



# Mathematical modeling of the viscosity of tomato, broccoli and carrot purees under dynamic conditions



Evelina Tibäck<sup>a</sup>, Maud Langton<sup>b</sup>, Jorge Oliveira<sup>d</sup>, Lilia Ahrné<sup>a,c,\*</sup>

<sup>a</sup> SIK – The Swedish Institute of Food and Biotechnology, Box 5401, SE-402 29 Gothenburg, Sweden

<sup>b</sup> The Swedish University of Agricultural Sciences, Box 7051, SE-756 51 Uppsala, Sweden

<sup>c</sup> Chalmers University of Technology, Department of Chemical and Biological Engineering, 41296 Gothenburg, Sweden

<sup>d</sup> Department of Process & Chemical Engineering, University College Cork, Cork, Ireland

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## ABSTRACT

Different viscosity models were developed to describe the viscosity of unprocessed fruit and vegetable purees under dynamic conditions. Temperature hysteresis cycles were carried out for three purees with different structural characteristics (tomato, carrot, and broccoli), with heating and cooling phases from 10 to 80 °C with isothermal (holding) phases at 10, 30, 60 or 80 °C. The apparent viscosity was measured continuously with a rotational rheometer and the data was analyzed with time-independent and time-dependent models (quantifying rheopexy, thixotropy, or both). The results revealed clear thixotropic behavior in tomato puree, attributed to shearing effects, and rheopectic in broccoli puree, attributed to gel formation at the higher temperatures. Although carrot puree data from the isothermal periods could be quantified satisfactorily with no time dependency, analysis of the nonisothermal periods proved that rheopectic effects also needed to be included.

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## 1. Introduction

A fruit or vegetable puree is a dispersed suspension of plant cell materials in a fluid phase containing soluble pectins. The pectins in the fluid phase bind water and may form gels. Heat treatment induces pectin degradation as well as gel formation. Pectin degradation can be caused either by enzymatic activity,  $\beta$ -elimination, or acid hydrolysis. Heating a fruit or vegetable puree also induces leakage and solubilization of pectins from the cell walls into the fluid phase, which allows for gel formation. The pectin chains form gels through a three-dimensional network together with water, solutes and suspended particles. Consequently, thermal processing of fruit and vegetable purees can result in both pectin degradation and in gel formation (Duvetter et al., 2009).

A viscous material flows when a force is applied. For a Newtonian fluid, the viscosity remains constant at different shear rates. If the viscosity decreases with increasing shear rate it is said to be shear-thinning. A yield stress may be defined as the minimum stress required for initiating flow, and reflects the deformation and breakage of network structures in the fluid (Rao, 1977). Time-dependent, non-Newtonian fluids can be categorized as rheopectic or thixotropic. At constant

temperature and shear rate, the viscosity of a rheopectic fluid increases with time while the viscosity of a thixotropic fluid decreases with time. Thixotropic (Armelin et al., 2006; Labanda et al., 2004) and rheopectic (Teyssandier et al., 2009) effects are widely studied in polymer science, and food science has applied many of the concepts developed there for various foodstuffs (Abu-Jdayil, 2003; Dolz et al., 2007; Hernandez et al., 2008). The rheological properties of a fruit or vegetable puree are influenced by the raw material composition, particle concentration, hardness and morphology, particle size distribution, particle–particle interactions, soluble pectins and pectins on the particle surfaces, temperature and shear conditions (Barrett et al., 1998; Hayes et al., 1998; Lopez-Sanchez et al., 2011). Fruit and vegetable purees generally behave as weak gels and highly non-Newtonian liquids. Gelling of a fruit or vegetable puree implies a rheopectic behavior, while degradation of the structure in the puree causes a thixotropic behavior to take place. The most widely used method of analyzing and quantifying these time-dependent effects is to perform a so-called hysteresis cycle at constant temperature, increasing the shear rate of the measurement to some limit and then decreasing it back to the original condition (Abu-Jdayil et al., 2004). The area defined by the two curves is a measure of the hysteresis and hence of the extent of the rheopexy or thixotropy (Roussel, 2006). Time-dependent effects are modeled as a function of shear rate and time at constant temperature (Maingonnat et al., 2005). In this study, time-dependent effects in

\* Corresponding author at: SIK – The Swedish Institute of Food and Biotechnology, Box 5401, SE-402 29 Gothenburg, Sweden. Tel.: +46 10 516 6623; fax: +46 31 833782 (L. Ahrné).

E-mail address: [lilia.ahrne@sik.se](mailto:lilia.ahrne@sik.se) (L. Ahrné).

unprocessed fruit and vegetable purees were analyzed by performing a temperature hysteresis cycle instead of a shear rate cycle. Temperature and time have a large impact on the rheological behavior of fruit and vegetable purees, which in turn is important for pumping, stirring, and storage as well as for sensory properties.

The objective of this work was to develop a mathematical model to describe the viscosity of unprocessed fruit and vegetable purees under dynamic conditions, in order to account for the shearing and/or gelling time and temperature dependent effects. The general model was applied to carrot, broccoli, and tomato puree, and the results were interpreted on the basis of the structural differences between the products.

## 2. Materials and methods

### 2.1. Preparation of purees

Carrots, broccoli, and tomatoes were purchased from a local supermarket in Sweden.

Each vegetable was mixed with ionized water to facilitate the blending process (carrot/broccoli puree 40% vegetable and 60% water, tomato puree 70% fruit and 30% water). Four peeled carrots were used for each carrot puree. In the top and bottom end of each carrot 2 cm was cut off and discarded and the carrots were cut in 0.5 cm thick slices. Two broccoli heads were used for each broccoli puree. In the end of the stem 3 cm was cut off and discarded. The broccoli heads were cut lengthwise in half, one of the halves was cut into approximately  $2 \times 2 \times 2$  cm pieces, and 100 g of broccoli pieces were used from each broccoli head. In the tomato purees 4–5 tomatoes were used for each sample batch. The tomatoes were cut into 4 pieces each, and the seeds were removed. Vegetable/fruit pieces (carrot/broccoli: 200 g, tomato 120 g) were crushed together with cold deionized water (carrot/broccoli: 300 g, tomato 280 g) in a kitchen blender (Moulinex, Ecully, Cedex, France) for a set time (carrot/broccoli 5 min, tomato 3 min). After crushing each sample was immediately ultra-high-speed homogenized in an Ultra Turrax T25 Basic (IKA, Staufen, Germany, speed  $24,000 \text{ min}^{-1}$ ) for a set time (carrot/broccoli 3 min, tomato 2 min), while cooled on ice. After homogenization air was removed from the purees using a vacuum pump under stirring while cooled on ice (carrot 10 min, broccoli 30 + 30 min (samples were divided into two batches to facilitate air removal, then remixed), tomato 60 min). The samples were kept chilled in a refrigerator until measurement. The pH of each puree was measured before viscosity measurement. For each replicate a new batch of puree was made from scratch.

### 2.2. Rheological measurements

The rheological measurements were carried out in a stress-controlled rotational rheometer (Stresstech, Rheologica Instruments, Lund, Sweden) with a four-bladed vane geometry (diameter = 23 mm, height = 42 mm) and a serrated cup (diameter = 25 mm). The vane measurement system was calibrated against a concentric cylinder geometry (diameter = 25 mm) using a Newtonian calibration oil (Viscosity standard, Brookfield Engineering Laboratories, Massachusetts, USA) and two shear-thinning polymer solutions (Carbopol 981 NF; 0.25% in deionized water (pH 7.14) and 0.5% in deionized water (pH 7.00)). The cup was completely filled ( $37 \text{ cm}^3$ ) and a lid was used to minimize evaporative losses. A constant shear rate of  $50 \text{ s}^{-1}$  was applied and viscosity values were recorded every 10 s. The heating rate was  $2 \text{ }^\circ\text{C}/\text{min}$  and the cooling rate was  $1 \text{ }^\circ\text{C}/\text{min}$ . In order to have a controlled history of the samples prior to measurement, a pre-shear step at  $10 \text{ }^\circ\text{C}$  and  $50 \text{ s}^{-1}$  for 3 min was applied.

The purees were subjected to an isothermal period of 30 min at 4 different temperatures, 10, 30, 60 and  $80 \text{ }^\circ\text{C}$ . In addition, measurements were taken of the heating and cooling periods of the 3 latter cases, following the product history from  $10 \text{ }^\circ\text{C}$  up at a constant rate of  $2 \text{ }^\circ\text{C}/\text{min}$ , and after the isothermal period, down to  $10 \text{ }^\circ\text{C}$  again, at a constant rate of  $1 \text{ }^\circ\text{C}/\text{min}$ . This created a temperature hysteresis cycle for the 3 different temperatures (illustrated in Fig. 1). If there were no time-dependent effects, viscosity would remain constant during the isothermal period and would be the same when the sample goes back to  $10 \text{ }^\circ\text{C}$  as it was in the beginning. As the heating and cooling rates were not the same, heating and cooling times were different: 10, 25 or 35 min of heating where the temperature was increased to 30, 60 and  $80 \text{ }^\circ\text{C}$ , respectively, and 20, 50 or 70 min of cooling where the temperature was decreased back to  $10 \text{ }^\circ\text{C}$ . These times are the programmed values, but for the data regression the actual times when the programmed temperatures were reached were considered, in order to minimize the error from sample to sample variability due to heating and cooling times. It is also noted that there was a small overshoot of temperature experimentally when reaching the set-point, which becomes a source of experimental error, as the models assume that once the temperature reaches the isothermal set-point it immediately remains constant. The temperature cycles are shown in Fig. 1, with experimental data from typical cycles, and showing the programme at  $80 \text{ }^\circ\text{C}$  to help identify the overshoot.

### 2.3. Development of the viscosity models

Models were developed independently for 4 possible situations: time-independent, thixotropic, rheopetic and both thixotropic and rheopetic. However, for condensing the information, they can all be described by a generic model, with some parameters switching on or off the components associated to thixotropic and rheopetic behavior.

The most usual structured kinetic models are n-order type of models, with the rate of change proportional to a power n of the deviation to equilibrium (Barnes, 1997). Assuming a first order model for thixotropy, the rate is simply proportional to the deviation from equilibrium:

$$-\frac{d(\mu - \mu_\infty)}{dt} = k_t(\mu - \mu_\infty) \quad (1)$$

where  $k_t$  is the thixotropy rate constant and  $\mu_\infty$  the value of viscosity for  $t \rightarrow \infty$ . This is also known as the stretched exponential model (Maingonnat et al., 2005).

To describe rheopetic behavior, where the apparent viscosity is evolving from a low to a high limit viscosity, a sigmoidal model

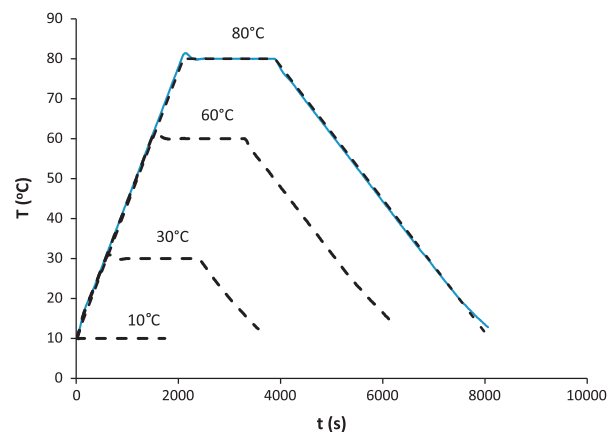


Fig. 1. Examples of experimental temperature cycles at  $10 \text{ }^\circ\text{C}$ ,  $30 \text{ }^\circ\text{C}$ ,  $60 \text{ }^\circ\text{C}$ , and  $80 \text{ }^\circ\text{C}$  and measured apparent viscosity for carrot, tomato and broccoli puree.

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