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A New Way to Improve Thermal Capacities of Lubricants for the Manufacture of Flint Glass Perfume Bottles: Part A—How to Combine Thermal Analysis and Physico-Chemical Observations at the Glass/Punch Interface

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ABSTRACT

In the hollow glass industry, the success of the forming process depends on controlling the thermal exchange at the glass/mold interface to prevent defects on the glass surface. In the manufacturing process for luxury perfume bottles, the current practice is to deposit a resin film on the inner faces of the mold at the beginning of the production process and regularly swab the mold with a lubricating paste. This study presents a new way to analyze the impact of lubrication on glass/tool thermal exchanges. The TEMPO Laboratory (Valenciennes, France) has an experimental Glass/Tool Interaction (GTI) platform, which is a reduced-scale production unit that allows researchers to reproduce the pressing cycle conditions encountered in the glass industry. To complete the analysis of the thermal exchange at the glass/tool interface, the BCR Center (Mons, Belgium) took physico-chemical measurements on the produced glass samples after the trials on the GTI platform. Part A presents the experimental conditions on the GTI platform and the thermal analysis with this platform for the first case of flint glass pressing cycles with a punch swabbed with a lubricating paste developed by our partner, SOGELUB® Special Lubricants Company (Marquain, Belgium). The analysis of the physico-chemical changes on the pressed glass samples produced with the swabbed punch were completed with our observations using a Scanning Electron Microscope (SEM) with Energy Dispersive Spectroscopy (EDS).

Keywords*: *Glass Forming, Lubrication, Pressing, Heat Transfer, Glass/Tool Contact, Physico-Chemical Analysis

1. Introduction

In hollow glass industry, the thermal exchanges at the glass/tool interface during the forming process are essential for the final quality of the glass products. Inappropriate thermal conditions lead to the formation of defects on the glass surface, the adhesion/sticking of the glass to the forming tools and the rapid damage of these tools. The heat exchange between the glass and the tools depends mainly on the initial temperatures of glass and tools, the materials composing the tools, the tool's roughness and the contact pressure between the glass and the tools. The presence of lubrication between the glass and the tools also contributes to the success of the forming process. By preventing the direct glass/metal contact, the lubrication limits the heat exchange between the glass and the tool, thus making it possible to not reach critical temperatures that lead to sticking.

For studies of the glass/tool contact, experimental platforms have been designed to better understand the heat exchange during the glass/tool contact [1-3] or to analyze the adhesion [4-6] and sticking [5-9] events between the glass and the tool. The main goal is to reproduce the industrial forming conditions for the glass container production [1-4,7,9] or for the precision molding of optical components [5,6,8]. These are high temperature experimental platforms in which the glass (e.g., soda lime silica glass, borosilicate, lead crystal, flint glass, coloured glass) is heated to a temperature between 900˚C and 1200˚C. The metal sample (e.g., cast iron, bronze, stainless steel) standing for the tool has an initial temperature between 450˚C and 750˚C.

All these platforms are instrumented with thermocouples in the metal sample near to the contact surface. Höhne *et al.* [2] also placed thermocouples in the glass. Depending on the platform, infrared pyrometers are used to measure the glass temperature before contact, effort sensors are used to control the contact force or to record the sticking force, and/or rapid cameras are used to film the process. In terms of the authors' objectives, the contact between glass and metal is made with a uncoated tool [1,2,7,9], a coated tool [5,6,8] or a lubricated tool [3,4].

In these experiments carried out on these experimental platforms, three approaches were used to bring the glass into contact with the metallic support.

- The first one involved spreading molten glass beads [4] or glass gobs [7] on flat metallic substrates, with the objective of analyzing the adhesion between the glass and the metal. This approach was based on a photoelectric timing unit that detected the progression of the glass flow [4] or a rapid camera that filmed the glass flow [7].
- In the second approach, the glass was contained inside a crucible. Either the punch contacted the hot glass according to the required contact pressure [2,3,5,8] or the glass contacted the punch after the blowing operation [1]. In our first three references [1-3], the glass/tool contact was thermally characterized using a temperature measurement obtained by the thermocouples located close to the tool/glass contact surface [1-3] and in the glass [2]. In Manns *et al.* [5] and Fischbach *et al.* [8], the glass/tool adhesion was investigated using the measurements from transducer load cell.
- In the third approach, small gobs (weight < 5g) were pressed between two flat heated substrates in a horizontal [6] or vertical position [9]. After pressing, the small gob was detached from the two flat substrates. Rieser *et al.* [6] measured the separation time, called sticking time, in terms of the initial substrate temperature. Falipou & Donnet [9] measured the strength of the glass/-substrate separation with a force transducer.

Among all these platforms for analyzing the thermal and mechanical behaviors at glass/tool interface, only three studies [5,6,9] allowed the glass to contact the tool during the pressing cycle, as in industrial situations. The solution of these authors was to reproduce the frequency and duration of the contact between the glass and the tool, with the goal being to reproduce the thermal cycle of industrial tools within the experimental platform's metallic support and to detect the important forming parameters that lead to the glass sticking to the tool.

In addition to the experiments on their platform, certain authors examined the metal and glass surfaces, before and after the contact, with Atomic Force Microscopy (AFM) [7] or optical profilometers [8]. The objective was to study the surface morphology or the corrosion state of the contact surface. Other authors determined the surface composition of the contacting materials using measurements made with an Scanning Electron Microscopy equipped with energy-dispersive X-ray Spectroscopy (SEM/EDX) [5,7-9] and X-ray Photoelectron Spectroscopy (XPS) [9].

This study proposes a new way to analyze the impact of the lubrication on glass/tool thermal exchanges in the manufacturing of luxury perfume bottles. The TEMPO Laboratory has an experimental Glass/Tool Interaction (GTI) platform, which is a reduced-scale production unit that allows researchers to reproduce the pressing cycle conditions encountered in the glass industry. Part A of this study defines the joint experiment carried out by the TEMPO Laboratory and the BCR Center to analyze the lubrication's impact on thermal exchanges and physico-chemical changes at the glass/tool interface. Part A presents our experimental conditions and the available thermal results on the GTI platform for the case of contact between flint glass and a punch swabbed with the lubricant developed by our partner, SOGELUB® Special Lubricants Company. To complete the analysis, the BCR Center took physico-chemical measurements on the glass samples produced after trial. Part B of this study presents our analysis of the different lubrication situations under industrial processing conditions.

2. The Glass/Tool Interaction Platform

In order to continue its investigations on the glass/tool interface [10], the Glass Forming and Tempering (GFT) research team in the TEMPO Laboratory (Valenciennes, France) purchased a Glass/Tool Interaction (GTI) platform in July 2007. This platform is a small production unit designed by the Philips Glass Research Center in Eindhoven (Netherlands). With GTI, this Center conducted investigations on the glass/tool interface with their production units to manufacture cathode ray tube (CRT) TV screens. For the GFT research team, the main objective of purchasing the GTI platform was to investigate the thermal impact of lubrication at the glass/tool interface under industrial processing conditions.

2.1. The GTI Platform and Its Components

First The GTI platform is composed of four main parts: the furnace, the cutting device, the pressing device, and the computer-aided control and acquisition system (**Figure 1(a)**).

• Furnace: The furnace is composed of an Inconel tank, which can be filled with 40 dm^3 of cullet. This tank is placed in a 10kW electric furnace. The maximum temperature for melting the glass is 1060˚C. At this temperature level, the molten glass flows by gravity through the cylindrical tube located in the tank center. There is an electrical resistance around the tube to increase the glass temperature up to a maximum of 1130˚C, thus adjusting the flow rate to the desired level.

• Cutting device: The glass flow is automatically cut into gobs by the controlled shear blades of the cutting device. The cutting time and the glass temperature at the furnace tube exit determine the gob weight. For example, with an interval of 7s between two cuts and a flint glass temperature of 1100˚C, the weight of the gob is close to 40g.

• Pressing device: The glass gob is then transferred into the pressing device via a delivery mechanism, which then drops one of three molds on the mold conveyor (**Figure 1(a)** and **1(b)**). The conveyor rotates, and the glass gob is pressed by the punch according to the preset forming parameters given by the control system (**Figure 1(a)**). (More details on the pressing operation are given in section 2.2.)

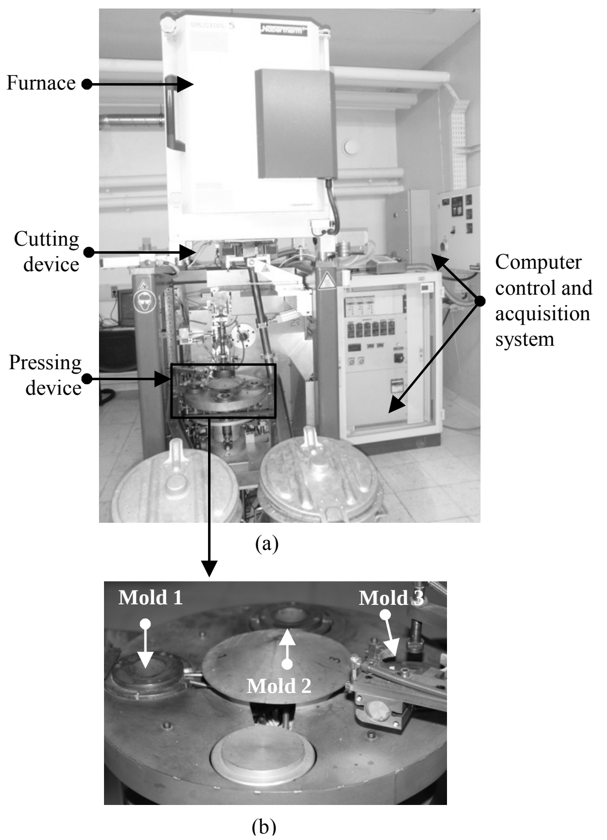

Figure 1. The Glass Tool Interaction platform: (a) the four main parts, (b) the three molds placed on the mold conveyor belt.

The punch temperature on the GTI platform may be adjusted between 20˚C and 700˚C, and the contact pressure between 0.1MPa and 0.6MPa. Depending on the glass weight, the sample produced by pressing on the GTI platform has a maximum diameter of 40mm and a maximum height of 10mm. After a second rotation of the conveyor, the glass sample is ejected to be cooled at ambient air temperature for the next physico-chemical analysis.

• Computer-aided control and acquisition system: The computer-aided control and acquisition system of the GTI platform is connected to specific components, such as local electrical heaters, shear blades, the mechanical components related to kinematics of the pressing device, and the thermal and mechanical sensors. For the control, these elements permit us to reproduce on the GTI platform the experimental conditions as close as possible to industrial practice. For the acquisition, the sensors on the GTI platform make it possible to analyze the thermal behavior at the glass/tool interface. (More details on the input and output data from the GTI platform are given in sections 2.2 and 2.3.)

One trial on the GTI platform will match the performance cycle of glass melting, glass cutting, the routing for the mold load cycle in the pressing device, the pressing of the glass in the mold by the punch, and glass ejection from the mold.

2.2. The GTI Platform's Pressing Cycle

The main purpose of the GTI platform is to analyze the glass/tool interface throughout successive pressing cycles. In this section, we describe the pressing cycle on the GTI platform in detail. The pressing device (**Figure 1(a)**) is the critical place where the contact between the glass and the punch occurs. On the top, there is a punch and a ring around it. On the bottom, there is a conveyor with three molds (**Figure 1(b)**). Each mold is made up of two parts: the cylinder and the ejector (**Figure 2(a)**). As shown in the cross-section in **Figure 2(a)** and **2(b)**, the punch, the ring, each ejector and each cylinder are instrumented by type-K thermocouples with a diameter of 1mm. For each thermocouple, the temperature measurement is carried out at 1.5mm from the surface of the metal items (*i.e.*, punch, ring, cylinder, and ejector). Two piezoelectric force sensors are respectively located below the ejector and above the punch to control the contact force effort of the contact between the punch and the glass. On the GTI platform, the acquisition rate is 1000 measurements per second for each channel.

After the shear blades cut the glass, the gob falls into the mold via the delivery mechanism (**Figure 2(a)**). One pressing cycle on the GTI platform includes the following six steps:

1) The conveyor rotates one quarter turn to carry the mold-gob set below the punch (**Figure 2(b)**).

2) The mold-gob set moves upwards so that the cylinder enters in contact with the ring. The gob and the ejector are in an intermediate position. Due to the space between the punch and the ejector at this time, the glass gob does not come in contact with the punch (**Figure 2(c)**). Temperature and effort measurements are begun at this time.

3) The ejector now moves upwards (**Figure 2(d)**) to bring the glass gob into contact with the punch, with the contact pressure and the contact duration defined by the user. The minimum contact duration is 20s to let a glass sample be sufficiently cooled to be ejected from the mold.

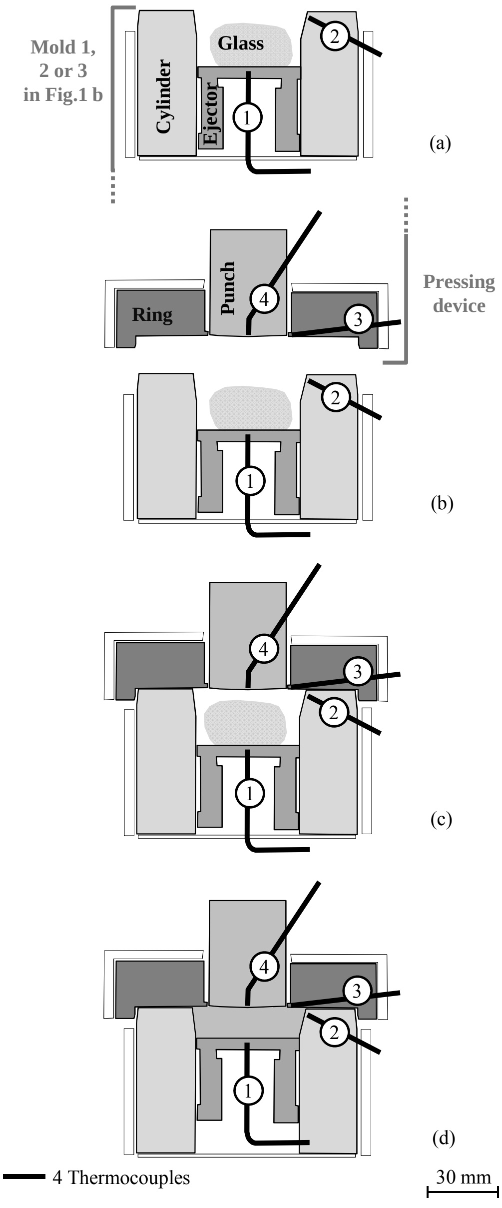

Figure 2. One pressing cycle on the GTI platform—(a) elements of the mold in the initial position; (b) elements of the pressing device and the location of thermocouples; (c) intermediate position during the pressing cycle; (d) position during the glass/punch contact.

4) At the end of the forming process, the punch moves upwards from the glass surface. Temperature and effort measurements are stopped at this time.

5) The glass sample and the ejector return to the intermediate position. The punch moves upwards to its initial position. The cylinder-ejector-glass sample set moves downwards.

6) The mold conveyor rotates one quarter turn, and the glass sample is ejected. Meanwhile, the next mold loaded with glass gob is brought under the punch to be pressed.

To perform one trial on the GTI platform means performing a number n of pressing cycles, with each pressing cycle consisting of the steps described above.

2.3. Experimental Procedure on the GTI Platform

The objective of the present trials performed on the GTI platform was to compare output temperatures under different lubrication conditions of the punch surface. For that purpose, we developed an experimental procedure to guarantee the repeatability of the trials for the same lubrication conditions. In addition, to accurately compare between the trials under different lubrication conditions, we had to guarantee that, except for the lubrication on the punch, the other GTI input data remained quite stable over a set of trials, even if they were performed over a large time interval.

The experimentation was carried out according to the following experimental procedure. Since the melted glass flows by gravity at the exit of the furnace, before each GTI trial, the same volume of cullet (25 dm^3) is loaded into the Inconel tank. Twenty-four hours before the trial, the cullet is melted to reduce most of the air bubbles in the melted glass. The surfaces of the cylinders, the ejectors of the three molds, and the punch are polished to obtain a surface roughness approximating the inner faces of a new blank mold used in the perfume bottle industry (*i.e.*, a roughness Ra from 1 µm to 2 µm). After polishing, the cylinders and ejectors are lubricated to prevent oxidation and binding during testing. Then, the molds (*i.e.*, cylinders and ejectors) and the punch are pre-heated to 450˚C for four hours before beginning the trial.

In this study, four lubrication conditions were analyzed: a bare punch and no lubrification, a punch swabbed with a lubricating paste, a punch coated with resin, and a punch first coated with resin and then swabbed with a lubricating paste. According to the lubrication conditions to be tested, the experimental procedure was thus different because of the punch lubrication. In Part A, we present the trials which used a punch swabbed with a lubricating paste. (The trials with the other lubrication conditions are presented in Part B).

In the trials with the lubricating paste, the swabbing of the punch was performed with a cotton swab just before the beginning of the trial. This cotton swab was saturated with the lubricant. To reproduce the same lubrication conditions for each trial, the cotton swab was only used for one single trial.

When the resin film was added, the coating on the punch is put on eight hours before the trial, and the roughness was measured again after drying in the open air. Pre-heating the punch to 450˚C for four hours allows the resin film to tighten and extends the lifetime of the resin film. In the case of the trial with a coated/swabbed punch, preheating the punch makes swabbing the punch surface with the lubricating paste optimal.

To perform a trial on the GTI platform, we distinguish input data, related to the instructions that are entered, from output data, given by different sensors on the platform. Input data for the GTI control system concern the temperature of the electric furnace, the temperature level of the heating resistance at the exit of the furnace, the cutting time, the initial temperature of the molds and the punch, the duration of the gob pressing, and the contact pressure between the glass and the punch. Values used for GTI trials in Part A and Part B were respectively 1060˚C, 1130˚C, 7s, 450˚C, 20s and 0.2MPa. We explain in Part B the choice of these input data values in terms of the industrial process used to manufacture perfume bottles.

A distinction was made between the output data used for control and the output data used for analysis. During the trial, the weight of each glass sample was the first output control data on the GTI platform. The glass weight was checked after each ejection throughtout the trial. With n pressing cycles in one trial, less than n glass samples were obtained because some samples were broken during the ambient-air cooling period. The objective was to keep the glass weight constant for each pressing during the trial in order to get a constant value for the pressing force delivered by the punch.

In fact, the thermal exchange at the punch/glass interface is totally dependent on the pressure level [3], and the pressing force must be constant during all the trials. With a cylindrical cavity for each mold, a minimum glass volume must be present to fill the mold in order to apply a given pressure distribution on the glass during each punch pressing cycle. As the mold diameter is 43.4 mm and we wanted to produce glass pieces with a height of at least 10mm, the total glass volume is 14 800g/mm3. With the goal of obtaining a 0.2MPa pressure on the glass during the pressing cycle, the preliminary trials on the GTI platform permitted us to determine that the weight of the glass sample has to be at least equal to 37g.

There are 6 other output control data elements issued from sensors at different locations in the GTI platform: the air temperature given by the type-K 1mm-diameter thermocouple located near the heating resistance, the glass temperature given by the bi-chromatic pyrometer just before the cutting area, the three ejector temperatures given by the three type-K 1mm-diameter thermocouples (**Figure 2**), and the force given by the piezoelectric force sensor in the ejector. They were used to verify that GTI trials were made under the same conditions.

Only one output analysis data element was used for the thermal analysis of the punch. This data element was obtained by the type-K 1mm-diameter thermocouple inside the punch (**Figure 2**). The output data recording started at the beginning of each pressing cycle. With a 20s contact duration, the total acquisition duration for each output data element during one trial is t = 22s x n, where n is the total number pressing cycles in one trial.

In the research presented in Part A and Part B, the single parameter that distinguished the different trials is the lubrication on the punch. Each trial is repeated three times for each lubrication condition. Consequently, for each output data element, the average value and the relative standard deviation along time t were computed. The preliminary trials made on the GTI platform permitted us to determine the maximum accepted value for the relative standard deviation for each output control data element. Thus, if the relative standard deviation during one trial reaches or exceeds the critical value of the output control data element, the trial was performed again with the same input data. By using the same input data, we could verify the repeatability of the trial made on the GTI platform for cases of very similar output control data.

3. Analysis of the Thermal Exchange at the Glass/Punch Interface with the GTI Platform

This section presents our main results for the analysis of the thermal exchange at the glass/punch interface. Part A concerns the trials performed with a punch swabbed with the lubricating paste. The punch was made of cast iron, like in industrial practice. The lubricating paste was designed by our partner, SOGELUB® Special Lubricants Company. According to the experimental procedure, the punch was lubricated only once at the beginning of the trial.

3.1. Temperature Evolution of the Punch During the Pressing Cycle

According to our experimental procedure, three trials were performed to guarantee trial repeatability, to estimate average temperature evolution, and to observe the impact of lubrication on the thermal exchange at the glass/punch interface. On the GTI platform, a hundred pressing cycles are possible with 25 dm^3 of cullet and 37g glass gobs. In the preliminary trials, we observed no signifycant results if the trials were performed in order to empty the cullet tank. In order to obtain a reasonable duration for one trial, we decided to perform 51 pressing cycles for each trial. Each of the 3 molds on the conveyor belt (**Figure 1(b)**) was filled 17 times, which led to 51 glass samples produced. With a 22s duration of the pressing cycle and a 20s contact time between the glass and the punch, the total time for the 51 pressing cycles is 1122s, and the total contact time between the glass and the punch is 1020s.

As mentioned in section 2.3, there were 7 output data elements from the GTI platform, which were used to check the trial validity. In Appendix, **Table A.1** gives the average values of these 7 output data elements, with the relative standard deviation in parentheses. These data were analyzed in two steps. In the first step, we analyzed the data, trial by trial, to check that no data drift appeared in the input data during the trial. In this analysis, the information in the **Table 1** had to be scrutinized, line by line, for the first three lines. In the second step, we analyzed the three trials together to prove that the experimental conditions were similar for the three trials. The last line in **Table A.1** gives the results for this second step for the 7 output data elements.

For the first analysis, the stability of the glass sample's weight during the trial was the first priority, given the pressing force during each pressing cycle throughout one trial. In **Table A.1**, the weight of the glass sample for the 3 trials is greater than 37g, allowing us to set the pressure equal to 0.2MPa during the 3 trials. For the 6 other output data elements, the relative standard deviations for the 3 trials are between 0.1% and 3.7%. We conclude that the output data were stable during each trial, and therefore the experimental conditions did not vary during each of the 3 trials. For the second analysis, given in the last line in **Table A.1**, the relative standard deviation for the 7 output data varies between 0.32% and 3.6%. We thus conclude that the output data were stable around similar values during each of the 3 trials, thus proving that the 3 trials were performed under the same experimental conditions.

Table 1. EDS results for the bare punch surface.

Elements	%
E1	96.6
E2	2.4
E3	0.6
E4	0.4

The analysis of the thermal exchange at the glass/punch interface is based on the temperature evolution during each pressing cycle, given by the type-K 1mm-diameter thermocouple located 1.5mm from the punch surface in contact with the glass gob during the pressing cycle (**Figure 2**). According to our previous analysis of the 7 output data elements for the 3 trials performed in the same experimental conditions, the average temperature inside the punch was computed from the evolution of the 3 temperatures obtained in the 3 trials.

This average temperature was used to analyze the impact of the lubrication on the thermal exchange at the glass/punch interface. During the 51 pressing cycles, the punch pressed the glass in mold 1, 2 or 3, one after the other, through the rotation of the mold conveyor belt (**Figure 1(b)**). As shown in **Figure 3**, among the 51 pressing cycles, we distinguished the 17 temperature evolutions for mold 1 (solid black lines), mold 2 (solid grey lines), and mold 3 (dotted black lines).

Our first global analysis of the temperature evolution led us to distinguish 2 phases in the punch's temperature evolution.

The first phase is related to the time interval between 0s (beginning of the trial) and 132s (end of the 6^{th} pressing cycle). During this interval, the temperature inside the punch gradually increases. Although the punch had been initially heated to 450˚C, the punch was now heated by coming into contact with the glass during the first 132s of the cumulated contact time. After 132s, the punch's temperature became stable, even though oscillations appeared due to the ambient air cooling the punch when it is no longer contact with the glass.

Considering now the pressing cycle for one mold (*i.e.*, 1, 2 or 3), the temperature inside the punch evolves quite similarly after 132s. The temperature increases followed by decreasing temperatures for a reduced time. This is explained by the fact that, during the 22s of the pressing cycle, the punch is in contact with the glass in the first 20s, after which the punch is no longer in contact with the glass and is cooled by ambient air. The temperature evolutions are similar in the 3 consecutive pressing cycles for mold 1, mold 2 and mold 3. However, the initial temperature of the punch at the beginning of the pressing cycle is different. The temperature of the punch has the lowest value for the pressing cycle for mold 1 and the highest value for mold 3. The explanation is related to the conveyor rotation between two consecutive pressing cycles. Between mold 1 and mold 2 and between mold 2 and mold 3, the conveyor (**Figure 1(b)**) rotates a quarter revolutions, but between mold 3 and mold 1, the conveyor has to rotate three-quarters of a revolution in the reverse direction to place mold 1 once again under the

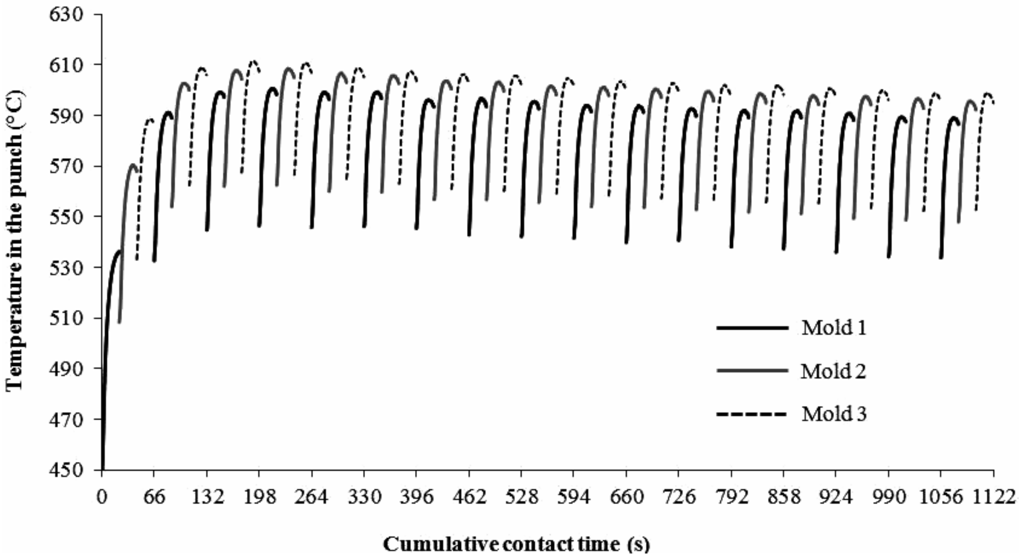

Figure 3. Temperature analysis using the GTI platform—average temperature evolution of the punch in the 3 trial repetitions for the pressing cycles for molds 1, 2 and 3.

punch. Consequently, the punch is cooled by ambient air much longer.

Guilbaut *et al.* [11] and Zhou *et al.* [12] have measured the temperature in the molds during industrial glass pressing. In their studies, the temperature in the industrial mold evolved in function of the cumulated pressing time and presented a similar evolution our results for mold 1, 2 or 3 (**Figure 3**). Guilbaut *et al.* [11] and Zhou *et al.* [12] also highlight a transient phase when the mold temperature increased after the beginning of production and a stable phase in which oscillations were observed depending whether the punch was in contact with the glass or was cooled by ambient air. Consequently, the GTI platform, with a reduced number of pressing cycles (*i.e.*, 51 on the GTI platform versus several hundreds for the studies Guilbaut *et al.* [11] and Zhou *et al.* [12]), was able to reproduce the thermal behavior at the glass/tool interface, as observed in a industrial context.

Using **Figure 3**, we analyzed the temperature evolution in the punch, looking each of the three molds, on case-by-case basis. **Figure 4** presents the temperature evolution in the punch when the pressing occurs for mold 1.

For this case, only the 17 temperature evolutions in solid black lines shown in **Figure 3** were plotted. For each pressing cycle, the punch's average temperature during the pressing cycle was computed from the 22000 temperature values, with a 22s pressing cycle and a 1000Hz acquisition rate.

By interpolating these 17 average temperatures with a 6th degree polynomial, the interpolated average temperature evolution was obtained (solid grey line in **Figure 4**).

Based on the numerous preliminary trials performed on the GTI platform, we conclude that this interpolated average temperature evolution for mold 1 is an important assessment criterion for analyzing the thermal exchange at the glass/punch interface. If the interpolated curves obtained for mold 2 and for mold 3 were considered, similar evolutions were obtained in our study of the impact of lubrication on the thermal exchange at the glass/punch interface. Moreover, using mold 1, mold 2 or mold 3 as a reference, identical conclusions are founded, even though temperature in the punch at each pressing cycle beginning is different. The comparison with other different lubrication conditions (Part B of this study) was based only on this interpolated average temperature obtained from the pressing cycles performed for mold 1. The information comes from the punch's average temperature evolution for the 51 pressing cycles after 3 trials repeated in the same experiment.

3.2. Temperature Evolution on Two Specific Pressing Cycles

Figure 3 shows the average temperature evolutions for all of the 51 pressing cycles during the trial with the punch swabbed with the lubricating paste. We now focus more specifically on one pressing cycle in the transient phase between 0s and 132s (132 s in **Figure 4** corresponds to the end of the 6th pressing cycle) and one pressing cycle in the stable phase after 132s.

In the transient phase, we selected the first pressing cycle. The first pressing cycle is specific because the punch's initial temperature, which is one of the input data for the GTI platform, is strictly identical in all the trials in this study, whatever the lubrication condition of the

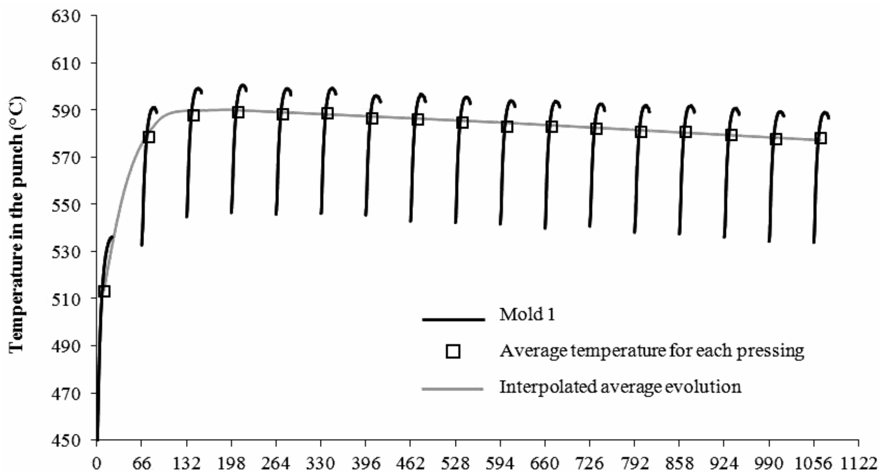

Figure 4. Temperature analysis using the GTI platform—average temperature evolution of the punch in the 3 trial repetitions for the pressing cycles for mold 1, evolution of the average temperature for each pressing cycle, and interpolated average evolution of the punch for the pressing cycle for mold 1.

punch. **Figure 5(a)** presents the 5 first seconds of the temperature evolution of the punch, obtained during the first pressing cycle with the swabbed punch. We selected only the 5 first seconds because 5s is representative of the industrial practice in terms of the length of the 2 successive blowing operations used to produce luxury perfume bottles. During these 5 first seconds, the temperature increase at 1.5mm from the punch surface is significant (+ 50˚C), and the variation over time is almost linear (**Figure 5(a)**).

In the stable phase, any pressing cycle located after the 6th pressing cycle could be chosen since the punch's temperature evolution during each pressing cycle is similar. The 10th pressing cycle was arbitrarily chosen. **Figure 5(b)** provides the average temperature evolution during the first 5s of this 10th pressing cycle. The temperature increase at 1.5mm from the punch surface is reduced compared to the first pressing cycle, with + 35˚C.

Moreover, after 6 pressing cycles, the punch's initial temperature at the beginning of the pressing cycle is now 546˚C (+ 102˚C compared to the first pressing cycle) because the punch has been heated by the hot glass during the first 9 pressing cycles. With the punch's initial temperature higher than at the beginning of the trial and a reduced temperature increase (+ 35˚C compared to + 50˚C), we may conclude that the heat exchange at the glass/punch interface in the stable phase is less significant than in the first pressing cycle. Thus, the temperature evolution in **Figure 5(b)** is no longer linear.

Since the evolution shown in **Figure 5(b)** is quite the same during the rest of the trial, we may also conclude than no significant change appears in the thermal exchange at the glass/punch interface during the 45 pressing cycles in the stable period. According to the GTI trials and observations

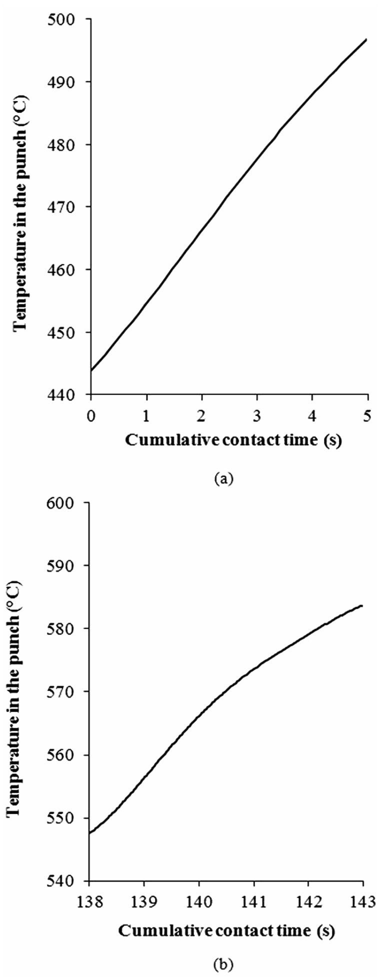

Figure 5. Average temperature evolution of the punch during 5 seconds: (a) for the 1st pressing cycle, (b) for the 10th pressing cycle.

of the temperatures inside the punch throughout the trial, we may also conclude that no significant loss of the lubricating paste on the punch has occurred during the 51 pressing cycles, as well as during the stable period after 132 s (**Figure 3, Figure 4**). The same pressing cycles (*i.e.*, the first one at the beginning of the transient period and 10^{th} pressing cycle in the stable period) were also used to compare the different lubrication conditions in Part B of this study.

4. Physico-Chemical Analysis on the Surfaces of the Glass Samples Pressed on the GTI Platform

4.1. Use of a Scanning Electron Microscope with Energy Dispersive Spectrocospy

We used a Scanning Electron Microscope (SEM) (JEOL JSM-5900LV) with Energy Dispersive Spectrocospy (EDS) detector on the SEM to analyze and characterize the glass, with the objective of tracing any lubricant transfer on the glass samples during the pressing cycles. The composition obtained by EDS should not be regarded as the exact composition of the analyzed sample, but it allowed us to compare the compositions of materials obtained on the same equipment under the same conditions. We used a constant primary electron beam (20 kV) and a constant sample/detector working distance (12mm). Due to the size of the SEM chamber, both the punches and the glass samples pressed on the GTI platform can be investigated in their entirety.

To identify the elements of the resin film or the lubricating paste as markers in order to detect any lubricant transfer on the glass samples, a first EDS analysis was made on the bare punch to produce the forming tools (**Table 1**). (To respect the confidentiality of our industrial partners, the elements are denoted with Ei, where i is equal to 1, 2, …i, depending on their presence in the successive EDS analyses in all the tables.) In industrial practice, the forming tools are coated before being swabbed with a lubricating paste. **Table 2 and Table 3** respectively present the EDS analysis made on a coated punch with the resin film and of a coated punch swabbed with the lubricating paste (Both the resin film and the lubricating paste are the products of the SOGELUB® Special Lubricants Company.) These punches were heated on the GTI platform to 450˚C for four hours, which is the same procedure before the GTI trials described in Section 2.3. Five new elements (E5 to E9) were detected in the resin film (**Table 2**) compared to the EDS analysis of the bare punch (**Table 1**). Two new elements (E10 and E11) were detected in the lubricating paste (**Table 3**).

Comparing the results obtained for the coated punch (**Table 2**) and the coated punch swabbed with a lubricating paste (**Table 3**) with those for the bare punch (**Table 1**), the characteristic elements were selected as markers for the lubrication products. The two main elements present in the resin film and in the lubricating paste (E5 and E6 in **Tables 2 and 3**) were not considered as markers because they were present in both the film and the paste. Since the markers only present either in the resin film or in the lubricating paste but not in the punch nor in the glass (**Table 4**) could not be determined, the following compromise was made:

- E2 and E7 were chosen as representative of the resin film because they are the predominant minor elements in the resin film (**Table 2**).
- E10 and E11 were chosen as representative of the lubricating paste for the same reason (**Table 3**).

Although the markers, E2 and E7, were present in the glass in different quantities (**Table 4**), a significant increase in the EDS signal for these elements highlights any lubrication transfer from the coated punch on the surface of the pressed glass sample. To realize the physico-chemical analysis of the glass samples pressed on the GTI platform, the EDS analysis of the Pochet de Courval flint glass is given in **Table 4**.

Table 2. EDS results for the coated punch surface with no lubricating paste.

Elements	%
E1	38.2
E5	33.8
E6	19.6
E2	4.3
E7	1.9
E8	1.4
E9	0.6
E3	0.3

Table 3. EDS results for the coated punch surface, swabbed by the lubricating paste.

Elements	%
E1	48.9
E6	23.0
E5	22.9
E2	1.4
E8	1.2
E10	0.9
E11	0.7
E3	0.6
E7	0.4

Table 4. EDS results for the flint glass.

Elements	%
E6	46.2
E2	33.8
E8	10.5
E10	6.2
E7	1.1
E12	1.0
E9	0.6
E13	0.5

4.2. Our Observations for the Glass Surfaces Pressed with a Punch Swabbed with a Lubricating Paste

To analyze the possible transfer of the lubricant on the glass samples during the GTI trials, we developed a protocol. The pressed glass samples were collected for each trial and were referenced by their pressing cycle order (*i.e.*, from 1 to 51 since 51 pressing cycles were performed on the GTI platform for each trial). The samples were taken at regular intervals—every tenth sample—for observation. In case a notable change in the aspect, size and/or quantity of markers on the glass surface was observed between two consecutive samples, further intermediate sampling was done.

The observation protocol on a glass sample pressed on the GTI platform was to use a SEM to scan the surface in contact with the punch during the pressing cycle, thus locating possible traces of foreign matter. Once the foreign matter on the surface was detected, a focused EDS analysis was done to identify the source of the foreign matter (*i.e.*, the resin film, lubricating paste or other matter). For Part A, we made similar observations on the three series of glass samples pressed for the three repetitions of the trial with the punch swabbed with the lubricating paste (designated as repetition 1, repetition 2 and repetition 3).

The presence of small dark spots was observed on the surface of the glass samples throughout the pressing cycles, not at the beginning or the end of the GTI trial. **Figure 6** to **9** shows typical pictures of these spots, respecttively obtained during the 20th, 30th and 50th pressing cycles of repetitions 1 and 3. The presence of these small spots was irregular on the surface of the glass samples, and the spots were mainly grouped on areas less than a few square millimetres large. Most of these spots were between 10 and 100 µm in size (**Figure 6, 8** and **9**). As shown in **Figure 7**, some were larger, about 500µm, and these larger spots, observed in series 1, 2 and 3, looked like clusters of small spots.

Figure 6 shows a representative SEM picture (sample 30/repetition 1) of the foreign matter traces on the glass sample surface. The dark traces are the foreign matter on the grey background of the glass. The EDS analysis for the trace marked by the white square □ in **Figure 6** was realized and the composition puts forward the presence of elements E10 (16.5%) and E11 (11.3%). The other traces in **Figure 6** were analyzed, and similar percentages were found. The elements E10 and E11 are markers for the lubricating paste. Lubricating paste residues were found in the dark grey spots, so we concluded that the lubricating paste spots were transferred on the pressed glass surface by the punch.

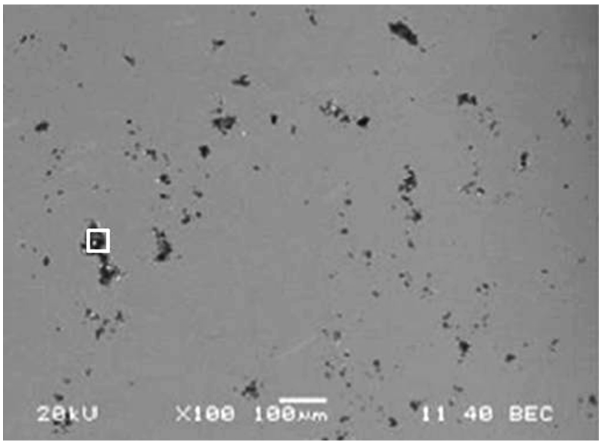

Figure 6. SEM image of lubricant traces (dark grey) on the surface of the 30th glass sample pressed with Punch 1 on the GTI platform.

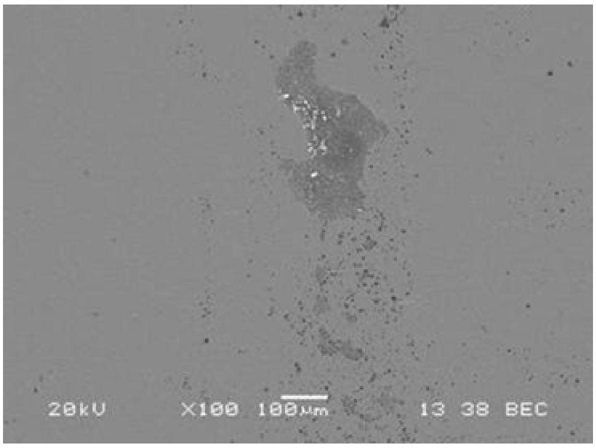

Figure 7. SEM image of lubricant traces (dark grey) on the surface of the 20th glass sample pressed with Punch 3 on the GTI platform.

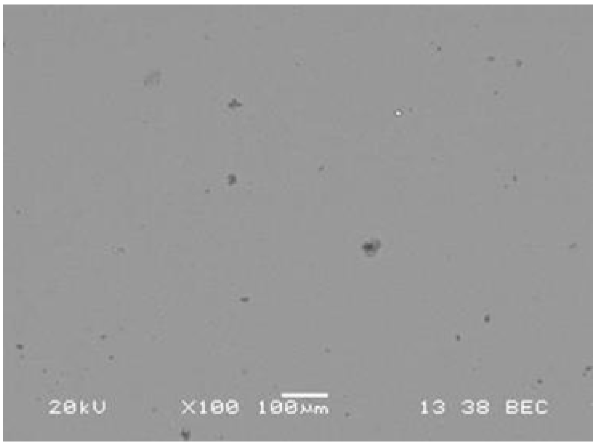

Figure 8. SEM image of lubricant traces (dark grey) on the surface of the 50th glass sample pressed with Punch 3 on the GTI platform.

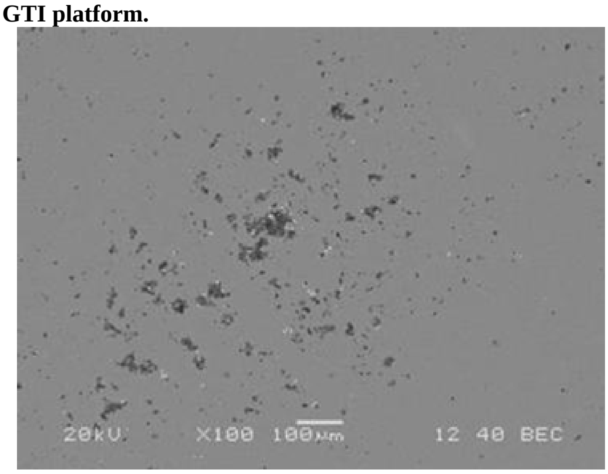

Figure 9. SEM image of lubricant traces (dark grey) on the surface of the 50th glass sample pressed with Punch 1 on the GTI platform.

Furthermore, tiny white points (**Figure 6** to **9**) were often observed in the area surrounding the lubricating paste traces. A closer EDS analysis on these points showed that they are rich in an element present in the lubricating paste but not already designated in **Table 3**, which reported the results of the EDS analysis of the swabbed punch. The explanation is that some of the elements present in a very small quantity in the resin film or in the lubricating paste were not detected by the global EDS analysis on the punch coating before the GTI trial. Consequently, they were not listed in **Table 2** for the coated punch, nor were they listed in **Table 3** for the coated and swabbed punch. Nevertheless, they became detectable by the close analysis of the areas where the traces of the lubricating paste or resin film from the punch were concentrated. According to the close EDS analysis done on the series of glass samples (not only for the coated and swabbed punch but also for the others presented in Part B), this element detected in the tiny white points, denoted E14, is a potential marker simultaneously in the lubricating paste and the resin film. It was an important element in the modification of the actual composition of the lubricating paste, as presented in Part B.

5. Conclusions

In this study, the TEMPO Laboratory (Valenciennes, France) and BCR Center (Mons, Belgium) has proposed a new way to analyze and improve thermal capacities of lubricants for the manufacturing of flint glass perfume bottles. In Part A, we considered the pressing of the flint glass with a punch swabbed with the lubricant developed by our partner, SOGELUB® Special Lubricants Company. For the GTI platform, we developed an experimental procedure to guarantee the repeatability of the trials that respects the initial input data conditions. After 3 trials on the GTI platform under the same input data conditions, the average temperature evolution of the punch was calculated throughout the pressing cycle. For the Physico-Chemical analysis using SEM and EDS observations, the BCR Center developed a procedure to detect the possible lubricant traces on the surfaces of the pressed glass samples, and EDS was used to analyze the samples, case by case.

The thermal analysis results with the GTI platform are:

- The average temperature evolution obtained for the 51 pressing cycles on the GTI platform presents a transient phase when the punch was heated by the hot glass during the first six pressing cycles.
- For the following pressing cycles, the average temperature during each pressing cycle is almost stable, with oscillations due to duration of the contact of the punch with the glass and the ambient-air cooling times.
- The temperature increase at 1.5mm from the surface of the punch during the first 5 seconds of the first pressing cycle is important (+ 50˚C), and the variation over time is almost linear.
- In the stable phase, during the first 5 seconds of the 10^{th} pressing cycle, the temperature increase in the punch is less in the first pressing cycle (+ 35˚C) due to its higher initial temperature at the beginning of the pressing cycle (+ 102˚C). The temperature evolution is no longer linear.

We conclude that the heat exchange at the glass/punch interface in the stable phase is less important than at the beginning of the trial. According to the GTI thermal measurements, with the stable state of the punch's average temperature over one pressing cycle after 6 pressing cycles and similar oscillations due to the contact duration and the ambient-air cooling, we also conclude that there is no significant increase of the heat transfer at the punch/glass interface, and thus no significant loss of the lubricating paste during the pressing cycle.

With the first EDS results on three punch conditions (*i.e.*, bare, coated and coated/swabbed) before the GTI trials, markers were identified for the resin film and for the lubricating paste. For the case of the GTI pressing cycles with a swabbed punch presented in Part A, the results of the physico-chemical analysis on the glass samples were:

- Typical small traces (between 10 and 100 µm) of the lubricating paste were detected on the glass samples, though not at the beginning or at the end of the GTI trial.
- Some clusters of these traces with a larger size (500µm) were also present on some glass samples.
- Other traces from one element of the lubricating paste were also detected because during GTI trials, there was a concentration of this element, present in a very small quantity in the lubricating paste.

We conclude that the total amount of the lubricant transferred after 51 pressings is very small, only witnessed by the presence of sporadic traces on the glass surface in very small quantities on the 51 pressed glass samples throughout the GTI trial, though not at the beginning or at the end. According to the GTI results and the results of physico-chemical analysis, no significant loss of lubricating paste was detected during the 51 pressing cycles (*i.e.*, during 1122 seconds of the swabbed punch pressed against the hot glass).

Part B presents the joint TEMPO Laboratory-BCR Center experiment to analyze the different lubrication conditions in an industrial context. For this purpose, the trials on the GTI platform were performed with a bare punch, a swabbed punch, which is before coated by a resin film, and a punch swabbed by a new lubricating paste.

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Appendix

A. Output data from GTI

Table A.1. Average output data for the reference trial with the relative standard deviation in % in parentheses.

Repetition number	Average glass sample mass (g)	Average temperature of the heating resistance (˚C)	Average glass temperature at the exit of the furnace (˚C)	Average temperature of the ejector n 1 (˚C)	Average temperature of the ejector n 2 (˚C)	Average temperature of the ejector n 3 (˚C)	Average pressing force (N)
S1	38.2 (1.4)	1100 (0.2)	966 (2.3)	556 (1.9)	561 (2.1)	566 (2.3)	405 (3.2)
S2	38.7 (3.3)	1106 (0.1)	958 (2.5)	552 (2.0)	557 (2.2)	562 (2.5)	402 (3.7)
S3	37.6 (1.3)	1101 (0.1)	963 (2.2)	556 (1.6)	562 (1.9)	566 (1.9)	396 (3.4)
For the 3 trials	38.2 (2.5)	1102 (0.32)	962 (2.6)	555 (1.9)	560 (2.1)	565 (2.3)	401 (3.6)

