Chemical and Process Engineering: New Frontiers, 2023, 44(3), e34
DOI: 10.24425/cpe.2023.146736

POSTER COMMUNICATION

Experimentation and configuring continuous ultrasound assisted operation of extraction pilot plant

Tuomas Koiranen[1]*, Jussi Tamminen[2], Dmitry Gradov[1]

[1] Lappeenranta-Lahti University of Technology, Department of Separation Science, Yliopiston katu 34, 53851 Lappeenranta, Finland
[2] Satakunta University of Applied Sciences, Satakunnankatu 23 28130 Pori, Finland

*** Corresponding author:**
e-mail:
tuomas.koiranen@lut.fi

Presented at 8th European Process Intensification Conference, 31.05–2.06.2023, Warsaw, Poland.

Article info:
Received: 31 July 2023
Revised: 20 August 2023
Accepted: 12 September 2023

Abstract
In this study a continuous pilot plant configuration for helical flow type Ultrasound Assisted Extraction (UAE) reactor is described and carotenoid extraction results from a carrot-water extraction system are presented. The pilot plant configuration consists of all necessary unit operations from washed raw material to extracted natural ingredient product. According to our best knowledge, the published data from the whole processing line of continuous UAE has not been described to date. One of the subjects in this study has been patented as an in-house UAE design which enables to prolong extractable solid material residence time under ultrasound field. Process operation was verified with several hours of pilot tests. Extraction results have been presented in case of carotenoids from fresh carrots and strawberry extract from frozen strawberries. The extracted carotenoid concentration was improved by 40% compared to macerated carrot results, and absorbance ratio of strawberry extract increased by a factor of 5.

Keywords
ultrasound extraction, continuous process, natural ingredients, pilot process

1. INTRODUCTION

Many process intensified extraction studies using ultrasound have been published for the last two decades but only few detailed process configuration studies can be found from continuous large scale processes. The reactor configurations have been circulating loop reactors or flow-through types; solid-liquid separation methods have been decanters, plate-frame filters or membrane filters (Alexandru et al., 2013; Eom et al., 2020; Grillo et al., 2020). Khadhraoui et al. (2021) reviewed hybrid techniques with UAE. The proposed solutions were based on pressure extruding, where filter cake was mixed with a solvent before ultrasound extraction for the rest of the cake. The total process configurations was not reported. Several scientific peer-reviewed journal articles on continuous UAE processes described pilot scale equipment and ultrasound effects but the whole process descriptions from raw materials to extracted products are not available (Clodoveo, 2017; Dorosh et al., 2020; Geow et al., 2021; Rodríguez et al., 2022). However, process descriptions of full-scale continuous UAE process for virgin oil extraction using flow-through ultrasound based extraction reactor were reviewed by Clodoveo et al. (2017) and by Amirante and Clodoveo (2017). The sonication residence times were less than 10 minutes due to installation of US probes directly to main pipe flow. Martinez-Guerra and Gude (2015) studied continuous UAE process for vegetable oil transesterification process. The process was described in more detail from oil feed to biodiesel product although oil extraction was not included in the description. In a patent survey including the keyword "ultrasound assisted extraction" between 2009-2022, 70 relevant patents were found related mainly to specific medicinal, pigment, oil and phenolic compounds separation and sonoreactor inventions. Durkacz and Cullen (2019) patented process alternatives as block diagram level for extraction of cannabis oil in which the main sequential unit operations were: raw material washing, shredding/grinding, solvent mixing, pumping, ultrasonication, filtering, solvent recovery and product separation. In another patent, Gu and Du (2016) presented their invention of combining US probes into a conveyor system, and reported an example of continuous processes at unit operation level.

In our previous work (Tamminen et al., 2022a), flow-through ultrasound assisted extraction (UAE) of chlorophylls and carotenoids was presented. A continuous flow-through reactor improved significantly chlorophyll and carotenoid yields compared to just mixing solvent extraction conditions. The novel reactor geometry was described by Tamminen et al. (2022b). The patented configuration consists of an ultrasound source and a helical flow channel inside the reactor including mixing elements which basically aid radial mixing in the reactor.

In this study, a pilot plant configuration for helical flow type UAE reactor is described and carotenoid extraction results from carrot-water extraction system are presented.

2. METHODS

The pilot plant configuration is presented in Fig. 1. The process configuration (see Table 1) consists of feed solvent tank,

Table 1. Main equipment in pilot configuration.

Equipment	Type and main specifications
Biomass shredder	Aple press ESE-055: motor nominal power 0.55 kW, blade opening 28 × 8.5 mm, rotation speed 1350 rpm
High-shear (HS) mixer	Silverson AX-5: motor nominal power 0.75 kW, max rotation speed 6000 rpm, max tip speed 16 m/s, Duplex mixer diameter 56.1 mm
Hose pump	LPP-D15: max. pressure 16 bar, max flow rate 2.5 l/min
Ultrasound extractor	Weber ultrasonics Sonopush mono: max. nominal power 2000 W, 25 kHz frequency, LUT in-house design reactor vessel, ultrasound sound dampening housing (60–68 dB)
Vibrating sieve	Virto VPB 450: Sieve size 0.15 mm, motor power 0.21 kW
Screw-press	Angel 20K-GS: motor nominal power 0.4 kW, max. screw speed 1450 rpm, 2 × 33.5 cm length screws
Auxiliary equipment	Lauda WK 4600 thermostat for temperature control, pressurized air operated AKO VMC 25 hose valve for pressure control at US reactor unit

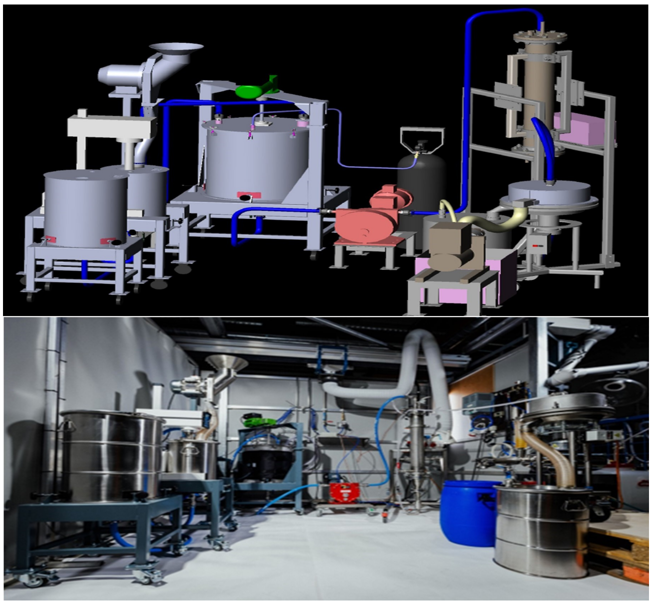

Figure 1. On-site extraction production line. Processing capacity of pulp 42 L/h. Power consumption 9 kWh, US power 2 kW, Solid-to-liquid separation efficiency 90%, nominal batch size 120 L. Flexibility in configuring equipment owing to hose pipe connections.

biomass shredder, high-shear (HS) mixer, hose pump, ultrasound (US) extractor, vibrating sieve for pre-treatment of solid-liquid removal, and screw-press for final mother liquor removal. The equipment configuration is flexible due to the camlock hose connections. For this specific reactor configuration solids tend to settle on the helical blade, and therefore intermittent fluid pressure variation is used to flush solids which is beneficial for increasing solid residence time while fluid residence time is kept constant. Carotenoid extraction analytics is based on UV/VIS spectrometer (Agilent Cary 8454 UV-Vis), and is described by Tamminen et al. (2022a), Pérez-Gálvez (2020), and Lichtenthaler and Buschmann (2001). Particle sizes were analyzed using Malvern Mastersizer 3000.

3. RESULTS

Pilot-scale extraction tests were performed for carrots as a model compound to extract total carotenoids. Process conditions are collected in Table 2 and in Fig. 2. In addition to carotenoid extraction from carrot-water feed, the pilot has also been successfully tested for production of strawberry extract in 50 vol-% ethanol at 23±1°C, see Table 3 and Fig. 3.

Table 2. Process conditions of carotenoid extraction from carrots.

Process parameters	Process values
Reactor residence time	10 minutes
Extraction temperature	31 ± 2°C
Raw material-to-solvent volume ratio	0.15
US power setting	1500 W, US power for 2–4 minutes and 1 minute power off
Total pilot run-time	180 minutes

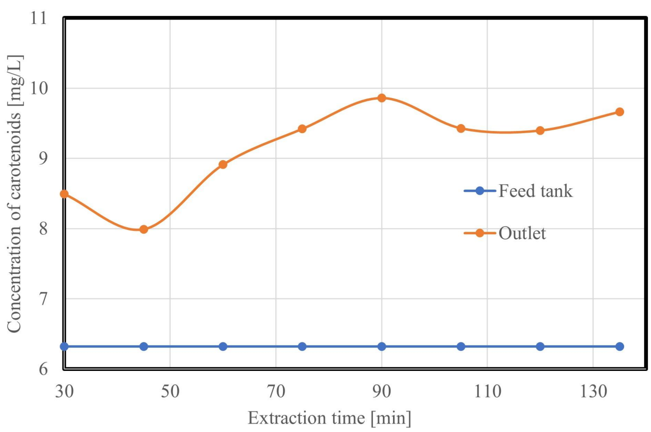

Figure 2. Carrot extraction results, product concentration as average of mother liquor outlets from vibrating sieve and screw press.

Table 3. Strawberry extract in 50 vol-% ethanol. Total pilot run-time 120 minutes.

Process parameters	Process values
Reactor residence time	10 minutes
US Power setting	2000 W continuously without pulses
Raw material-to-solvent volume ratio	0.8

Extraction results (Fig. 2) show a significant increase in total carotenoid concentration 8.5–9.7 mg/l compared to feed tank 6.3 mg/l. The total carotenoid concentration in carrots was between 14.9–40 mg/l with R/S-ratio 0.15 and data from Khoo et al. (2011). The solid particle size D90 was 1.4–1.6 mm after shredding and HS-mixing. The overall extraction intensification is due to high shear wet milling and ultrasound reactor effect. Reactor pressure variation cycles with AKO VMC 25 valve from 1 bar to 2.5–3 bar enables solid collection to the reactor and flushing from the reactor during operation. Solid flushing cycle in experiments was 20 minutes, and solid collecting time to the reactor was 35 minutes. In this manner, ultrasound treatment time to extractable solids was prolonged compared to solid suspension residence time in reactor zone. However, the carotenoid concentration in screw press filtrate was 13.7–15.8 mg/L which still leaves room for R/S-ratio optimization as vibrating sieve mother liquor flow could be decreased. In addition to carotenoid extraction from carrot-water feed, the pilot has also been successfully used for production of strawberry extracts from frozen strawberries to 50 vol-% ethanol at 23 ± 1°C, see Fig. 3. Substantial extraction improvement was based on UV/Vis absorbance with Pilot in comparison to macerated extraction. Particle size in solid outlet was 1.4–1.6 mm (strawberries).

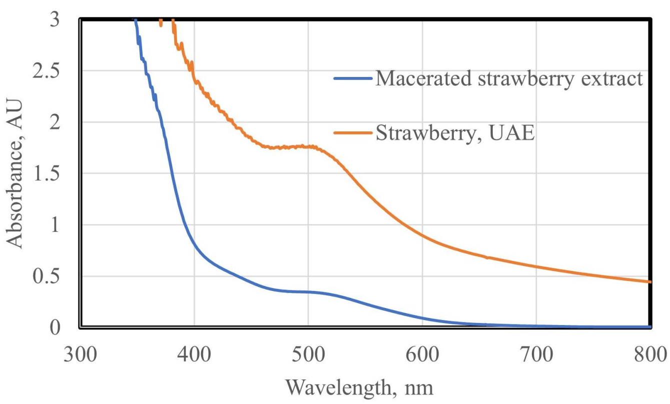

Figure 3. UV/Vis absorbances of strawberry extract in 50 vol-% ethanol and macerated extract. Absorbance-ratio: UAE Strawberry extract / Macerated Strawberry extract = 5.

4. CONCLUSIONS

The presented pilot plant has been developed for extraction intensification where a combination of high-shear mixing, ultrasound processing and screw pressing takes place. The patented ultrasound device also allows the prolongation of ultrasound time for extracted solids.

ACKNOWLEDGEMENTS

The Authors wish to thank Business Finland for financial support NATINREC – project (1195/31/2019).

REFERENCES

Alexandru L., Cravotto G., Giordana L., Binello A., Chemat F., 2013. Ultrasound-assisted extraction of clove buds using batch- and flow-reactors: A comparative study on a pilot scale. *Innovative Food Sci. Emerg. Technol.*, 20, 167–172. DOI: 10.1016/j.ifset.2013.07.011.

Amirante R., Clodoveo M.L., 2017. Developments in the design and construction of continuous full-scale ultrasonic devices for the EVOO industry. *Eur. J. Lipid Sci. Technol.*, 119, 1600438. DOI: 10.1002/ejlt.201600438.

Clodoveo M.L., Moramarco V., Paduano A., Sacchi R., Di Palmo T., Crupi P., Corbo F., Pesce V., Distaso E., Tamburrano P., Amirante R., 2017. Engineering design and prototype development of a full scale ultrasound system for virgin olive oil by means of numerical and experimental analysis. *Ultrason. Sonochem.*, 37, 169–181. DOI: 10.1016/j.ultsonch.2017.01.004.

Dorosh O., Moreira M.M., Rodrigues F., Peixoto A.F, Freire C., Morais S., Delerue-Matos C., 2020. Vine-canes valorisation: Ultrasound-assisted extraction from lab to pilot scale. *Molecules*, 25, 1739. DOI: 10.3390/molecules25071739.

Durkacz A.J., Cullen M.T., 2019. *Method for extracting compositions from plants*. U.S. Patent No. US10851077B2.

Eom S.J., Kim Y.E., Kim J.E., Park J., Kim Y.H., Song K.M., Lee N.M., 2020. Production of *Undaria pinnatifida* sporophyll extract using pilot-scale ultrasound-assisted extraction: Extract characteristics and antioxidant and anti-inflammatory activities. *Algal Res.*, 51, 102039. DOI: 10.1016/j.algal.2020.102039.

Geow C.H., Tan M.C., Yeap S.P., Chin N.L., 2021. A review on extraction techniques and its future appilcations in industry. *Eur. J. Lipid Sci. Technol.*, 123, 200302. DOI: 10.1002/ejlt.202000302.

Grillo G., Boffa L., Talarico S., Solarino R., Binello A., 2020. Batch and flow ultrasound-assisted extraction of grape stalks: Process intensification design up to a multi-kilo scale. *Antioxidants*, 9, 730. DOI: 10.3390/antiox9080730.

Gu J., Du J., 2016. *System and method for continuous extraction of material*. U.S. Patent No. US9333441B2.

Khadhraoui B., Ummat V., Tiwari B.K., Fabiano-Tixier A.S., Chemat F., 2021. Review of ultrasound combinations with hybrid and innovative techniques for extraction and processing of food and natural products. *Ultrason. Sonochem.*, 76, 105625. DOI: 10.1016/j.ultsonch.2021.105625.

Khoo H.-E., Prasad K.N., Kong K.-W., Jiang Y., Ismail A., 2011. Carotenoids and their isomers: Color Pigments in fruits and vegetables. *Molecules*, 16, 1710–1738. DOI: 10.3390/molecules16021710.

Lichtenthaler C. Buschmann C., 2001. Chlorophylls and carotenoids: Measurement and characterization by UV-VIS spectroscopy. *Curr. Protocol. Food Anal. Chem.*, 1, F4.3.1–F4.3.8. DOI: 10.1002/0471142913.faf0403s01.

Martinez-Guerra E., Gude V.G., 2015. Continuous and pulse sonication effects on transesterification of used vegetable oil. *Energy Convers. Manage.*, 96, 268–276. DOI: 10.1016/j.enconman.2015.02.073.

Pérez-Gálvez A., Viera I., Roca M., 2020. Carotenoids and chlorophylls as antioxidants. *Antioxidants*, 9, 505. DOI: 10.3390/antiox9060505.

Rodríguez Ó., Bona S., Stäbler A., Rodríguez-Turienzo L., 2022. Ultrasound-assisted extraction of polyphenols from olive pomace: Scale up from laboratory to pilot scenario. *Processes*, 10, 2481. DOI: 10.3390/pr10122481.

Tamminen J., Holappa J., Gradov D.V., Koiranen T., 2022a. Scaling up continuous ultrasound-assisted extractor for plant extracts by using spinach leaves as a test material. *Ultrason. Sonochem.*, 90, 106171. DOI: 10.1016/j.ultsonch.2022.106171.

Tamminen J., Varis J., Koiranen T., Gradov D., 2022b. *An ultrasound processing device*. Patent No. FI20205291A1.

Lappeenranta-Lahti University of Technology / LUT university, Finland

Experimentation and configuring continuous ultrasound assisted extraction pilot plant operation

Tuomas Koiranen*[a], Jussi Tamminen[b], Dmitry Gradov[a]
[a] Department of Separation Science, LUT School of Engineering Sciences, LUT University, P. O. Box 20, FIN-53851 Lappeenranta, Finland.
[b]Satakunta University of Applied Sciences, Pori, Finland
*tuomas.koiranen@lut.fi

Introduction

Large amount of process intensified extraction studies using ultrasound have been published during last two decades but only few detailed process configuration studies can be found from continuous large scale processes [1, 2, 3]. The reactor configurations have been circulating loop reactors or flow-through types; solid-liquid separation methods have been decanters, plate-frame filters or membrane filters.
In our previous work [4] flow-through ultrasound assisted extraction (UAE) of chlorophylls and carotenoids was presented. Continuous flow-through reactor improved significantly chlorophyll and carotenoid yields compared to just mixing solvent extraction conditions. The reactor geometry is described in [5]. Patented configuration consists of ultrasound source and helical flow channel inside reactor including mixing elements which basically aid radial mixing in the reactor.
In this study pilot plant configuration for helical flow type UAE reactor is described, and carotenoid extraction results from carrot-water extraction system are presented.

Methods

Pilot plant configuration is presented in Fig. 1. The process configuration consists of feed solvent tank, biomass shredder, high-shear (HS) mixer, hose pump, ultrasound (US) extractor, vibrating sieve for pre-treatment of solid-liquid removal, and screw-press for final mother liquor removal. The equipment configuration is flexible due to the camlock hose connections. For this specific reactor configuration the solids tend to settle on the helical blade, and therefore intermittent fluid pressure variation is used to flush solids which is beneficial for increasing solids residence time while fluid residence time is kept constant.

Carotenoid extraction analytics is based on UV/VIS spectrometer, and is described in more detail by Tamminen et al. [4], Particle sizes were analyzed using Malvern Mastersizer 3000. The process conditions are described in Table 1.

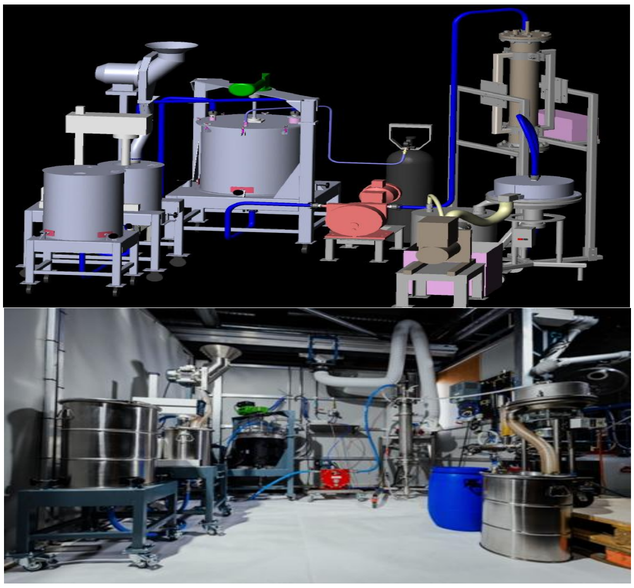

Figure 1. On-site extraction production line. Processing capacity of pulp is 42 L/h, Power consumption 9 kWh, US power 2 kW, S/L separation efficiency 90%, Batch size 120 L. Flexibility in configuring equipment due to hose connections.

Table 1. Pilot Hall Test (PHT) conditions.

PHT TEST	PROCESS CONDITIONS
Reactor residence time:	10 min.
Extraction temperature:	31±2 °C
Raw material shredded and wet-milled with HS-mixer.	
Raw material / Solvent volume-ratio (R/S):	0.15
US nominal power:	1500 W/L
Total run-time:	180 min.

CONTACT
Professor Tuomas Koiranen
LUT University, Finland.
Tuomas.Koiranen@lut.fi
+358 50 4357414

Results and discussion

Extraction results (Fig. 2) show significant increase in product flow (mother liquor streams from vibrating sieve and from screw press) carotenoid concentration 8.5-9.7 mg/L compared to feed tank 6.3 mg/L. The average total carotenoid concentration in carrots is 18.6 mg/L. The solids particle size D90 was 1.4-1.6 mm after shredding and HS-mixing. The overall extraction intensification is due to high shear wet milling and ultrasound reactor effect. It is noted that reactor pressure variation cycles from 1 bar to 2.5-3 bar enables solids flushing from reactor during operation. Solids flushing cycle in experiments was 20 minutes, and solids collecting time to reactor was 35 minutes. The carotenoid concentration in screw press filtrate was 13.7-15.8 mg/L which still leaves room for R/S-ratio optimization as vibrating sieve mother liquor stream could be decreased.
In addition to carotenoid extraction from carrot-water feed, the pilot has also been successfully used for production of strawberry and herb mixture extracts to 50 vol-% and 60 vol-% ethanol at 23±1 C°, see Fig. 3. Substantial extraction improvement was found based on UV/Vis absorbance with Pilot in comparison to macerated extraction. Particle size in solids outlet was 1.4-1.6 mm (strawberries), and 2-3 mm (herbs).

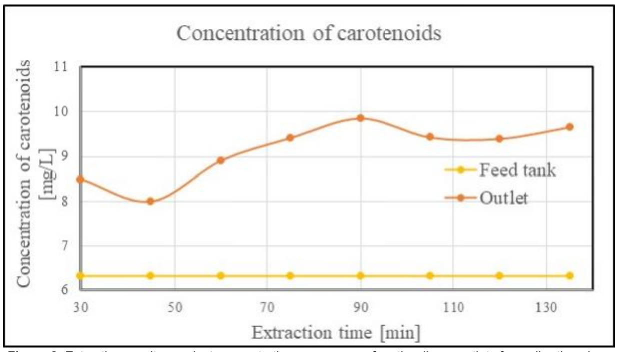

Figure 2. Extraction results, product concentration as average of mother liquor outlets from vibrating sieve and screw press.

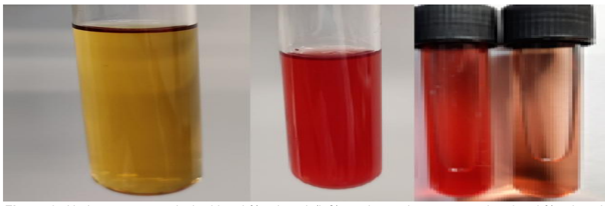

Figure 3. Herb extract sample in 60 vol-% ethanol (left), and strawberry extract in 50 vol-% ethanol (middle), Pilot strawberry extract comparison to macerated strawberry extract (right). Absorbance-ratio: UAE Strawberry extract / Macerated Strawberry extract = 5.

Conclusions

The presented pilot plant has been developed for extraction intensification where combination of high-shear mixing, ultrasound processing and screw pressing takes place. The patented ultrasound device also allows the prolongation of ultrasound time for extracted solids.
The Authors wish to thank Business Finland for financial support NATINREC -project (1195/31/2019).

References

[1] Eom, S.J., Kim, Y.E., Kim, J-E. et al, Production of Undaria pinnatifida sporophyll extract using pilot-scale ultrasound-assisted extraction: Extract characteristics and antioxidant and anti-inflammatory activities, Algal Research 51, (2020), 102039
[2] Grillo, G., Boffa, L., Talarico S. et al., Batch and Flow Ultrasound-Assisted Extraction of Grape Stalks: Process Intensification Design up to a Multi-Kilo Scale, Antioxidants 9, (2020), 730.
[3] Alexandru, L., Cravotto, G., Giordana, L., Binello, A., Chemat, F., Ultrasound-assisted extraction of clove buds using batch- and flow-reactors: A comparative study on a pilot scale, Innovative Food Science and Emerging Technologies 20, (2013), 167–172.
[4] Tamminen, J., Holappa, J., Gradov, D.V., Koiranen, T., Scaling up continuous ultrasound-assisted extractor for plant extracts by using spinach leaves as a test material, Ultrasonics Sonochemistry 90, (2022) 106171.
[5]An ultrasound processing device, Tamminen, J., Varis, J., Koiranen, T., Gradov, D., FI 130028B, patent granted 30.12.2022.

Lappeenranta-Lahti University of Technology