Numerical modeling of the rheological characteristic of olive paste under different conditioning treatments: traditional malaxation, high-frequency ultrasound and microwave

A. Tamborrino¹, P. Catalano², R. Romaniello³, B. Bianchi¹ and A. Leone¹

¹Department of Agricultural and Environmental Science, University of Bari Aldo Moro, Bari, Italy; ²Department of Agriculture, Environment and Food, University of Molise, Campobasso, Italy; ³Department of the Science of Agriculture, Food and Environment, University of Foggia, Foggia, Italy.

Abstract

Olive paste, a mixture of olive oil, vegetation water and solid particles, have a complex rheological behavior. Its viscosity (µ) cannot be considered as constant and depends on several parameters. The olive paste changes its rheological characteristics from the inlet to the outlet of the olive oil extraction line because of temperature increase and variation in fluid composition (i.e., solid-liquid m). A numerical analysis was carried out using different mathematical models to predict the apparent viscosity of olive paste as a function of the solids and olive oil volume fractions. Experimental trials were carried out processing the olive paste using different techniques: traditional malaxing (TM), the use of megasound (MS) and the use of microwaves (MW). The collected data consisted of apparent viscosity values, the related shear strain rates and the composition of the olive paste. These data were interpolated using a power law model whose parameters were determined by means of a linear regression in a bilogarithmic scale at each step of the olive milling process. As a result of comparison with the experimental data, the different models were found to be quite effective for describing the relative viscosity behavior and the obtained solid volume fraction obtained after the three different processing methods confirms the best behavior of the MS technique. As a final consideration, the results of this work represent another step toward full comprehension of the physical characteristics of the olive paste finalizes to improve the solid-liquid separation in olive oil centrifugal decanters.

Keywords: food processing, ultrasound applications; microwave application, olive paste, relative viscosity model

INTRODUCTION

The viscosity affects the velocity gradients and, therefore, the motion of suspended solid particles that must be removed by centrifugation. Based on this consideration, modeling the rheological behavior of olive paste is essential to investigate on the evolution of rheological properties of the olive paste in order to have more tools to use correct operation and parameters choose to optimize the decanter sedimentation (Boncinelli et al., 2013; Leone et al., 2013, 2015; Tamborrino et al., 2014a, 2015; Ayr et al., 2015; Squeo et al., 2017). The viscosity of olive paste has been measured under a range of extraction processing conditions in many experimental studies. The results show the strong dependence of the viscosity on the processing history undergone during process extraction (Ayadi et al., 2009; Tamborrino, 2014; Romaniello et al., 2017; Tamborrino et al., 2014a, 2019; Kalogianni et al., 2019). In the last decade microwave (MW) and ultrasound (US) are considered the novel olive paste processing technology and it has found wide experimental applications in the olive oil industry. It has been considered to introduce a change in the actual kneading phase that is considered a very crucial phase to improve the qualitative characteristics of the olive oil as well as the extraction efficiency of the actual process (Bejaoui et al., 2016, 2018; Leone et al., 2018a; Jiménez et al., 2007; Tamborrino et al., 2010 2019; Juliano et al., 2017; Leong et al.,



2015). Analyzing the results of many previous studies when both US and MW treatments are used, a significant decrease of apparent viscosity is observed. US and MW treatments result in a number of physicochemical changes in the olive paste matrix, in particular, on the solid/liquid fraction ratio and on the apparent viscosity (Leone et al., 2018b; Amarillo et al., 2019; Tamborrino et al., 2014b, c; Leone et al., 2017). Nowadays, the first consideration that led to think that these technologies could be conveniently used in the olive oil extraction process is related to the effects that they generate on the olive paste. (Ashokkumar, 2011; Chemat et al., 2017; Bejaoui et al., 2017; Caponio et al., 2018; Leone, et al., 2018b; Amarillo et al., 2019; Tamborrino et al., 2017).

In this article, experimental data concerning viscosity measurements carried out on olive paste treated with ultrasound waves and microwave are reported and analyzed to investigate the rheological behavior of olive paste with different treatments. The collected data consist of apparent viscosity values and the related shear rates. These data were interpolated using a power law model (Darby, 2001) whose parameters were determined by means of a linear regression in a bi-logarithmic scale at each step of the olive milling process. Moreover, further analysis was carried out using a specific mathematical model to predict the apparent viscosity of olive paste as a function of the solids volume fraction.

MATERIALS AND METHODS

Industrial olive mill and experimental equipment with MW continuous system and MS prototype

Trials were conducted with Coratina olives in an industrial olive oil mill using three different technologies to condition the olive paste. The industrial olive mill included a series of units, built by MORI-TEM s.r.l., Tavernelle Val di Pesa (FI, Italy) and represented by a leaf remover and washing machines group, a knives crusher, two vertical malaxer vessels with internal kneading blades connected to the top lid, and two phase solid-liquid horizontal centrifugal decanter. The industrial plant has a capacity of 750 kg h⁻¹; the olives were processed without water added to the decanter. The continuous MW system and MS vessel were connected in parallel with the two malaxers using two 3-way valves. The MW continuous system was built by a polypropylene tube of 65.4 mm of diameter and 2 m of length, in a reverberant chamber made by stainless steel wall. Several electromagnetic shields were placed on both ends of the pipe. The system was equipped with a generator attached to a magnetron and has a maximum power draw of 6.0 kW at 2.45 GHz. Besides, the generator head is connected to a power supply. A PLC was need to control in a continuous mode the output power of the system. The MW system was fabricated and assembled by Emitech s.r.l. (Corato, Italy). The MS vessel prototype, the first design for a large scale trials, is a stainless steel rectangular reactor with a 200 L of capacity. The reactor is equipped with a set of four transducers with frequencies 400 and 600 kHz and a power consuming of 2 kW. The transducers are positioned parallel to the opposite wall to create a standing sound wave field inside the reactor. The system is completed by a thermocouples placed at the inlet and the outlet of the reactor and a pressure gage at the inlet.

Experimental design

Experimental trials were carried out processing the olive paste by different techniques: traditional malaxing (TM), the use of megasound (MS) and the use of microwaves (MW). Data where acquired concerning viscosity measurements carried out on olive paste to validate the models. The collected data consist of apparent viscosity values, the related shear rates and the composition of the olive paste. These data were interpolated using a power law model according to the equations below, whose parameters were determined by means of a linear regression in a bi-logarithmic scale at each step of the olive milling process.

The treatments are summarize following:

- TM: Control treatment was conducted by malaxing 350 kg of olive paste for 30 min at 28°C as malaxing temperature;
- MW: Microwave treatment was conducted by passing in continuous way the olive

paste thought the MW continuous system, setting the temperature of 28°C;

 MS: Megasonic treatment was conducted by passing in continuous way thought the MS prototype with a registered temperature processing of about 28°C;

For each test a sample of olive paste was taken for the rheological measurements and for oil and moisture analysis.

Rheological measurements

Viscosity determinations were carried out using a rotational rheometer (model DV2-HBT Brookfield Engineering Laboratories, Inc., Stoughton, MA, USA) equipped with a disc spindle (model RV/HA/HB-4; Brookfield DVII + Brookfield Engineering Laboratories). Viscosity measurements were carried using 500 mL of olive paste, loaded into a 1000-mL glass container conditioned at 27°C in a thermostatic bath. Ten rotational speeds, ranged 0.5-100 rpm, were used to record the apparent viscosity. To interpret the experimental results in terms of viscosity, the torque-speed data and scale readings were converted into shear stressshear rate relationships using numerical conversion values as for Tamborrino et al. (2014a, b).

Oil and moisture analysis in olive paste

The total oil content was determined on 30 g of sample, previously dehydrated until reaching constant weight. Moisture removed from the sample was recorded. Oil in the dried sample was extracted with hexane in an automatic extractor (Randall 148, Velp Scientifica, Milan, Italy) following the analytical technique described by Cherubini et al. (2009). The sample was initially immersed directly in the boiling solvent at 139°C for 60 min. The sample was then subjected to washing at 139°C for 40 min; the sample container was removed from the solvent and reflux washed. Results were expressed as percentage of oil on wet and dry matter.

Power-law model

Viscosity directly affects velocity gradients and fluid flow fields as it describes the link between shear stress τ_{ij} and shear strain rate $\dot{\gamma}_{ij}$:

$$\dot{\gamma}_{ij} = \frac{\partial v_i}{\partial x_j} + \frac{\partial v_j}{\partial x_i} \tag{1}$$

where (v_1, v_2, v_3) describes fluid velocity field in (x_1, x_2, x_3) coordinate system.

If the ratio between shear stress and shear rate:

$$\mu(\dot{\gamma}_{ij}) = \frac{\tau_{ij}}{\dot{\gamma}_{ij}} \tag{2}$$

is constant the fluid behavior is called Newtonian (μ is the fluid dynamic viscosity).

Otherwise, if the above ratio is not constant, the fluid is called non-Newtonian and an empirical law is necessary to relate shear stress (or the viscosity that is called apparent viscosity: $\mu(\dot{\gamma}_{ij}) = \mu_{app}$ and it is independent on the shear plane *ij*) to shear rate. The power-law model can be used to calculate the apparent viscosity from the shear rate:

$$\mu_{app} = m \dot{\gamma}^{n-1} \tag{3}$$

where μ_{app} is the apparent viscosity, *n* is the flow behavior index and *m* is the consistency index.

Olive paste rheological model

As a result of comparison with the experimental data, different models were found (Zhang and Evans, 1989; Quemada, 1977; Yaghi, 2003; Arefinia and Shojaei, 2006) to be quite effective for describing the relative viscosity behavior and the obtained solid volume fraction



 φ_s , whose maximum value (maximum packing factor) is φ_{max} , defined in (Servais et al., 2002).

Typically, all models provide values for the non-dimensional relative viscosity

$$\mu_r = \frac{\mu_{app}}{\mu_0} \tag{4}$$

where μ_0 is a reference value given by the viscosity of the fluid with suspended solids removed. In particular, in Boncinelli et al. (2013) it was proved that the Zhang-Evans model

(Zhang and Evans, 1989) was the model best fitting the experimental data:

$$\mu_r = \left(\frac{1 - c\frac{\varphi_S}{\varphi_{max}}}{1 - \frac{\varphi_S}{\varphi_{max}}}\right)^2 \tag{5}$$

The model parameters *c* is determined fitting the model with experimental data.

RESULTS AND DISCUSSION

In Table 1 the characteristics of the olive paste used in the different processing conditions are shown.

Processing condition	Water content (%)	Oil content (%)	Solids content (%)	ϕ_{s} / ϕ_{max}
TM ₁	56.58	16.85	26.57	0385
MS ₁	55.77	19.47	24.76	0.358
MS ₂	62.87	17.09	20.04	0.288
MW ₁	55.76	16.50	27.74	0.404
MW ₂	56.20	17.19	26.61	0.386
MW ₃	56.76	16.41	26.83	0.390

Table 1. Olive paste physical characteristics.

TM – Traditional malaxer; MS – Megasound treatment; MW – Microwave treatment.

In Figure 1 the trend of the apparent viscosity is shown measured at different values of the shear rate.

As one can easily see all the lines are very close each other except the traditional malaxing: MS and MW affect viscosity in the same way giving quite low viscosity values, while TM leaves the viscosity quite higher than in the other processing conditions.

More insight gives the Zhang model applied using the measured values of the ratio $\frac{\varphi_S}{\varphi_{max}}$

between the solids volume fraction and the maximum packing factor. The same value for the constant c=1.3588 is used in Equation 5 for all the samples. In

Figure 2 the relation between measure and calculated values of the relative viscosity μ_r is shown. The model fits all the data with a quite high R^2 value as the same olives batch has been used during all the tests. Possible differences in olive paste composition can occur, so also in the solids volume to maximum packing factor ratio, and they are explained as sampling the olive paste during and/or after malaxing process is a quite difficult operation. In any case, the results are quite independent of this variability and the method used here allows also standardising the olive paste characteristics not simply on the base of the olives samples but on calculated and measured parameters.

Best model fitting points correspond to MW applications while for MS (and more for TM) the model gives a little worst approximation of the measured values. Moreover, MW samples behavior (c=1.3488 if only MW samples are used) is very near to (but slightly smaller than) the mean one (c=1.3588), MS (also very near the mean line but below the regression line in Figure 2) is characterized by a higher constant c=1.3600 and TM by a quite smaller

value c=1.3249 (the sample is above the regression line). Smaller values of c lead to higher values of the viscosity at equal values of the other parameters.

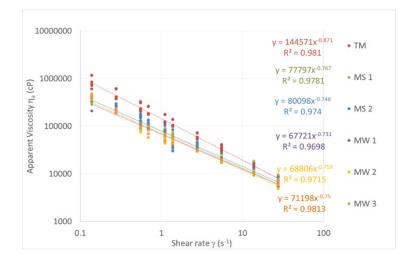
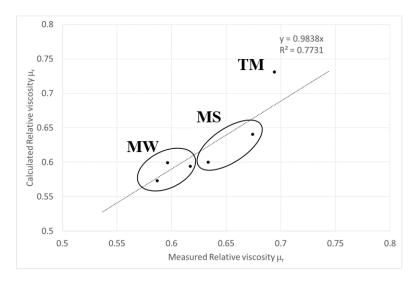
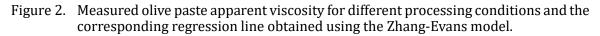


Figure 1. Measured olive paste apparent viscosity for different processing conditions and the corresponding regression lines (Power law model).





CONCLUSIONS

The model studied in this paper allows to standardize the comparison between differently treated olive paste through their standard characteristic parameters: μ_0 which is the viscosity of the olive paste with all solids removed (liquid phase) and the ratio φ_S/φ_{max} representing the packing status of the solids. Following the previous considerations, the non-treated paste is characterized (at equal values of the other parameters) by the highest value of the relative viscosity (smallest constant *c*), while MS treated paste by the smallest value (highest constant *c*), confirming that MW and MS allow significant reduction of the viscosity even MS behaves slightly better than MW.

ACKNOWLEDGEMENTS

The authors have contributed to the same extent to the present study. The authors are



grateful to Dr. Domenico Tarantino for technical assistance.

Literature cited

Amarillo, M., Pérez, N., Blasina, F., Gambaro, A., Leone, A., Romaniello, R., Xu, X.Q., and Juliano, P. (2019). Impact of sound attenuation on ultrasound-driven yield improvements during olive oil extraction. Ultrason Sonochem *53*, 142–151 https://doi.org/10.1016/j.ultsonch.2018.12.044. PubMed

Arefinia, R., and Shojaei, A. (2006). On the viscosity of composite suspensions of aluminum and ammonium perchlorate particles dispersed in hydroxyl terminated polybutadiene–New empirical model. J. Colloid Interface Sci. 299 (2), 962–971.

Ashokkumar, M. (2011). The characterization of acoustic cavitation bubbles - an overview. Ultrason Sonochem *18* (*4*), 864–872 https://doi.org/10.1016/j.ultsonch.2010.11.016. PubMed

Ayadi, M.A., Grati-Kamoun, N., and Attia, H. (2009). Physico-chemical change and heat stability of extra virgin olive oils flavoured by selected Tunisian aromatic plants. Food Chem Toxicol 47 (10), 2613–2619 https://doi.org/10.1016/j.fct.2009.07.024. PubMed

Ayr, U., Tamborrino, A., Catalano, P., Bianchi, B., and Leone, A. (2015). 3D computational fluid dynamics simulation and experimental validation for prediction of heat transfer in a new malaxer machine. J. Food Eng. *154*, 30–38 https://doi.org/10.1016/j.jfoodeng.2014.12.022.

Bejaoui, M.A., Beltran, G., Aguilera, M.P., and Jimenez, A. (2016). Continuous conditioning of olive paste by high power ultrasounds: response surface methodology to predict temperature and its effect on oil yield and virgin olive oil characteristics. Lebensm. Wiss. Technol. *69*, 175–184 https://doi.org/10.1016/j.lwt.2016.01.048.

Bejaoui, M.A., Sánchez-Ortiz, A., Sánchez, S., Jiménez, A., and Beltrán, G. (2017). The high power ultrasound frequency: effect on the virgin olive oil yield and quality. J. Food Eng. 2017, 10–17 https://doi.org/10.1016/j.jfoodeng.2017.03.013.

Bejaoui, M.A., Sánchez-Ortiz, A., Aguilera, M.P., Ruiz-Moreno, M.J., Sánchez, S., Jiménez, A., and Beltrán, G. (2018). High power ultrasound frequency for olive paste conditioning: effect on the virgin olive oil bioactive compounds and sensorial characteristics. Innov. Food Sci. Emerg. Technol. *47*, 136–145 https://doi.org/10.1016/j.ifset.2018.02.002.

Boncinelli, P., Catalano, P., and Cini, E. (. (2013). Olive paste rheological analysis. Trans. ASABE 56 (1), 237–243 https://doi.org/10.13031/2013.42574.

Caponio, F., Squeo, G., Brunetti, L., Pasqualone, A., Summo, C., Paradiso, V.M., Catalano, P., and Bianchi, B. (2018). Influence of the feed pipe position of an industrial scale two-phase decanter on extraction efficiency and chemical-sensory characteristics of virgin olive oil. J Sci Food Agric *98* (*11*), 4279–4286 https://doi.org/10.1002/jsfa.8950. PubMed

Chemat, F., Rombaut, N., Sicaire, A.-G., Meullemiestre, A., Fabiano-Tixier, A.-S., and Abert-Vian, M. (2017). Ultrasound assisted extraction of food and natural products. Mechanisms, techniques, combinations, protocols and applications. A review. Ultrason Sonochem *34*, 540–560 https://doi.org/10.1016/j.ultsonch.2016.06.035. PubMed

Cherubini, C., Migliorini, M., Mugelli, M., Viti, P., Berti, A., Cini, E., et al. (2009). Towards a technological ripening index for olive oil fruits. Journal Science of Food Agriculture, 89, 671e682.

Darby, R. (2001). Chemical Engineering Fluid Mechanics (New York, N.Y.: Marcel Dekker).

Jiménez, A., Beltrán, G., and Uceda, M. (2007). High-power ultrasound in olive paste pretreatment. Effect on process yield and virgin olive oil characteristics. Ultrason Sonochem *14* (6), 725–731 https://doi.org/10.1016/j.ultsonch.2006.12.006. PubMed

Juliano, P., Bainczyk, F., Swiergon, P., Supriyatna, M.I.M., Guillaume, C., Ravetti, L., Canamasas, P., Cravotto, G., and Xu, X.Q. (2017). Extraction of olive oil assisted by high-frequency ultrasound standing waves. Ultrason Sonochem *38*, 104–114 https://doi.org/10.1016/j.ultsonch.2017.02.038. PubMed

Kalogianni, E.P., Georgiou, D., and Exarhopoulos, S. (2019). Olive oil droplet coalescence during malaxation. J. Food Eng. 240, 99–104.

Leone, A., Romaniello, R., and Tamborrino, A. (2013). Development of a prototype for extra-virgin olive oil storage with online control of injected nitrogen. Trans. ASABE *56* (*3*), 1017–1024.

Leone, A., Romaniello, R., Zagaria, R., and Tamborrino, A. (2015). Mathematical modelling of the performance parameters of a new decanter centrifuge generation. J. Food Eng. *166*, 166 https://doi.org/10.1016/j.jfoodeng.2015.05.011.

Leone, A., Romaniello, R., Tamborrino, A., Xu, X., and Juliano, P. (2017). Microwave and megasonics combined technology for a continuous olive oil process with enhanced extractability. Innov. Food Sci. Emerg. Technol. 42, 56–

63 https://doi.org/10.1016/j.ifset.2017.06.001.

Leone, A., Romaniello, R., Juliano, P., and Tamborrino, A. (2018a). Use of a mixing-coil heat exchanger combined with microwave and ultrasound technology in an olive oil extraction process. Innov. Food Sci. Emerg. Technol. *50*, 66–72 https://doi.org/10.1016/j.ifset.2018.09.005.

Leone, A., Romaniello, R., Tamborrino, A., Urbani, S., Servili, M., Amarillo, M., and Juliano, P. (2018b). Application of microwaves and megasound to olive paste in an industrial olive oil extraction plant: impact on virgin olive oil quality and composition. Eur. J. Lipid Sci. Technol. *120* (1), 1700261 https://doi.org/10.1002/ejlt.201700261.

Leong, T., Knoerzer, K., Trujillo, F.J., Johansson, L., Manasseh, R., Barbosa-Canovas, G.V., and Juliano, P. (2015). Megasonic separation of food droplets and particles: design considerations. Food Eng. Rev. 7 (3), 298–320 https://doi.org/10.1007/s12393-015-9112-4.

Quemada, D. (1977). Rheology of concentrated disperse systems and minimum energy dissipation principle. Rheol. Acta *16*, 82–94.

Romaniello, R., Leone, A., and Tamborrino, A. (2017). Specification of a new de-stoner machine: evaluation of machining effects on olive paste's rheology and olive oil yield and quality. J Sci Food Agric 97 (1), 115–121 https://doi.org/10.1002/jsfa.7694. PubMed

Servais, C., Jones, R., and Roberts, I. (2002). The influence of particle size distribution on the processing of food. J. Food Eng. *51* (*3*), 201–208 https://doi.org/10.1016/S0260-8774(01)00056-5.

Squeo, G., Tamborrino, A., Pasqualone, A., Leone, A., Paradiso, V.M., Summo, C., and Caponio, F. (2017). Assessment of the influence of the decanter set-up during continuous processing of olives at different pigmentation index. Food Bioprocess Technol. *10* (*3*), 592–602 https://doi.org/10.1007/s11947-016-1842-7.

Tamborrino, A. (2014). Olive paste malaxation. The extra-virgin olive oil handbook, p.127–137.

Tamborrino, A., Clodoveo, M.L., Leone, A., Amirante, P., and Paice, A.G. (2010). The malaxation process: Influence on olive oil quality and the effect of the control of oxygen concentration in virgin olive oil. Olives and Olive Oil in Health and Disease Prevention, p.77–83.

Tamborrino, A., Catalano, C., and Leone, A. (2014a). Using an in-line rotating torque transducer to study the rheological aspects of malaxed olive paste. J. Food Eng. *126*, 65–71 https://doi.org/10.1016/j.jfoodeng.2013.09.024.

Tamborrino, A., Pati, S., Romaniello, R., Quinto, M., Zagaria, R., and Leone, A. (2014b). Design and implementation of an automatically controlled malaxer pilot plant equipped with an in-line oxygen injection system into the olive paste. J. Food Eng. 141, 1–12 https://doi.org/10.1016/j.jfoodeng.2014.05.002.

Tamborrino, A., Romaniello, R., Zagaria, R., and Leone, A. (2014c). Microwave-assisted treatment for continuous olive paste conditioning: impact on olive oil quality and yield. Biosyst. Eng. *127*, 92–102 https://doi.org/10.1016/j.biosystemseng.2014.08.015.

Tamborrino, A., Leone, A., Romaniello, R., Catalano, P., and Bianchi, B. (2015). Comparative experiments to assess the performance of an innovative horizontal centrifuge working in a continuous olive oil plant. Biosyst. Eng. *129*, 160–168 https://doi.org/10.1016/j.biosystemseng.2014.10.005.

Tamborrino, A., Romaniello, R., Caponio, F., Squeo, G., and Leone, A. (2019). Combined industrial olive oil extraction plant using ultrasounds, microwave, and heat exchange: impact on olive oil quality and yield. J. Food Eng. 245, 124–130 https://doi.org/10.1016/j.jfoodeng.2018.10.019.

Yaghi, B. (2003). Rheology of oil-in-water emulsions containing fine particles. Petroleum Sci. and Eng. 40 (3–4), 103–110 https://doi.org/10.1016/S0920-4105(03)00106-2.

Zhang, T., and Evans, J.R.G. (1989). Predicting the viscosity of ceramic injection moulding suspensions. J. Eur. Ceram. Soc. 5 (3), 165–172 https://doi.org/10.1016/0955-2219(89)90032-0.

