Available online at www.sciencedirect.com
SciVerse ScienceDirect
Procedia Engineering 44 (2012) 1736 – 1738
Procedia Engineering
www.elsevier.com/locate/procedia

Euromembrane Conference 2012

[P3.032]

Towards a membrane process based parth to concentrate willow extract

K.V. Christensen*[1], M.L. Ohm[1], V.G. Horn[2]
[1]*University of Southern Denmark, Denmark,* [2]*Palsgaard A/S, Denmark*

Introduction

Extracts from willow have been known as an analgesic, antipyretic and anti-inflammatory drug for over 3000 years. Since the 1820s the main therapeutically effect has been associated with the component salicine isolated from the extract and from 1899 sold in the form of salicylic acid as Aspirin. Aspirin belongs to the group of NonSteroid Antiinflamatory Drugs (NSAID) sold around most of the Globe as counter drug requiring no medical prescription. Aspirin is one of the most sold NSAID world-wide and has shown to be a cheap and effective pain reliever and anti-inflammatory agent. Unfortunatedly it has also been shown to have unwanted side effects. It is suspect of causing bleeding and perforation of the gastrointestinal system and it is estimated that around 7600 deaths and 76000 hospitalisations in the USA alone can be attributed to NSAID intake [1,2]. Therefore an alternative with less side effects is desirable.

The obvious starting point is to go back to the original extract from willow (*salix* spp.). Apart from salicine and salicine derivatives, the extract contains flavonoids and condensed tannins [3].

Experiments carried out with willow extracts show that the extracts have higher analgesic and anti-inflammatory effects than would be expected if only salicine would be responsible for these effects [2], [4]. At the same time the side effects associated with Aspirin was considerable reduced. This indicates that willow extract in itself has potential as a future NSAID, but also that synergetic effects between the chemical constituents of the extract is important. Thus when producing a potential NSAID from willow extract recovering most if not all the extracted components is of importance.

This paper describes the use of a combination of microfiltration (MF), ultrafiltration (UF) and reverse osmosis (RO) as part of a path to produce willow extract concentrates.

Experimental and Methods

Aqueous willow extract was delivered from Ny Vraa Bioenergi I/S. Prior to MF the willow extract was passed through a 45 µm sieve to remove major solids like leaves and stems.

For the MF, UF and RO the experimental setup consisted of a LabStak® M20 DSS from Alfa Laval with appropriate external cooling system to keep the willow extract temperature constant around 10°C to reduce the risk of microbial activity. The purpose of the MF is to remove minor fouling components and reduce potential microbial hazards. For this purpose a FSMO,45pp (AlfaLaval) membrane with a mean pore size of 0,45 µm was chosen. The total filtration area being 0.216 m^2. The UF should remove virus and endospores before the RO-concentration. An Alfa-Laval-GR81PP membrane with a MWCO of 10000 was chosen for this operation as none of the known beneficial flavonoids, tannins and salicine derivatives has a molecular size close to this MWCO. The total UF-filtration area was 0.288 m^2. The RO concentration was performed using an Alfa-Laval RO98pHt membrane the total membrane area being 0.288 m^2.

As an indicator for the content of salicine derivatives saligenine was chosen and for flavonoids epicatechin was tried. Both was analysed using HPLC with an UV detector.

doi:10.1016/j.proeng.2012.08.929

Results and discussion

The driving force for MF, UF and RO is the total pressure difference modified by the osmotic pressure difference between retentate and permeate. Thus the flux in all cases can be related to an expression of the form:

$$J_{flux} = k_m \cdot (\Delta P - \Delta \Pi)$$

where k_m is an experimentally determined mass transfer coefficient while ΔP and ΔΠ is the transmembrane pressure and osmotic pressure difference between retentate and permeate respectively. In the present case though only for RO is the osmotic pressure difference of importance.

The MF was performed with a transmembrane pressure of 2 bar. The MF produced 400 litre of willow extract permeate. During the filtration severe membrane fouling was observed leading to a permanent flux decline from 60 LMH to 15 LMH over time even with an intermediate CIP with hot water, a pH 10 NaOH solution followed by a pH 3 citric acid solution and finally hot water.

The UF process following the MF was performed at a transmembrane pressure of 7.2 bar. During the UF an initial permanent flux decline from 13 LMH to 6 LMH was observed. After CIP a pure water flux of 60% of the clean membrane flux could be re-established. During the remaining UF experiment no further permanent flux decline was observed, the flux between CIP's being between 8 and 5 LMH. A total of 214 litres willow extract was processed leading to 212 litres of permeat.

The final RO concentration was carried out at a transmembrane pressure of 34 bar. The willow extract total drymatter content (DM) could be increased from 0.4% to a maximum of 5% at which the permeate flux becomes close to zero as seen from figure 1. Further the DM and the saligenine concentration follow each other closely indicating little or no loss in the content of essential constituents in the willow extract concentrate (se figure 2).

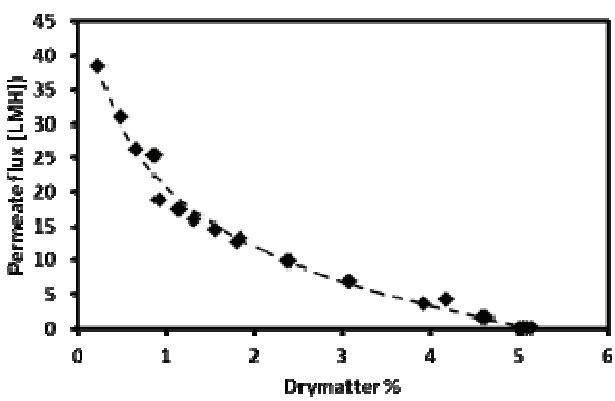

Figure 1 RO-permeate flux as a function of retentate DM-%

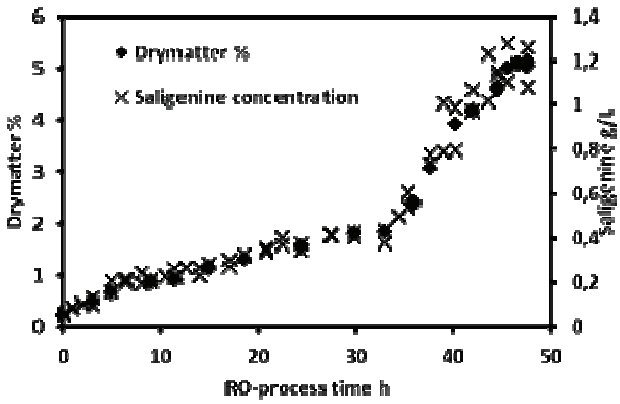

Figure 2 RO-concentrate drymater and saligenine content as a function of RO-processing time

Conclusion

Using a combination of MF, UF and RO industrial concentration of willow extract from 0.4% DM to 4-5 % DM is possible. Higher fluxes though would be necessary for the RO-process. This could be obtained with RO-membranes able to withstand higher transmembrane pressures. Further a higher end concentration would be desirable. This could be attained by including a membrane distillation step as the final concentration step.

References

[1] C. Sostros, C.J. Gargello, M.T. Arroyo,A. Lanas, (2010), Adverse effects of non-steroidal anti-inflammatory drugs (NSAIDs, aspirin and coixbs) on upper gastrointestianal tract, Best Practise & Research Clinical Gastroenterology, ***24****, 121-132.*

[2] S. Chuibasik, E. Eisenberg, E. Balan, T. Weinberger, R. Luzzati, C. Conradt (2000), Treatment of Low Back Pain Exacerbations with Willow Bark Extract: A Randomized Double-Blind Study, The American Journal of Medicine, ***109****, 9-14.*

[3] R. Julkunene-Tiitto, S. Sorsa, (2000), Testing the effect of Drying Methods on Willow Flavonoids, Tannins and Salicylates,Journal of Chemical Ecology ***27****, 779-789*

[4] G.A. Bonaterra, E.U. Heinrich, O. Kelber, D. Weiser, J. Metz, R. Kinscherf, (2010), Anti-inflammatory effects of the willow bark extract STW 33-I (Proaktiv) in LPS-activated human monocytes and differentiated macrophages, Phytomedicine, ***17****, 1106-1113*

Keywords: Willow Extract Concentrate, Microfiltration, Ultrafiltration, Reverse Osmosis