

Thermo-Optical Set-Up to Investigate Non-Isothermal Glass-Metal Contact

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Abstract. The development of innovative mold materials and coatings for glass forming, relies on a profound understanding of the interaction between glass melt and the respective metallic or oxidic surface. In order to revise the existing theories on sticking temperatures and viscosities, a new thermo-optical setup was constructed that enables the investigation of the non-isothermal bonding behavior of glass melts on different substrates. A glass gob is generated in an upper furnace at temperatures of up to 1200 °C and is then poured onto an individually heated substrate (the respective contact material) at temperatures similar to container glass forming. The resulting movement of the gob at pouring and on the substrate is observed with a high-speed camera. The set-up was tested with a soda lime silicate glass melt in contact with various metallic, carbon-based and ceramic materials. Sticking temperatures for the different materials were determined and compared to the results found in literature. Contact temperatures at which sticking occurs varied significantly with the investigated material. A critical interface temperature (respectively viscosity of $10^{8.8}$ Pas) as obtained by previous researchers could not be found, instead the temperature range for sticking was almost 200 K. Some materials resisted sticking even at temperatures up to 600 °C with the interface viscosity being below $10^{8.8}$ Pas. Interestingly, those substrates also showed non-wetting behavior in previous heating micro-copy trials, suggesting that wettability plays a more important role for sticking than assumed so far.

Keywords: Glass-Metal Contact, Glass-Oxide Contact, Wetting, Sticking

1. Introduction

The interactions between viscoelastic glass and metallic materials play a crucial role in industrial container glass forming. After the glass leaves the spout and the gob is formed at around 10^3 Pas, regular contact of the glass with different materials occurs at multiple stages of the process, and mainly during the forming process, between the cooling glass melt and the blank- and blow-molds [1], [2], [3]. With prolonged use, explicitly the thermally, mechanically and tribologically stressed blank-molds experience increased wear and oxidation. Due to this wear and corrosion process, the glass melt may stick to the mold, causing surface defects in the final product [4], [5]. To delay this sticking and to allow smooth loading of the gob into the blank-mold, a mineral oil- and graphite-based lubricant is applied to the surface of the mold at regular intervals [2], [3], [4]. However, this procedure results in production downtime and raises issues concerning occupational safety and environmental protection. For the development of improved mold materials as well as coatings that can enable a lubricant-free, so-called “dry” loading process in the future, it is necessary to better understand the interaction of a glass melt with metallic and oxidic surfaces.

Common theories on the interaction between glass and a respective material assess the contact in terms of the so-called sticking temperature, which corresponds to a certain viscosity. This temperature at the glass/substrate interface defines the point at which a glass begins to adhere to the contact material. According to this hypothesis, sticking occurs when the glass viscosity which corresponds to the contact temperature at the glass-mold interface falls below a critical value, regardless of the actual contact material or glass composition. In a series of non-isothermal pressing experiments this critical viscosity was empirically found to be at the unique value of $10^{8.8}$ Pas [6], [7], [8], [9].

2. Experimental

The constructed thermo-optical set-up is based on a conventional heating microscope which can be used for sessile drop experiments to determine the wetting and spreading behavior of liquids, details of which can be taken from a recent model description of this behavior [10]. However, two of the biggest downsides of this conventional set up should be avoided, namely allowing only for “static” conditions (i.e. glass and substrate are in contact during the whole trial time) and only allowing isothermal trials (i.e. glass and substrate are at the same temperature during the whole trial).

Also, both restrictions do not precisely reflect the conditions in industrial container glass forming, where the cast iron blank-molds are at temperatures of 400–600 °C and the gob is loaded into those at temperatures around 1000–1100 °C [2], [3]. The setup presented avoids those limitations as it comprises an upper furnace (built by Thermo-Star GmbH) and a lower heating chamber being placed at an optical bench (built by DataPhysics Instruments GmbH). In the upper furnace, the glass gob is generated while the substrate is mounted in the lower chamber. Moreover, the set-up allows the investigation of materials and coatings containing low-melting components like aluminum which cannot be investigated in a conventional heating microscope. A detailed sketch of the set-up is shown in Figure 1.

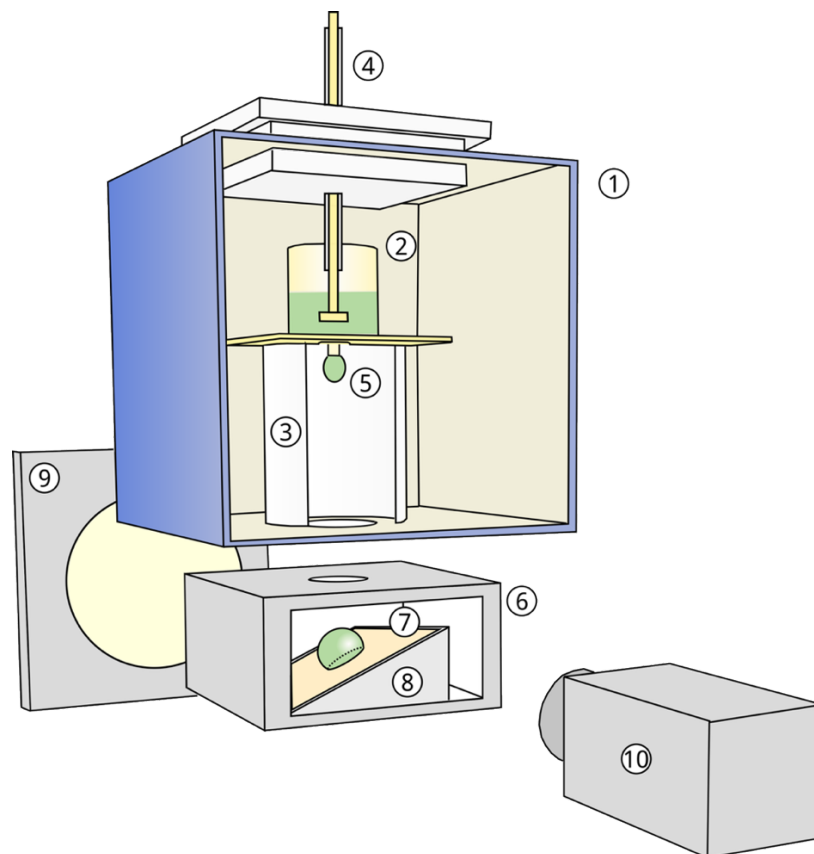


Figure 1. Sketch of the thermo-optical setup: Inside an electrically heated upper furnace (1), the glass is melted inside a crucible (2) which is placed on a support resting on a refractory tube (3). A plunger (4) can be raised to let a glass drop (5) fall into a lower chamber (6). The vertical position of the upper furnace can be changed allowing variable distances between outlet and substrate between 20 and 35 cm. The sample (7) is placed on an (if desired inclined) refractory brick (8). The individually heated chamber is mounted on an optical bench in-between the light source (9) and the camera (10).

In a first sample series all experiments were carried out with a conventional soda lime silica (SLS) glass which main constituents and composition can be found in Table 1.

Table 1. Composition of the investigated soda lime silicate glass.

Oxide	SiO ₂	Na ₂ O	CaO	Al ₂ O ₃	MgO	K ₂ O	Fe ₂ O ₃
Mass fraction (%)	71.6	13.7	11.1	1.9	0.8	0.6	0.3
Molar fraction (%)	71.9	13.3	11.9	1.1	1.2	0.4	0.1

The viscosity temperature curve was described by using the Vogel-Fulcher-Tamman (VFT) equation

$$\log(\eta) = A + \frac{B}{T - T_0} \quad (1)$$

where η is the viscosity and T the temperature of the glass. The empirical VFT parameters A , B and T_0 of the glass were determined by undertaking three different viscometry measurements. Glass transition temperature was determined by dilatometry, beam bending was used for the glass softening range and rotational viscometry for the melting range by carrying out three measurements at 1200 °C, 1300 °C and 1400 °C. The VFT equation was fitted to the collected experimental data by use of the non-linear least square fit from SciPy's `curve_fit` function. The obtained VFT parameters are displayed in Table 2 and were compared to VFT parameters calculated with the statistical Fluegel model [11]. This viscosity model predicts the complete viscosity curve of a glass from its chemical composition based on multiple regression using a global statistical approach with more than 2200 composition–viscosity data. Calculated VFT parameters from the composition given in Table 1 are displayed in Table 2.

Table 2. VFT parameters of the investigated glass composition, determined from viscometry trials and calculated with Fluegel model.

VFT parameter	A	B	T_0
Experiment	-2.753	4322.5	265.0
Fluegel	-2.538	4125.8	277.1

Figure 2 shows the course of the viscosity with temperature. The temperature corresponding to a viscosity of $10^{8.8}$ Pas is 640 °C and was accentuated. The density ρ and the heat capacity c_p of the glass can likewise be estimated using the composition from Table 1 and statistical models based on multiple regression. Fluegel's model for the glass melt density at 1200 °C [12] gives $2367 \text{ kg}\cdot\text{m}^{-3}$ while the heat capacity was calculated with GlassPy's deep neural network prediction model GlassNet [13] to be $1443 \text{ J}\cdot\text{kg}^{-1}\cdot\text{K}^{-1}$. The values are generally consistent with experimental data of similar glass melt compositions in [14]. The thermal phonon conductivity of the glass λ is estimated by the empirical model by van der Tempel [15] to $1.4 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$.

Twelve substrate materials were investigated: Platinum (Pt), a platinum-gold alloy (PtAu5) and alumina (Al₂O₃) were investigated, being typical crucible materials for glass melting. Grey cast iron (EN-GJL-200) was used being a typical blank-mold material. A brass (CuZn37) and a bronze alloy (CuSn6), and four metals (Al, Cu, Fe, Ni) completed the series. Finally, graphite and glassy carbon were chosen due to their known non-wetting behavior in contact with glass melts. Both materials react with atmospheric oxygen to form CO₂ at elevated temperatures. To

minimize the oxidation during the trials a steady flow of argon was applied inside the chamber. Due to the open nature of the system oxygen contents lower than 500 ppm could not be reached.

The investigated substrates were cut to plates with a dimension of 30 x 30 mm². To minimize the influence of the substrate surface roughness, the plates were grinded on a 600-grit diamond plate or SiC abrasive paper and polished up to 1 µm polycrystalline diamond suspension on a semi-automatic grinding machine. Temperature-dependent thermal conductivity and density data of the substrate materials were taken from [16] while temperature dependent heat capacity data was taken from NIST Chemistry WebBook [17].

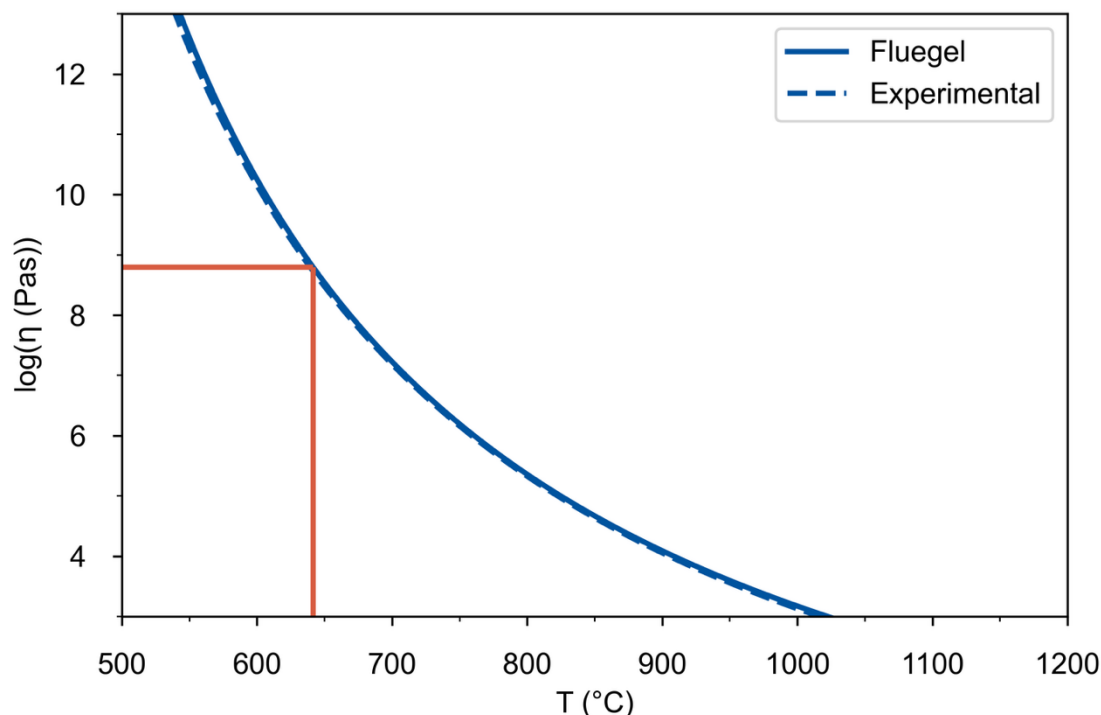


Figure 2. Viscosity-temperature curve of the investigated glass, determined from viscometry and calculated with Fluegel model.

For each of the twelve substrates the following measuring procedure was used. The glass was melted from cullet, heated to 1200 °C with a heating rate of 10 K·min⁻¹ and kept at this temperature for at least one hour to equalize the temperature, while the plunger being in lowered position and closing the crucible's outlet. Meanwhile, the substrate was placed in the lower heating chamber in a way that it was inclined by 25° to the horizontal and resting on a fire brick (see Figure 1). The substrate was heated to 300 °C and held at this temperature for one hour. The temperature of the substrate inside the chamber was measured with a type K thermocouple contacting the substrate. The plunger was then opened and a drop allowed to fall onto the substrate. The temperature of the substrate was then increased in 10 K steps till the gob stuck to the substrate. The reproducibility of this method was tested with cast iron substrates, and the accuracy of the sticking temperature was within an interval of around 10 K. Figure 3 shows a sample recording of the trial, the upper sequence of images showing the last temperature at which no sticking occurred, while the lower sequence shows the trial for the lowest temperature at which sticking arose.

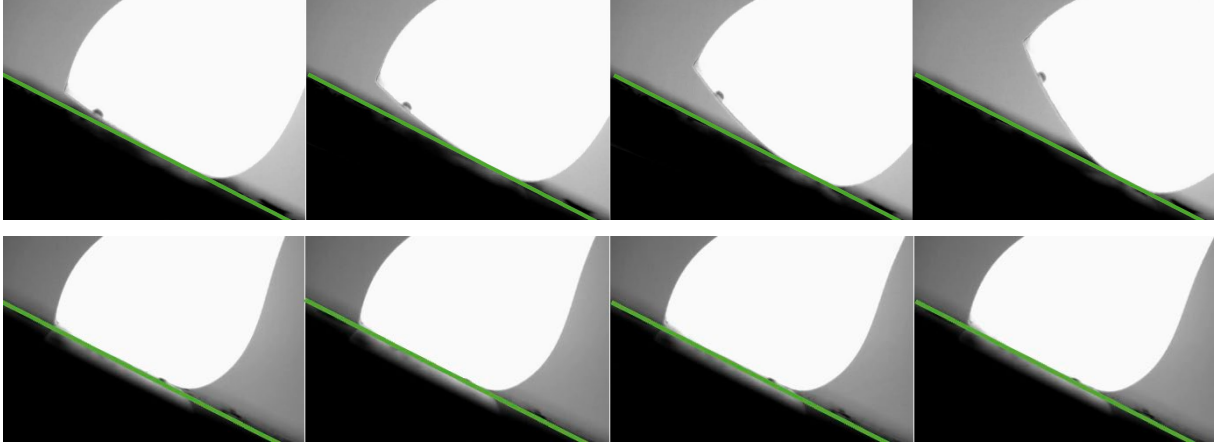


Figure 3. Optical recordings of Glas Metal Contact taken during the trials in case of non-sticking (upper row) and sticking (lower row).

3. Results and Discussion

The measured substrate temperature T_s and the glass temperature T_{gl} at which sticking occurred were used to calculate the interface temperature T_i . The thermal effusivity b

$$b = \sqrt{\lambda \cdot c_p \cdot \rho} \quad (2)$$

which describes the ability of a material to absorb and release heat at its surface, was calculated for the contact material (b_s) and the glass (b_{gl}) according to equation 2 from their respective thermal properties (see chapter 2). Taken into account the measured temperatures of glass T_{gl} and contact material T_s and calculated thermal effusivities b_s and b_{gl} , the interface temperature T_i at which sticking occurred was calculated to

$$T_i = \frac{b_{gl}}{b_{gl} + b_s} T_{gl} + \frac{b_s}{b_{gl} + b_s} T_s \quad (3)$$

[18]. The glass temperature was measured at the outlet of the crucible using a type S thermocouple at the end of the plunger. Thus, some, primary radiative, heat loss will occur during the fall of the gob. Stefan and Boltzmann's law

$$V \cdot c_p \cdot \rho \cdot \frac{dT}{dt} = -\sigma \cdot \varepsilon \cdot A \cdot (T^4 - T_0^4) \quad (4)$$

will apply to radiation with A and V being the surface area and volume of the gob, ε its emissivity, c_p its specific heat capacity, ρ its density, T_0 the ambient temperature and σ the radiation constant. Equation 4 was used to obtain an initial approximation for the cooling of the glass. With a fall duration of not more than 0.5 s and $T_0 > 573$ K (the ambient temperature was at least 300 °C) the drop (with $V = 0.79$ cm³ and $A = 4.7$ cm²) would cool down not more than 20 K until contacting the substrate, the exact amount being dependent on the temperature gradient on the gob's way. Resulting in changes of the interface temperature of no more than 5 K, the cooling of the gob was neglected for the calculation of T_i .

The viscosity of the glass at the interface η_i was calculated using the VFT parameters from Table 1 according to

$$\eta_i = 10^{A + \frac{B}{T - T_0}} \quad (5)$$

Assuming sticking to occur at $10^{8.8}$ Pas, the glass should adhere to the substrate at an interface temperature above $T_i > 640^\circ\text{C}$. Table 3 shows the actual measured substrate temperatures T_s , the calculated thermal effusivity b_s at T_s and the calculated interface temperatures T_i at which sticking occurred during thermo-optical trials for all investigated contact materials.

Table 3. Substrate temperature T_s (measured) and interface temperatures T_i (calculated) at which sticking occurred.

Substrate	T_s ($^\circ\text{C}$)	b_s ($\text{kJ}\cdot\text{K}^{-1}\cdot\text{m}^{-2}\cdot\text{s}^{-0.5}$)	T_i ($^\circ\text{C}$)
GJL 200	440	13.23	548
Fe	420	15.48	517
Cu	390	37.71	434
CuZn37	400	19.59	480
CuSn6	410	18.05	495
Ni	510	17.81	585
Al	310	25.35	381
Al ₂ O ₃	330	8.32	511
Pt	470	15.34	561
PtAu5	>600 $^\circ\text{C}^*$	-	-
Graphite	>600 $^\circ\text{C}^*$	-	-
Glassy Carbon	>600 $^\circ\text{C}^*$	-	-

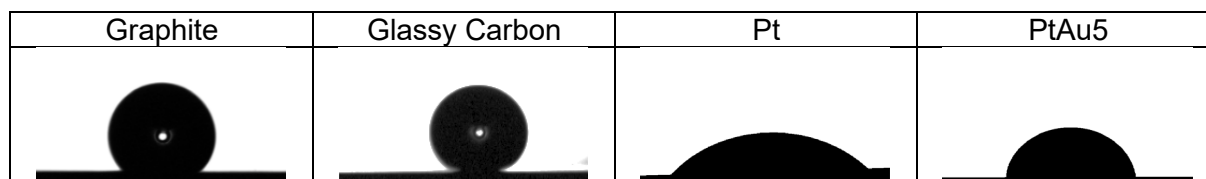
* Sticking was not reached until a substrate temperature of 600 $^\circ\text{C}$

As can be seen from the results in Table 3, no constant interface temperature, resp. viscosity at which sticking occurred could be observed. The interface temperature varied in an interval of around 200 K. Moreover, it can be observed that some materials resisted sticking even at a substrate temperature as high as 600 $^\circ\text{C}$, resulting in interface temperatures above 640 $^\circ\text{C}$ resp. viscosities lower than $10^{8.8}$ Pas. Interestingly, those materials resisting sticking in the trials (see Table 3) also are known for their reduced wettability by glass melts.

Especially graphite and amorphous carbon show no wettability in contact with the investigated glass melt. Static contact angles of 130° and 150° were observed in additional isothermal trials carried out in a conventional heating microscope at 1000 $^\circ\text{C}$ in an atmosphere of argon (see Table 4). No sticking of the glass to the substrate occurred at the end of the trial. On the other hand, most metallic materials showed (almost) complete wetting in the same trials, platinum and its alloy being an exception. It becomes apparent that while platinum is wetted readily by the glass, platinum gold showed reduced wettability (see Table 4). Again, the same tendency can be seen in the sticking experiments. This behavior might be explained by the higher inertness of the platinum gold alloy and its reduced tendency to adsorb oxygen from the atmosphere as was discussed before [19].

As apparent, due to the trials being conducted under air, the majority of the substrates suffers oxidation. Hence, not the pure metal surface is tested but the respective oxides. However, the generated oxide-layer has an insignificant influence on the thermal effusivity due to its very limited thickness resulting from the test duration. This inevitably leads to the conclusion that, in addition to the pure thermal properties of a substrate, such as λ , c_p and ρ , other aspects also influence the bonding behavior. As recently discussed [10] for both a typical SLS-glass and metal oxides the oxygen anion occupies most of the volume and largely determines the interaction between melt and contact material, rendering the initial type of metal less significant. In a recent isothermal heating microscopy experiment at 1000 $^\circ\text{C}$, an almost non-wetting behavior and a contact angle of 100° could be observed for a SLS glass melt on a copper substrate in a very pure and largely oxygen free atmosphere. In future experiments more non-noble metals will be investigated under atmospheres as oxygen free as possible to investigate the true influence of the metal atoms.

Table 4. Silhouette images of investigated glass in contact with different substrate materials at 1000 °C in isothermal heating microscopy trials.



4. Conclusion

A thermo-optical setup for non-isothermal sticking trials was build and tested successfully on soda lime silicate glass. The results of a first sample series show that the temperature at which sticking occurs varies significantly with the investigated substrate material. A single, universally applicable critical interface viscosity (respectively temperature) as reported before in the literature [6], [7], [8], [9] could not be found and the observed interface temperature range for sticking was almost 200 K. This clearly indicates that additional interface parameters beyond the thermal effusivity influence the sticking behavior. Moreover, it is important to note that materials which show only partial wetting or even non-wetting behavior resisted to sticking even below interface viscosities of $10^{8.8}$ Pas. This again connects sticking to wetting and hence highly surface dependent interactions, beyond thermal conductivity, density and heat capacity.

The results therefore show that wettability plays an important role in the glass to metal contact and the sticking tendency and suggest revising existing theories on sticking. Especially if a wettability of the substrate by the glass can be avoided, it seems to be possible to exceed the predicted sticking temperatures. While so far due to a too fast cooling of the gob, the wetting and spreading behavior of the melt cannot be investigated in the presented setup to a satisfactory extent, a modification of the setup to allow for such trials is under construction. The authors expect a correlation between wetting and sticking tendency to become even more apparent then.

Data availability statement

Data of presented results can be provided upon request.

Author contributions

Paweł Engelmann: Conceptualization, Investigation, Data analysis, Visualization, Writing – original draft. Christian Roos: Conceptualization, Project administration, Resources, Funding acquisition, Writing – review and editing.

Competing interests

The authors declare that they have no competing interests.

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