The newly reported results (LC-012a, LC-015a, LC-017a) were included in the Q/Hampel calculations to get a final x_{pt} and $u(x_{pt})$.

Two laboratories used weighing pans made of nonconductive material. LC-001 used silica crucibles in addition to the provided stainless steel pans and LC-004 performed the experiment with ceramic pans. Since the provided test method strictly stipulates the use of electroconductive weighing pans, those two set of results were also excluded from the calculation, even though these two sets of data showed no abnormalities. Thus, the parameters x_{pt} and $u(x_{pt})$ were calculated from a total of 13 data sets. However, the reported mean values (x_i) and the resulting z scores (z_i) are also shown for the six excluded data sets.

Table 2 summarizes the relevant parameters needed for scoring, namely, the assigned values (x_{pt}) of relative mass losses after tempering [%], its associated expanded uncertainty ($U(x_{pt})$

calculated with a coverage factor k=2), and the standard deviation for the PT assessment (σ_{pt}) as well as the repeatability standard deviation (s_r).

Table 2: Assigned ranges related to the determination of volatile compounds in silicone materials.

Sample	x_{pt} [%]	$U(x_{pt})$[a)] [%]	σ_{pt}[b)] [%]	σ_{pt} [% of x_{pt}]	$u(x_{pt})/\sigma_{pt}$[c)]	s_r[d)] [%]
A	1.204	±0.052	0.095	7.9	0.270	4.4
B	0.357	±0.030	0.049	13.9	0.301	5.6
C	0.138	±0.015	0.025	17.9	0.299	7.7

x_{pt} and $U(x_{pt})$ values were estimated using mean results reported by the participants (n=13)
a) $U(x_{pt})$ is the expanded uncertainty at a given coverage factor (k=2).
b) reproducibility standard deviation
c) The uncertainty of the standard value may be considered to be negligible if $u(x_{pt})/\sigma_{pt} < 0.3$ [13].
d) repeatability standard deviation

As the proportion $u(x_{pt})/\sigma_{pt}$ was never found to be significantly higher than 0.3 only ***z*** scores were used.

To evaluate the influence of the exclusion of results obtained with nonconductive pans, x_{pt} and $u(x_{pt})$ were also calculated including the data sets of the two laboratories LC-001 and LC-004. The results (table 3) are only shown for comparison and will not be used further in the report.

Table 3: Assigned ranges including LC-001 and LC-004 data sets.

Sample	x_{pt} [%]	$U(x_{pt})$[a)] [%]	σ_{pt}[b)] [%]	σ_{pt} [% of x_{pt}]	$u(x_{pt})/\sigma_{pt}$[c)]	s_r[d)] [%]
A	1.204	±0.048	0.089	7.4	0.268	3.9
B	0.355	±0.024	0.050	14.1	0.245	5.9
C	0.141	±0.014	0.026	18.7	0.264	8.1

x_{pt} and $U(x_{pt})$ values were estimated using mean results reported by the participants (n=15)
a) $U(x_{pt})$ is the expanded uncertainty at a given coverage factor (k=2).
b) reproducibility standard deviation
c) The uncertainty of the standard value may be considered to be negligible if $u(x_{pt})/\sigma_{pt} < 0.3$ [13].
d) repeatability standard deviation

A comparison of these analyses revealed no significant differences between exclusion and inclusion of the results obtained with nonconductive pans.

7 Evaluation of results

7.1 Scores and evaluation criteria

The individual laboratory performance was expressed in terms of *z* scores according to ISO 13528:2015 [13].

The *z* scores for the proficiency test results x_i were calculated as:

$$z_i = \frac{x_i - x_{pt}}{\sigma_{pt}} \qquad \text{(Eq. 2)}$$

where:

x_i	is the mean value, calculated from single values reported by the participant "i",
x_{pt}	is the assigned value,
σ_{pt}	is the standard deviation for proficiency test assessment.

The interpretation of the *z* performance scores is done according to ISO 13528:2015 [13]:

$\lvert z_i \rvert \leq 2.00$	satisfactory performance	(green in chapter 11),
$2.00 < \lvert z_i \rvert < 3.00$	questionable performance	(yellow in chapter 11),
$\lvert z_i \rvert \geq 3.00$	unsatisfactory performance	(red in chapter 11).

The *z* score demonstrates the deviation between the participants' mean and assigned values in terms of the standard deviation for proficiency test assessment (σ_{pt}).

7.2 Laboratory results and scoring

Seventeen laboratories from seven EU member states participated in this MES, including nine OCLs, seven NRLs, and one university. Sixteen of those reported results for mass losses after tempering of the provided samples. One laboratory was not able to report results due to COVID-19 limitations.

The reported results for each participant and each sample in the form of tables and graphs are included in 11 Annex - Mass loss of silicone elastomers after tempering. The laboratory performance for the determination of mass losses after tempering was assessed using *z* scores.

Figure 1 depicts the *z* scores of all 19 data sets (see 6.3 Standard deviations for proficiency assessment – preliminary evaluation). The majority of *z* scores were satisfactory. In case of sample A, only one result was questionable, while four were unsatisfactory and one questionable for each of samples B and C. Without exception, all unsatisfactory results arose from experiments that deviated from the stipulated test method by performing the test with ventilation turned on in the oven. Therefore, in this MES unsatisfactory *z* scores were a clear indication for problems with the ventilation of the ovens used in the labs. On the other hand, experiments that complied with the specified method achieved satisfactory results, with the only exception of one questionable result for sample B.

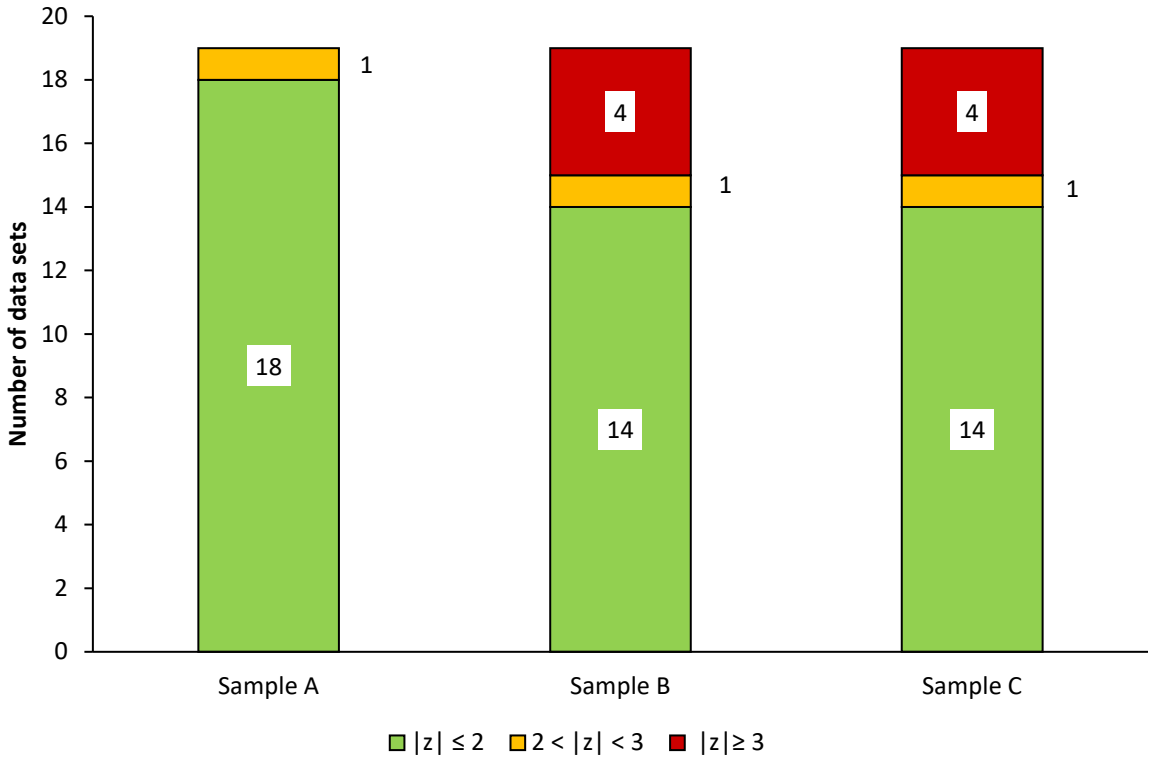

Fig. 1: Overview of laboratory performance according to *z* scores. Corresponding number of data sets are included in the graphs. Satisfactory, questionable and unsatisfactory performances are indicated in green, yellow and red, respectively.

Figure 2 summarizes the results reported for the mass losses of samples A, B, and C after tempering as box plots. Table 4 states the respective values for the 1. and 3. quartiles as well as the interquartile ranges (IQR). Both box plots and interquartile ranges show a narrow distribution of the reported values and indicate that the adapted test method leads to comparable results within this MES.

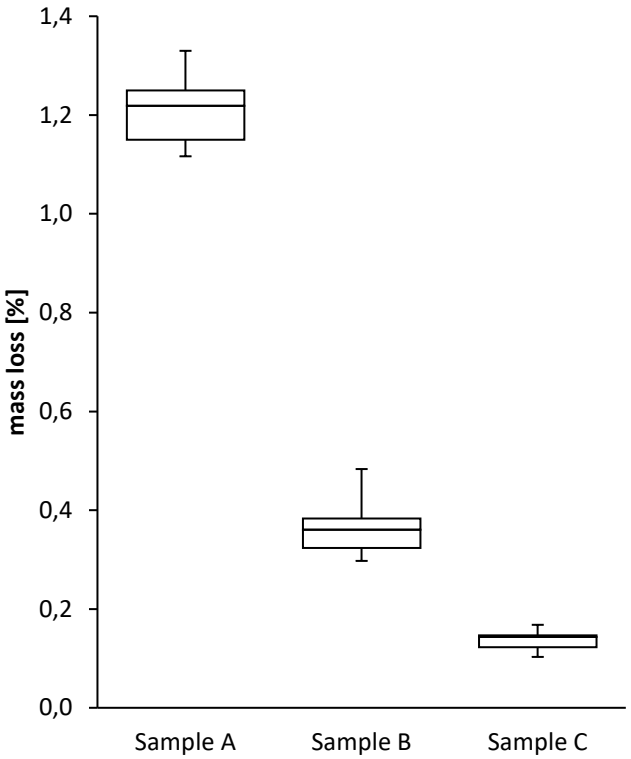

Fig 2: Box plots for the reported values of mass losses of silicone samples A, B, and C after tempering for 4 h at 200 °C (n = 13).

Table 4: Relevant box plot parameters related to the determination of volatile compounds in silicone materials.

Sample	1. Quartile [%]	3. Quartile [%]	IQR[a)] [%]
A	1.150	1.250	0.100
B	0.323	0.383	0.060
C	0.123	0.147	0.024

[a)] Interquartile range

The experimental details, method parameters, and performance characteristics provided by the participants are included in chapter 12 Annex - Questionnaire.

7.3 Additional information extracted from the questionnaire

Most of the participants followed the stipulated method; however, some reported deviations, which have been described in the following text. Chapter 6.3 describes in detail how the deviations from the stipulated method were handled.

Two participants did not use the provided stainless steel weighing pans but their own, which were made of platinum and ceramic, respectively. Two other laboratories used their own pans in addition to the provided ones, namely such made of silica and stainless steel. One laboratory covered samples B and C with a watch glass for weighing to stabilize the weighing process.

Two laboratories used ventilation during the test for temperature stabilization and in two additional cases ventilation was switched on unintentionally.

The test method allowed for several options while performing the experiments and the participants were asked to state which ones they chose. Two laboratories used calcium chloride as desiccant whereas the others used silica gel. Three participants used multiple desiccators, one used a desiccator cabinet and the others used a single desiccator. Four laboratories tested all three samples in a single experiment, whereas the others performed three separate tests. Even though ventilation in the oven was not allowed during the experiment itself, it may be used to speed up the heating-up phase. Four participants used this option.

Additionally, the participants were asked to report information regarding the sample handling including the time required for weighing the samples and transferring them from the desiccator to the oven and vice versa. The time required for weighing varied between 10 and 120 s per sample and between 67 and 960 s for all three samples. The stated transfer time varied between 15 and 180 s for the three replicates of one experiment. The temperature in the ovens before conditioning directly after sample transfer was between 88 and 99 °C for 13 participants. In two cases, the temperature dropped down to 78–79 °C, and one laboratory reported 103 °C. The temperature before tempering after sample transfer was 141 °C for two laboratories, 205 °C for another laboratory, and varied between 174 and 190 °C for the remaining 13 participants.

Three laboratories reported deviations in the duration of the cooling down and storage periods in desiccators. One prolonged the cooling down time from 60 to 90 min after the first sample, one stored the samples after conditioning and weighing in the desiccator for 20 h before tempering and one did not further specify the deviations.

Two participants remarked about the oven temperature. In one case it took about 10 min to reach the original temperature after sample transfer. The other laboratory stated difficulties to keep the stipulated temperature without ventilation.

8 Conclusions

The method evaluation study NRL-D-FCM-02/2020 was organized to determine the robustness of an adapted method for gravimetric determination of volatile compounds in silicone materials and to assess the analytical capabilities of OCLs and NRLs.

All laboratories that followed the stipulated test method achieved satisfactory *z* scores (≤ 2) with the exception of one questionable result for sample B ($2 < |z| < 3$). The distribution of the results was narrow for all three samples. Unsatisfactory high results and *z* scores could be attributed to a deviation from the test method, i.e. ventilation in the oven during tempering. Thus it was possible to identify inaccurately performed tests by means of the reported results.

The two laboratories that used nonconductive weighing pans achieved satisfactory *z* scores with the exception of one questionable result in case of sample C. The statistical parameters were comparable with only minor differences, regardless of whether the respective data sets were excluded from the calculation or not. Even though the use of nonconductive weighing pan material was identified as a source of error in preliminary experiments, it seems to have only a minor influence on the results in this MES.

Some participants reported difficulties to immediately achieve and hold the required temperature in the oven after loading of the samples. However, this had no significant influence on the results. Thus, the method can be assumed to be robust regarding this problem. Presumably, the tempering duration of 4 h is more than sufficient to remove all significant amounts of volatile compounds from the silicone material, so that minor temperature variations at the beginning are negligible.

The assigned values of the mass losses for the samples were 1.204% (A), 0.357% (B), and 0.138% (C). Since the level of interest (LI) for volatile compounds in silicone materials is 0.5%, these values roughly correspond to 2*LI, LI and 0.2*LI, covering the working range of the method [16]. The mean value of all three relative standard deviations is 13%.Taking into ccount the higher inhomogeneity of sample C, this results in an estimated expanded measurement uncertainty of 25% (95% significance, k=1.96).

This method evaluation study revealed that the adapted test method is suitable for the gravimetric determination of volatile compounds in silicone materials. It leads to comparable and reproducible results and is robust against minor disturbances of the process. However, distinct deviations from the test method lead to considerably flawed results and must be avoided.

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10 Annex – General Information

10.1 Instructions

Please follow these instructions as close as possible. In particular, swift handling and weighing of the samples is crucial to achieve comparable results due to fast sorption of water from the laboratory air to the samples. This can hamper settling of the balance and the achievement of stable results. Thus, the lowest value at the beginning of the measurement is to be recorded.

Weighing pans made of materials without or with poor electroconductivity like laboratory or quartz glass are not allowed since they do not discharge static electricity in a reasonable time. Please use the supplied stainless steel weighing pans or pans with comparable properties. Ventilation and/or forced circulation in the drying cabinet can lead to poor reproducibility and is not allowed. However, ventilation may be used during the heat-up phases of the drying cabinet to achieve faster heat-up. All experiments are to be carried out in triplicates. Please check the questionnaire before starting so that you are able to answer all questions!

Please report the sample masses before tempering, the sample masses after tempering, and the relative mass losses after tempering. Please report all individual results. In addition, please report the weighted sample amounts, the weighing room conditions (temperature, relative humidity), the time required for weighing, the time required for sample transfer between drying cabinet and desiccator, and the drying cabinet temperatures directly after sample transfer.

Required equipment

- Balance with a tolerance of ±1 mg or better
- Drying cabinet for temperatures of 100±5 °C and 200±5 °C
- Desiccator with calcium chloride or silica gel with moisture indicator as desiccant

1. Preparation

The weighing pans are to be washed, e.g. in a laboratory dishwasher, heated in the drying cabinet for 4 h at 200 °C, cooled down in the desiccator for at least 1 h and kept there until the beginning of the actual experiment.

2. Weighing

The cleaned and empty weighing pan is weighed in precisely to ±1 mg and the mass determined (M_{empty}). Weigh in approximately 10 g of the sample on the pan. The individual pieces must overlap as little as possible.

3. Conditioning

The weighing pans with samples are kept for 60±5 min in the drying cabinet at 100±5 °C without air circulation and with closed ventilation valve. Let the samples cool down in the desiccator for 30 min.
The weighing pan with the conditioned sample is weighed in precisely to ±1 mg and the mass determined (M_{before}).

4. Tempering

The weighing pans with samples are kept for 240±5 min in the drying cabinet at 200±5 °C without air circulation and with closed ventilation valve. Let the samples cool down in the desiccator for 1 h.

The weighing pan with the tempered sample is weighed in precisely to ±1 mg and the mass determined (M_{after}).

5. Calculation of mass loss

The amount of volatile compounds is calculated according to:

Volatile compounds [%] = $((M_{before}-M_{after})/(M_{before}-M_{empty}))*100$

10.2 Homogeneity test results

The homogeneity assessment of the samples was performed after preparation and mixing and before distribution to the participants. Results were evaluated according to ISO 13528:2015 [13].

The estimate of the between-sample standard deviation s_s often becomes negative when the standard deviation of sample averages $s_{\overline{x}}$ is smaller than the within-sample standard deviation s_w. In this case, the material is considered to be highly homogenous and s_s is set to zero. Since it is not possible to conduct two analyses of the same subsample, 16 single experiments were performed and the results distributed randomly into an 8×2 matrix. This approach is somewhat arbitrary and may not be sufficient to assess the homogeneity of the material. As an alternative for destructive test methods, ISO 13528:2015 allows to set the standard deviation of all results s_r as s_s [13]. Both approaches were used for homogeneity assessment of the samples.

Table 5: Homogeneity study sample A

ISO 13528:2015	Mass loss [%]	
	1	2
1	1.145	1.276
2	1.281	1.232
3	1.156	1.260
4	1.182	1.262
5	1.152	1.183
6	1.216	1.278
7	1.185	1.207
8	1.077	1.197
Mean	1.206	
$s_{\overline{x}}$	0.039	
s_w	0.059	
s_s	0	
$\sigma_{pt}(11\% \text{ of Mean})$	0.133	
σ_{allow}	0.040	
$s_s \leq \sigma_{allow}$	passed	
Assessment	highly homogeneous ($s_s = 0$)	
ISO 13528:2015 alternative	Mass loss [%]	
Mean	1.206	
$s_s = s_r$	0.0577	
$\sigma_{pt}(16\ \% \text{ of Mean})$	0.1929	
σ_{allow}	0.0579	
$s_s \leq \sigma_{allow}$	passed	
Assessment	homogeneous with $\sigma_{pt} = 16\%$	

where:
- $s_{\overline{x}}$ standard deviation of sample averages,
- s_w within-sample standard deviation,
- s_s estimate of between-sample standard deviation,
- s_r repeatability standard deviation,
- σ_{pt} standard deviation for proficiency assessment,
- σ_{allow} $\sigma_{allow} = 0.3\ \sigma_{pt}$; criterion of sufficient homogeneity.

Table 6: Homogeneity study sample B

ISO 13528:2015	Mass loss [%]	
	1	2
1	0.324	0.327
2	0.339	0.339
3	0.323	0.318
4	0.318	0.339
5	0.321	0.329
6	0.351	0.362
7	0.356	0.342
8	0.332	0.365
Mean	0.336	
$s_{\overline{x}}$	0.014	
s_w	0.0112	
s_s	0.0110	
$\sigma_{pt}(11\% \text{ of Mean})$	0.0370	
σ_{allow}	0.0111	
$s_s \leq \sigma_{allow}$	passed	
Assessment	homogeneous with $\sigma_{pt} = 11\%$	
ISO 13528:2015 alternative	Mass loss [%]	
Mean	0.336	
$s_s = s_r$	0.0154	
$\sigma_{pt}(16\ \% \text{ of Mean})$	0.0538	
σ_{allow}	0.0161	
$s_s \leq \sigma_{allow}$	passed	
Assessment	homogeneous with $\sigma_{pt} = 16\%$	

Table 7: Homogeneity study sample C

ISO 13528:2015	Mass loss [%]	
	1	2
1	0.134	0.130
2	0.146	0.126
3	0.137	0.126
4	0.150	0.128
5	0.138	0.117
6	0.131	0.140
7	0.109	0.134
8	0.127	0.134
Mean	0.132	
$s_{\overline{x}}$	0.005	
s_w	0.012	
s_s	0	
$\sigma_{pt}(11\% \text{ of Mean})$	0.014	
σ_{allow}	0.004	
$s_s \leq \sigma_{allow}$	passed	
Assessment	highly homogeneous ($s_s = 0$)	
ISO 13528:2015 alternative	Mass loss [%]	
Mean	0.132	
$s_s = s_r$	0.0102	
$\sigma_{pt}(26\ \% \text{ of Mean})$	0.0342	
σ_{allow}	0.0103	
$s_s \leq \sigma_{allow}$	passed	
Assessment	homogeneous with $\sigma_{pt} = 26\%$	

Where:
- $s_{\overline{x}}$ standard deviation of sample averages,
- s_w within-sample standard deviation,
- s_s estimate of between-sample standard deviation,
- s_r repeatability standard deviation,
- σ_{pt} standard deviation for proficiency assessment,
- σ_{allow} $\sigma_{allow} = 0.3\ \sigma_{pt}$; criterion of sufficient homogeneity.

10.3 Stability tests

The stability of the test items was monitored over a period of about eight months for each sample with repeated tests. The amount of volatile compounds was determined as mass loss after tempering for 4 h at 200 °C, following conditioning at 100 °C for 1 h. The mean values and relative standard deviations of the respective experiments are stated in table 8.

There was no observable trend in the determined mass losses over time. According to ISO 13528:2015, the test item can be considered to be stable if the difference in the measurements before and after the proficiency test is smaller than or equals to 0.3 times the standard deviation for proficiency assessment σ_{pt}[13]. Additionally, the uncertainty due to homogeneity of the samples was taken into account on the basis of ISO 13528:2015 [13] The test items were considered to be sufficiently stable.

Table 8: Stability test results related to the determination of volatile compounds in silicone materials

Sample	mean [%]	RSD [%]	$n^{a)}$ [-]	period [d]	slope[b)] [%/d]	Δ during MES[c)] [%]	stability criterion[d)]
A	1.217	5.8	20	256	$-5.3*10^{-4}$	0.0705	0.0862
B	0.369	10.3	6	265	$-3.4*10^{-5}$	0.0045	0.0301
C	0.143	12.4	6	230	$9.9*10^{-5}$	0.0132	0.0177

[a)] number of experiments
[b)] slope of the regression line for the complete test period
[c)] difference of values during duration of MES (133 days between shipment of samples on February 25, 2020 and receipt of the last set of results on July 7, 2020)
[d)] calculated with the standard deviation σ_{pt} from the MES and the uncertainty of the homogeneity tests with $0.3\sigma_{pt} + s_r(homogeneity)$

11 Annex – Mass loss of silicone elastomers after tempering

11.1 Sample A

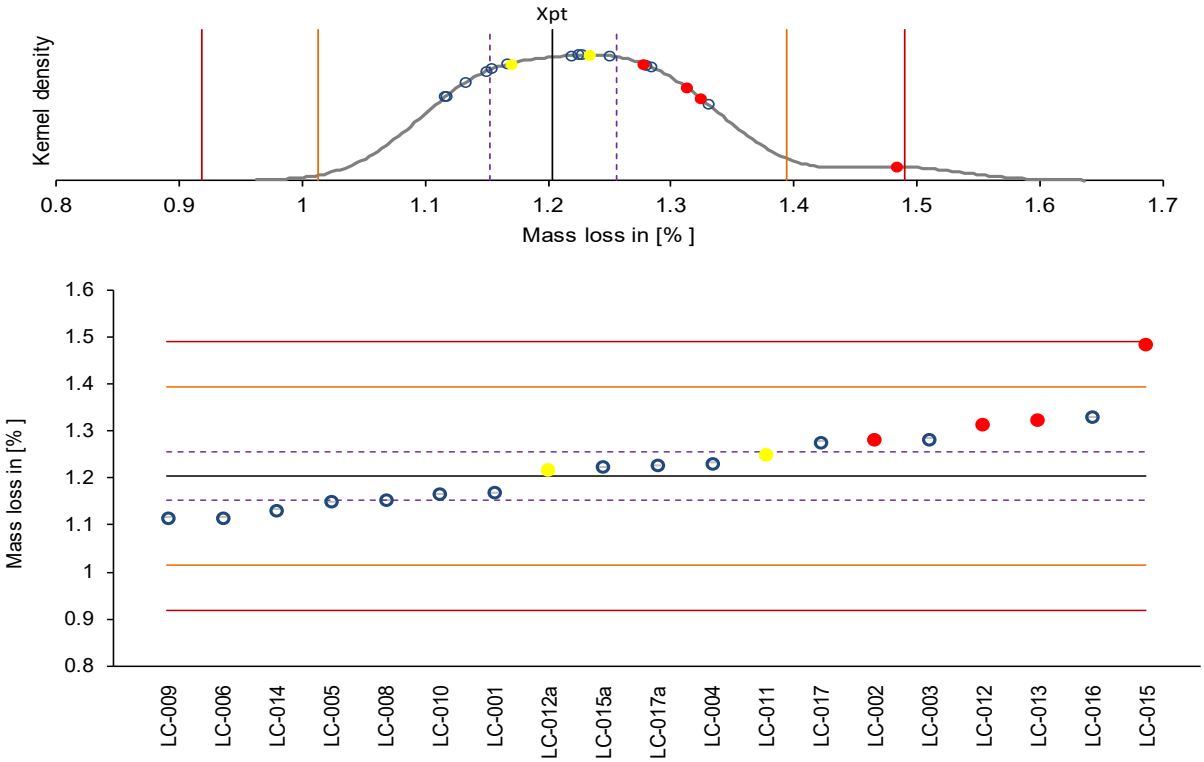

Fig 3: Measurement result range reported by the participants for the mass loss of sample A after tempering. Orange lines represent a *z* score of 2 ($x_{pt} \pm 2\ \sigma_{pt}$), red lines a z score of 3 ($x_{pt} \pm 3\ \sigma_{pt}$), the black line the assigned value x_{pt}. The dashed purple line represents the expanded uncertainty of the assigned value ($x_{pt} \pm U(x_{pt})$) and the circles the individual results x_i. Filled circles indicate the results that were excluded from the calculation of statistical values due to deviations from the stipulated test method (red: ventilation turned on; yellow: use of nonconductive weighing pans).

Table 9: Results for the mass loss of sample A after tempering. Assigned range: x_{pt} = 1.204%; σ_{pt} = 0.095%. Results of repeated experiments from laboratories that did not follow the stipulated method in the first place are stated at the end of the table and marked with the suffix "a"

Laboratory Code	x_i [%]	*z* score
LC-001	1.170	-0.35
LC-002	1.280	0.80
LC-003	1.283	0.83
LC-004	1.232	0.30
LC-005	1.150	-0.56
LC-006	1.117	-0.91
LC-008	1.154	-0.52
LC-009	1.116	-0.92
LC-010	1.166	-0.40
LC-011	1.250	0.48
LC-012	1.313	1.14
LC-013	1.323	1.25
LC-014	1.133	-0.74
LC-015	1.483	2.93
LC-016	1.330	1.32
LC-017	1.277	0.77
LC-012a	1.219	0.15
LC-015a	1.224	0.21
LC-017a	1.227	0.24

11.2 Sample B

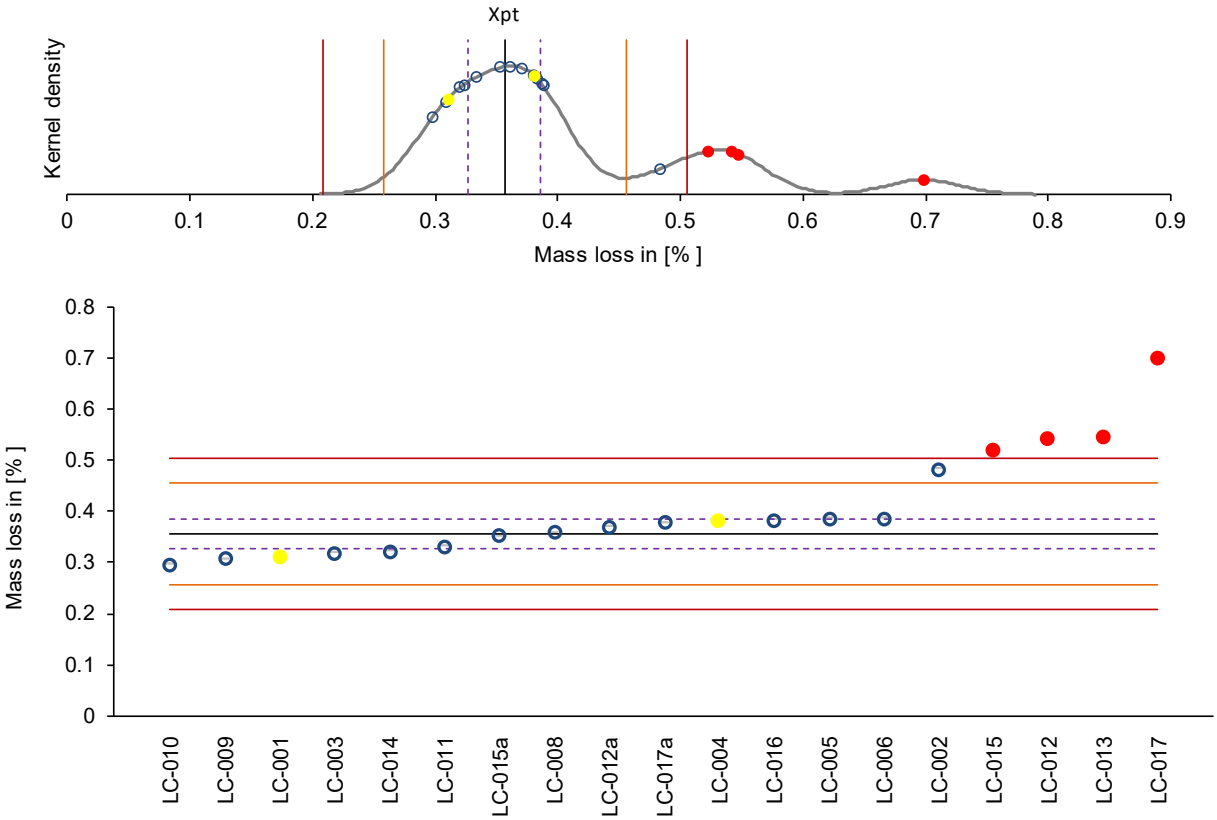

Fig 4: Measurement result range reported by the participants for the mass loss of sample B after tempering. Orange lines represent a *z* score of 2 ($x_{pt} \pm 2\ \sigma_{pt}$), red lines a z score of 3 ($x_{pt} \pm 3\ \sigma_{pt}$), the black line the assigned value x_{pt}. The dashed purple line represents the expanded uncertainty of the assigned value ($x_{pt} \pm U(x_{pt})$) and the circles the individual results x_i. Filled circles indicate the results that were excluded from the calculation of statistical values due to deviations from the stipulated test method (red: ventilation turned on; yellow: use of nonconductive weighing pans).

Table 10: Results for the mass loss of sample B after tempering. Assigned range: x_{pt} = 0.357%; σ_{pt} = 0.049%. Results of repeated experiments from laboratories that did not follow the stipulated method in the first place are stated at the end of the table and marked with the suffix "a"

Laboratory Code	x_i [%]	*z* score
LC-001	0.310	-0.94
LC-002	0.483	2.56
LC-003	0.320	-0.74
LC-004	0.381	0.49
LC-005	0.387	0.61
LC-006	0.388	0.63
LC-008	0.361	0.08
LC-009	0.308	-0.98
LC-010	0.297	-1.20
LC-011	0.333	-0.47
LC-012	0.541	3.73
LC-013	0.547	3.84
LC-014	0.323	-0.67
LC-015	0.521	3.33
LC-016	0.383	0.54
LC-017	0.698	6.90
LC-012a	0.371	0.28
LC-015a	0.353	-0.07
LC-017a	0.380	0.47

11.3 Sample C

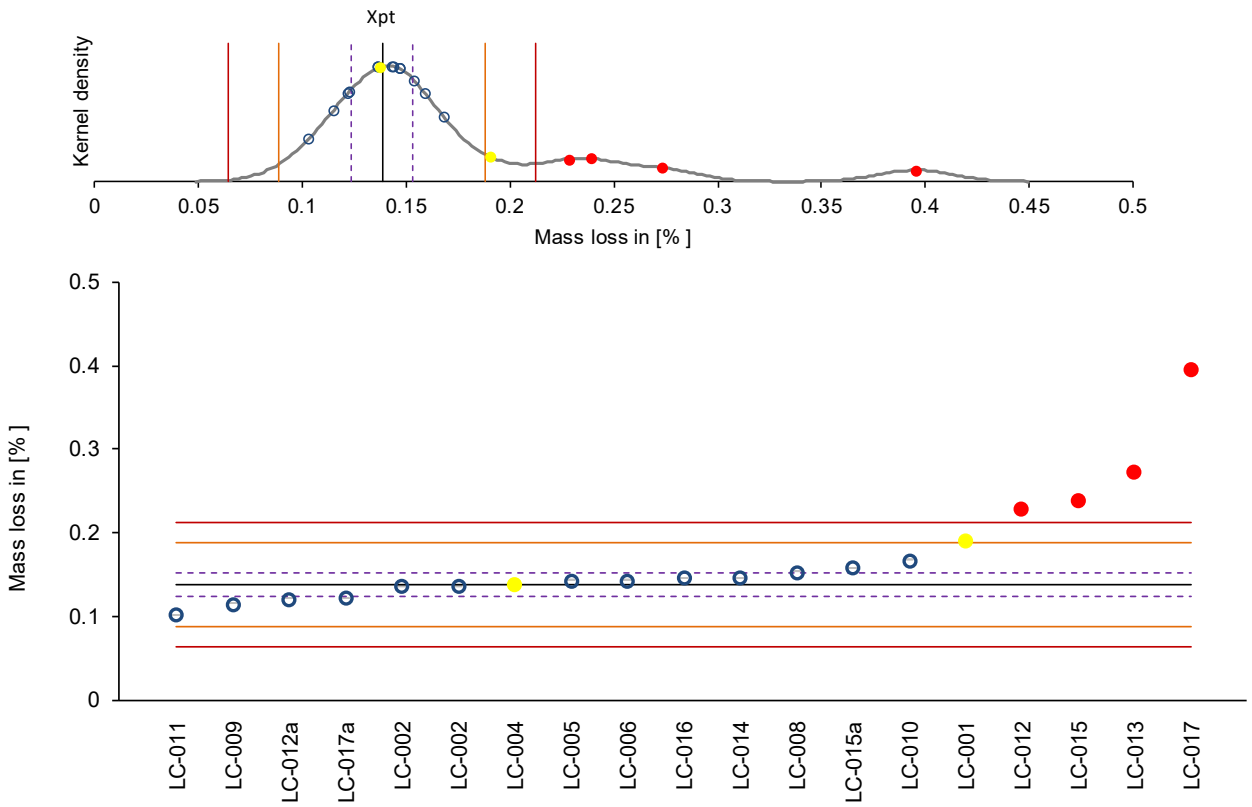

Fig 5: Measurement result range reported by the participants for the mass loss of sample C after tempering. Orange lines represent a *z* score of 2 ($x_{pt} \pm 2\ \sigma_{pt}$), red lines a z score of 3 ($x_{pt} \pm 3\ \sigma_{pt}$), the black line the assigned value x_{pt}. The dashed purple line represents the expanded uncertainty of the assigned value ($x_{pt} \pm U(x_{pt})$) and the circles the individual results x_i. Filled circles indicate the results that were excluded from the calculation of statistical values due to deviations from the stipulated test method (red: ventilation turned on; yellow: use of nonconductive weighing pans).

Table 11: Results for the mass loss of sample C after tempering. Assigned range: x_{pt} = 0.138%; σ_{pt} = 0.025%. Results of repeated experiments from laboratories that did not follow the stipulated method in the first place are stated at the end of the table and marked with the suffix "a"

Laboratory Code	x_i [%]	*z* score
LC-001	0.190	2.09
LC-002	0.137	-0.06
LC-003	0.137	-0.06
LC-004	0.137	-0.04
LC-005	0.143	0.21
LC-006	0.144	0.23
LC-008	0.154	0.63
LC-009	0.115	-0.94
LC-010	0.168	1.20
LC-011	0.103	-1.43
LC-012	0.228	3.62
LC-013	0.273	5.46
LC-014	0.147	0.34
LC-015	0.239	4.07
LC-016	0.147	0.34
LC-017	0.395	10.40
LC-012a	0.122	-0.67
LC-015a	0.159	0.85
LC-017a	0.123	-0.63

12 Annex – Questionnaire

12.1 General

Table 12: General Information

Laboratory code	Does your laboratory have a quality management system?	if Yes, please specify	Do you provide an uncertainty statement to your customer?
001	Yes	ISO 17025	Yes
002	Yes	ISO 17025	Yes
003	Yes	ISO 17025	Yes
004	Yes	ISO 17025	Sometimes
005	Yes	ISO 17025	Yes
006	Yes	ISO 17025	Sometimes
008	Yes	ISO 17025	Sometimes
009	Yes	ISO 17025	Yes
010	Yes	ISO 17025	Sometimes
011	Yes	ISO 17025	Sometimes
012	Yes	ISO 17025	Yes
013	Yes	ISO 17025	Sometimes
014	Yes	ISO 17025	Yes
015	Yes	ISO 17025	Yes
016	Yes	ISO 17025	Sometimes
017	Yes	ISO 17025	Sometimes

Table 13: Analytical Method (Part 1)

Laboratory code	Please state the reading accuracy of the balance used for the experiments [mg]	Please state the error of measurement/repeatability of the balance [mg]	What type of desiccator and how many did you use for this experiment?	What kind of desiccant did you use?
001	0.10	0.22	Single desiccator	Silica gel
002	0.10	0.10	Single desiccator	Silica gel
003	0.10	0.05	Multiple desiccators	Silica gel
004	0.10	0.44	Single desiccator	Silica gel
005	0.01	0.10	Single desiccator	Silica gel
006	0.10	0.10	Single desiccator	Silica gel
008	0.10	0.001	Single desiccator	Silica gel
009	0.01	0.007	Desiccator cabinet	Calcium chloride
010	0.01	0.10	Single desiccator	Silica gel
011	0.10	0.06	Single desiccator	Silica gel
012	0.01	0.001	Single desiccator	Calcium chloride
013	0.1	1	Single desiccator	Silica gel
014	0.10	0.10	Multiple desiccators	Silica gel
015	0.10	2.00	Single desiccator (Multiple desiccators for results 15a)	Silica gel
016	1.00	3.00	Multiple desiccators	Silica gel
017	1.00	0.05	Single desiccator	Silica gel

Table 14: Analytical Method (Part 2)

Laboratory code	Is your oven equipped with air circulation and/or a ventilation valve?	Did you use ventilation in the oven for heating-up?	How did you monitor the temperature in the oven?
001	Ventilation valve	No	Temperature display
002	Both	No	Temperature display
003	Neither	No	Internal temperature logging
004	Both	No	Temperature display
005	Air circulation	Yes	Temperature sensor
006	Ventilation valve	No	Temperature display
008	Both	No	Temperature sensor
009	Both	Yes	Internal temperature logging
010	Neither	No	Temperature display
011	Both	Yes	Temperature display
012	Ventilation valve	No	Temperature display
013	Air circulation	No	Temperature sensor
014	Both	No	Thermometer
015	Both	Yes	Thermometer
016	Neither	No	Thermometer
017	Both	No	Temperature sensor

Table 15: Analytical Method (Part 3)

Laboratory code	Did you use the provided weighing pans?	if No, please describe your weighing pan material	if No, please state your weighing pan diameter [mm]	if No, please state your weighing pan height [mm]	Did you perform 3 separate experiments or did you test all 3 materials in a single experiment?
001	Yes	Silica	150	40	Single experiment
002	Yes				3 Separate experiments
003	Yes				3 Separate experiments
004	No	Ceramic	60	20	Single experiment
005	Yes				3 Separate experiments
006	Yes				3 Separate experiments
008	Yes				3 Separate experiments
009	Yes				3 Separate experiments
010	Yes				3 Separate experiments
011	Yes				3 Separate experiments
012	No	platin	75	27	Single experiment
013	Yes				3 Separate experiments
014	Yes				3 Separate experiments
015	Yes	additional use of own pans: metallic for results 15a			3 Separate experiments
016	Yes	additional use of own pans: stainless steel	100	30	Single experiment
017	Yes				3 Separate experiments

Table 16: Sample handling/accompanying parameters (Part 1)

Laboratory code	Please state the time required for weighing the samples [s]	Please state the time required for sample transfer oven<>exsiccator [s]	Oven temperature before conditioning directly after sample transfer [°C]	Oven temperature before tempering directly after sample transfer [°C]
001	960	30	103	205
002	16	19	96	190
003	mean: 68 sec	mean: 9 sec	A: 92,9°C; B:89,0°C; C: 79,1°C	A: 195,2°C; B: 170,2°C: C: 141,0
004	10 each replicate	15 all the pans	99	189
005	16	12	88 to 94	178 to 182
006	18 s per sample	20 s for 3 samples	95.00	179
008	for 3 samples: about 150	for 3 samples: about 40	for 3 samples: about 88	for 3 samples: about 174
009	67-103 for 3 samples	12-16 for 3 samples	96; 96; 96	187; 188; 185
010	20	14	93	186
011	15	5	92	190
012	20-60	20	98	177
013	30	60	95	175
014	26	9	98	189
015	162	14	78	141.7
016	120 sec per sample on average	180 sec for all samples	95.7	190.0
017	60	30	97.9	174.5

Table 17: Sample handling/accompanying parameters (Part 2)

Laboratory code	Weighted sample [g]								
	A1	A2	A3	B1	B2	B3	C1	C2	C3
001	9.993	10.007	10.005	9.998	10.001	9.997	10.005	10.017	9.994
002	10.110	10.017	10.109	10.056	10.454	10.236	10.109	10.072	10.147
003	10.158	10.131	10.097	10.084	9.978	10.210	10.103	10.194	10.131
004	10.076	10.021	10.089	10.074	10.015	10.033	10.027	10.066	10.045
005	not recorded	not recorded	not recorded	not recorded	not recorded	not recorded	not recorded	not recorded	not recorded
006	10.148	9.961	9.962	10.135	10.143	9.998	10.111	10.190	10.108
008	10.059	10.102	10.094	10.123	10.080	10.017	10.099	10.027	10.052
009	10.001	10.030	10.004	10.005	10.168	10.114	10.022	10.126	10.158
010	10.017	10.041	10.271	10.103	10.141	10.154	10.196	10.201	10.186
011	10.237	10.195	10.004	10.035	10.059	10.043	10.042	10.006	10.081
012	10.057	10.586	10.547	10.320	10.135	10.585	10.104	10.063	10.046
013	10.107	10.060	10.077	10.078	10.076	10.073	10.048	10.120	10.197
014	10.044	9.906	9.935	10.074	9.960	9.976	10.001	10.005	10.043
015	10.004	10.041	10.084	10.432	9.957	10.024	10.183	10.116	10.035
016	10.490	10.266	10.161	10.154	10.175	10.108	10.176	10.166	10.256
017	10.038	9.868	9.986	10.285	9.85	9.979	10.097	9.849	9.772

Table 18: Additional Information

Laboratory code	Did you apply any treatment to the delivered samples other than those specified in the instructions?	if Yes, please specify	Did you encounter any problems with the analysis?	if Yes, please specify
001	No		No	I used both the provided stainless steel pans and our in-house silica crucibles
002	No		No	
003	No		Yes	The weighing pans should have a larger diameter than the delivered pans, so the sample pieces overlap could be reduced.
004	No		Yes	10 g of sample C were too much for the pan size
005	No		No	However, it was difficult to keep the temperature in the oven in the required interval 100°C±5°C and 200°C±5°C because no air circulation was used.
006	No		No	size and shape of samples varied quiet a lot after first sample we prolonged the cool down time from 60 min to 90 min => it seemed the weight was more stable
008	Yes	after conditioning, cooling down and weighing, the samples were stored for about 20 hours in the exsiccator and then continued tempering	No	but you have to be quick with weighing; the relative humidity in the weighing room was about 30% during the first experiment;
009	No		No	
010	Yes	For the samples B and C we additionally took watch glasses to cover the samples during weighing in order to stabilize the weighing process. The watch glasses were also treated like the weighing pans (preparation, conditioning and tempering). Before each weighing we performed a deionization. After sample transfer into the oven it took 10 minutes until the original temperature was reached again.	Yes	During the weighing process with sample we noticed intense deviations.
011	No		No	
012	No		No	The material was cut rather coarsely compared to our usual approach (we cut it to 1x1 cm)
013	No		No	
014	No		Yes	Conditioning at 100 °C is very time-consuming and not practicable in a routine laboratory
015	Yes	Time in exsiccator.	Yes	We have to use very mild ventilation in the oven to stabilize the temperature.
016	No		No	
017	No		No	

12.2 Results

Table 19: Measured masses and calculated mass loss of sample A, measurements 1 and 2

Laboratory code	Measurement 1			Measurement 2		
	Sample mass before tempering [g]	Sample mass after tempering [g]	Mass loss after tempering [%]	Sample mass before tempering [g]	Sample mass after tempering [g]	Mass loss after tempering [%]
LC-001	9.8999	9.7818	1.19	9.9016	9.7872	1.16
LC-002	10.032	9.9053	1.26	9.9326	9.8019	1.32
LC-003	10.0676	9.9334	1.33	10.0405	9.916	1.24
LC-004	9.9727	9.8519	1.2113	9.9226	9.8016	1.2194
LC-005	10.39261	10.2755	1.13	10.35451	10.23412	1.16
LC-006	10.1482	10.032	1.145	9.9612	9.8487	1.129
LC-008	9.9597	9.8359	1.243	9.9901	9.8756	1.1461
LC-009	9.92787	9.81751	1.111618101	9.95386	9.84284	1.115346207
LC-010	9.92141	9.81443	1.0783	9.94632	9.82652	1.2045
LC-011	10.1456	10.0151	1.28	10.1109	9.9929	1.17
LC-012	9.97288	9.84194	1.31296075	10.4864	10.34304	1.367104059
LC-012a	10.49363	10.36129	1.261146048	10.05891	9.93691	1.212855071
LC-013	9.9764	9.8516	1.25	9.9336	9.7962	1.38
LC-014	10.0436	9.9281	1.15	9.9064	9.7912	1.16
LC-015	9.9181	9.7698	1.495246065	9.9496	9.8064	1.439253839
LC-015a	10.3955	10.2784	1.1264	10.0807	9.9483	1.3134
LC-016	10.388	10.251	1.32	10.172	10.035	1.35
LC-017	9.94197	9.80957	1.332	9.78159	9.66042	1.239
LC-017a	10.0841	9.95684	1.262	9.86731	9.7474	1.215

Table 20: Measured masses and calculated mass loss of sample A, measurement 3 and accompanying parameters

Laboratory code	Measurement 3			Temperature [°C]	relative humidity [%]
	Sample mass before tempering [g]	Sample mass after tempering [g]	Mass loss after tempering [%]		
LC-001	9.9018	9.7869	1.16	21.3	42.3
LC-002	10.0367	9.9099	1.26	25.8	31
LC-003	10.0044	9.876	1.28	22.6	30.6
LC-004	9.9813	9.8549	1.2664	22	60
LC-005	10.44549	10.32448	1.16	23.9	35
LC-006	9.9623	9.855	1.077	22.5	48.1
LC-008	10.0014	9.894	1.0738	about 21	about 22
LC-009	9.93325	9.82175	1.122492638	22.1/22.1/22.2	34.8/31.2/30.4
LC-010	10.16853	10.04496	1.2152	23.1	41
LC-011	9.9184	9.7899	1.3	23	35
LC-012	10.45387	10.32238	1.2578117	22	39.5 (at weighing in) / 43.3 (after conditioning) / 50.9 (after tempering)
LC-012a	10.39037	10.26758	1.181767348	between 20.5 and 23.7	between 43.9 and 55.2
LC-013	9.9479	9.8146	1.34	not reported	not reported
LC-014	9.9347	9.8268	1.09	21.6	19
LC-015	10.003	9.8515	1.514545636	22.4	23.9
LC-015a	10.04	9.9163	1.2321	23.2	48.4
LC-016	10.062	9.929	1.32	22	30 - 40
LC-017	9.8907	9.76593	1.261	21.5	22
LC-017a	9.91101	9.79164	1.204	21.5	22

Table 21: Measured masses and calculated mass loss of sample B, measurements 1 and 2

Laboratory code	Measurement 1			Measurement 2		
	Sample mass before tempering [g]	Sample mass after tempering [g]	Mass loss after tempering [%]	Sample mass before tempering [g]	Sample mass after tempering [g]	Mass loss after tempering [%]
LC-001	9.9498	9.9206	0.29	9.947	9.9184	0.29
LC-002	10.0212	9.9713	0.5	10.4185	10.3708	0.46
LC-003	10.0038	9.9736	0.3	9.8804	9.8442	0.37
LC-004	10.0146	9.9722	0.4234	9.9589	9.9209	0.3816
LC-005	1.033329	10.29168	0.4	10.38178	10.34261	0.38
LC-006	10.1352	10.0966	0.381	10.1434	10.1037	0.391
LC-008	10.0735	10.0342	0.3901	10.0319	9.9949	0.3688
LC-009	9.97662	9.94473	0.319647335	10.13818	10.10831	0.294628819
LC-010	10.04918	10.02012	0.2892	10.08634	10.05634	0.2974
LC-011	9.9875	9.9551	0.32	10.0104	9.9768	0.34
LC-012	10.26915	10.21526	0.524775663	10.08707	10.03574	0.508869275
LC-012a	10.02492	9.98221	0.426038313	10.32772	10.29263	0.339765214
LC-013	10.0206	9.9614	0.59	10.0207	9.9706	0.5
LC-014	10.0735	10.0392	0.34	9.9595	9.9298	0.3
LC-015	10.3835	10.3321	0.495016131	9.9111	9.8595	0.520628386
LC-015a	10.24323	10.2078	0.3456	10.0276	9.9929	0.346
LC-016	10.108	10.069	0.39	10.127	10.091	0.36
LC-017	10.23164	10.16067	0.694	9.80382	9.73533	0.699
LC-017a	10.15191	10.11326	0.381	9.95886	9.92223	0.368

Table 22: Measured masses and calculated mass loss of sample B, measurement 3 and accompanying parameters

Laboratory code	Measurement 3			Temperature [°C]	relative humidity [%]
	Sample mass before tempering [g]	Sample mass after tempering [g]	Mass loss after tempering [%]		
LC-001	9.9533	9.9183	0.35	21.3	42.3
LC-002	10.2018	10.1517	0.49	25.9	31
LC-003	10.1581	10.1286	0.29	22.9	10.7
LC-004	9.9761	9.9425	0.3368	22	60
LC-005	10.248	10.20921	0.38	23.9	37
LC-006	9.9983	9.9591	0.392	22.6	48.5
LC-008	9.9695	9.9373	0.323	about 21	about 22
LC-009	10.08596	10.05467	0.310233235	21.7/22.0/21.9	12.3/10.7/8.9
LC-010	10.10078	10.06992	0.3055	23.3	31
LC-011	9.9946	9.9605	0.34	22	38
LC-012	10.53645	10.47431	0.589762206	22.6	48.0 (at weighing in) / 50.3 (after conditioning) / 42.2 (after tempering)
LC-012a	10.41741	10.38137	0.345959312	between 20.5 and 23.7	between 43.9 and 55,2
LC-013	10.0181	9.9631	0.55	not reported	not reported
LC-014	9.976	9.9428	0.33	21.7	19
LC-015	9.9774	9.9227	0.54823902	22.3	20.3
LC-015a	10.1079	10.0707	0.368	23.3	48.2
LC-016	10.061	10.021	0.4	22	30 - 40
LC-017	9.92854	9.85885	0.702	21.5	22
LC-017a	10.18736	10.14756	0.391	21.5	22

Table 23: Measured masses and calculated mass loss of sample C, measurements 1 and 2

Laboratory code	Measurement 1			Measurement 2		
	Sample mass before tempering [g]	Sample mass after tempering [g]	Mass loss after tempering [%]	Sample mass before tempering [g]	Sample mass after tempering [g]	Mass loss after tempering [%]
LC-001	10	9.981	0.19	10.0109	9.9939	0.17
LC-002	10.106	10.0915	0.14	10.069	10.0561	0.13
LC-003	10.0546	10.0443	0.1	10.2	10.187	0.13
LC-004	10.0105	9.998	0.1249	10.0514	10.037	0.1433
LC-005	10.26392	10.24941	0.14	10.28299	10.26747	0.15
LC-006	10.1107	10.0961	0.144	10.1898	10.175	0.145
LC-008	10.0947	10.0784	0.1615	10.0238	10.0082	0.1556
LC-009	10.01889	10.00866	0.10210712	10.12324	10.11171	0.113896341
LC-010	10.18836	10.17361	0.1448	10.19548	10.17689	0.1823
LC-011	10.0385	10.0287	0.098	10.0013	9.9913	0.1
LC-012	10.09787	10.07319	0.244407979	10.0573	10.03367	0.234953715
LC-012a	10.35112	10.33792	0.127522432	10.12559	10.11483	0.106265413
LC-013	10.0415	10.0137	0.28	10.1136	10.0906	0.23
LC-014	10.0007	9.986	0.15	10.0053	9.9906	0.15
LC-015	10.1781	10.1486	0.289837985	10.1117	10.0883	0.231415093
LC-015a	10.0647	10.0487	0.159	10.0515	10.0349	0.1651
LC-016	10.173	10.155	0.18	10.167	10.152	0.15
LC-017	10.09216	10.05465	0.372	9.44017	9.39798	0.447
LC-017a	10.07302	10.06086	0.121	10.1466	10.13481	0.116

Table 24: Measured masses and calculated mass loss of sample C, measurement 3 and accompanying parameters

Laboratory code	Measurement 3			Temperature [°C]	relative humidity [%]
	Sample mass before tempering [g]	Sample mass after tempering [g]	Mass loss after tempering [%]		
LC-001	9.9847	9.9634	0.21	21.3	42.3
LC-002	10.1439	10.1302	0.14	24.5	32
LC-003	10.1348	10.1163	0.18	23.2	11.9
LC-004	10.033	10.0186	0.1435	22	60
LC-005	10.31978	10.30551	0.14	23.7	33.7
LC-006	10.108	10.0936	0.143	22.5	48
LC-008	10.0493	10.0348	0.1443	about 21	about 22
LC-009	10.15503	10.1419	0.129295531	21.8/22.0/22.1	11.4/12.0/12.3
LC-010	10.18052	10.16251	0.1769	25.1	34
LC-011	10.0765	10.0653	0.111	23	37
LC-012	10.0397	10.0192	0.204189368	22.9	33.8 (at weighing in) / 29.6 (after conditioning) / 21.4 (after tempering)
LC-012a	10.29684	10.28336	0.13091395	between 20.5 and 23.7	between 43.9 and 55.2
LC-013	10.1902	10.1587	0.31	not reported	not reported
LC-014	10.0426	10.0286	0.14	21.8	19
LC-015	10.031	10.0114	0.195394278	22.9	26.8
LC-015a	10.2269	10.2112	0.1535	23.4	47.8
LC-016	10.25	10.239	0.11	22	30 - 40
LC-017	10.17175	10.17175	0.367	21.5	22
LC-017a	10.13845	10.12512	0.131	21.5	22

13 Figures

14 Tables