EVALUATION OF REVERSE OSMOSIS AND NANOFILTRATION MEMBRANES IN CONCENTRATION OF HAWTHORN FRUIT AND ANISE SEED EXTRACT.

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MAGYAR AGRÁR- ÉS ÉLETTUDOMÁNYI EGYETEM
Élelmiszertudományi és Technológiai Intézet

ITRODUCTION:

Hawthorn (*Crataegus. Monogyna*) is one of the most popular edible fruits and has been used to make wines and sweet or tinned foods, as well as jams and juices. It is also used to make medicinal products and functional foods for the treatment of chronic heart failure and high blood pressure (Liu et al.,2010). The antioxidant capacity of phenolic compounds present in different organs of the hawthorn has been reported in several studies (Caliskan et al.,2012).

Pimpinella anisum L. (anise) is one of the herbal spices. belong to the Apiaceae family which includes more than 3,500 species and many of them are considered nutraceuticals because of their nutritional, medicinal, and therapeutical properties, and as a source of raw materials used in the pharmaceutical, cosmetic, flavor, and perfumery industries (Sayed-Ahmad et al., 2017).

The main step in obtaining these active substances are purification and concentration. In recent years, researchers have paid a lot of attention to membrane technology, and they have considered it an environmentally benign technology for purifying natural extracts.

Despite the several advantages of this technology, however, choosing the appropriate membrane, optimal operating conditions, and membrane fouling are still the challenges of this technology. It is clear that the membrane efficiency depends on membrane type, solute, and the mutual interaction between them. Temperature, pH, pressure, and concentration also have an influence on rejection (Li et al., 2010). Nanofiltration and reverse osmosis membranes are basically used in water purification with the purpose of desalination. Even so, their applications in food and beverage industries are continuously expanding mostly in the concentration of juices (Alsobh et al.,2022) The main purpose of this investigation was to evaluate the efficiency of nanofiltration and reverse osmosis membranes on the concentration of valuable compounds from Anise seed and hawthorn fruit extracts.

MATERIALS AND METHODS:

Hawthorn fruit was collected from several trees grown in different regions in Hungary. After removing the sticks, the fruits were washed, cleaned, wiped to remove the water, and shredded to different portions using GM 200 pulverizer, the extraction of phenolic compounds process was performed by using (OS20-S Electric LED Digital Overhead Stirrer (220V)) at 55 °C, by using ethanol 56 v/v% as a solvent (10 g of the fruit in 100 mL solvent) for 80 min. For anise seed, the extractions were completed using water as solvent at 37 °C for 100 min..

Determination of total phenolic content

Total phenolic content was estimated by the Folin-Ciocalteu colorimetric method based on the method of Singleton and Rossi (1965).

Determination of total flavonoids content

The colorimetric method was used to measure flavonoids. Where the absorbance of the reaction mixture was measured at 415 nm using a spectrophotometer.

Determination of Antioxidant activity

Antioxidant activity was determined spectrophotometrically according to the method previously described by Benzie and Strain (1996).

Membrane separation:

Table 1: Filtration parameters

Membrane:	NF270,RO99,X20	Initial liquid volume:	3000 mL
Equipment:	DDS	Pump freq:	32.2
Surface of one membrane:	0.018 m^2	Recirc flow rate:	400 L/h
Pieces of membrane:	10	p1:	28 bar
Membrane Surface:	0.18 m^2	p2:	32 bar
Temperature:	35 °C	TMP:	30 bar

RESULTS:

Total Phenolic Compounds and flavonoids (TPC, TFC), Antioxidant Activity (AA):

Table 1: Total phenolic compounds, flavonoids, and antioxidant activity in crudes and retentates of X20 type reverse osmosis membrane separation

Plant	Sample	TPC (mg GAE/g dw)	TFC (mg QUE/g dw)	AA (mg ASE/g dw)
Anise	Initial	28.12 ±1.93	7.5±4.68	8.15±0.44
	R(400 mL)	32.52±0.45	8.91±0.32	9.15±1.84
	R(800 mL)	44.14±11.4	13.22±12.6	11.75±1.11
	R(1200 mL)	56.81±0.84	15.67±5.45	12.83±1.93
	R(1600 mL)	110.13±1.76	23.88± 2.66	22.92±0.78
	R(2000 mL)	115.77±1.81	25.12±1.93	24.76±0.45
Hawthorn	Initial	45.31±0.8	18.38±0.41	31.12±1.55
	R(400 mL)	52.58±2.67	20.47±0.12	33.46±1.57
	R(800 mL)	61.53±1.76	25.26±0.47	35.37±0.99
	R(1200 mL)	74.22±1.93	30.68±0.33	40.49±11.2
	R(1600 mL)	154.11±3.78	53.25±0.98	87.55±0.7
	R(2000 mL)	166.72±0.45	57.83±1.58	113.72±0.65

Table 2: Total phenolic compounds, flavonoids, and antioxidant activity in crudes and retentates of RO-99 type reverse osmosis membrane separation

Plant	Sample	TPC (mg GAE/g dw)	TFC (mg QUE/g dw)	AA (mg ASE/g dw)
Anise	Initial	28.12±1.93	7.5±4.68	8.15±0.44
	R(400 mL)	33.86±0.84	8.1±1.34	10.13±0.82
	R(800 mL)	37.26±5.67	8.92±0.88	12.87±1.6
	R(1200 mL)	55.79±3.69	9.88±3.7	13.21±0.81
	R(1600 mL)	62.57±0.48	12.36±2.89	15.52±1.23
	R(2000 mL)	87.68±3.78	18.82±0.46	19.86±2.46
Hawthorn	Initial	45.31±0.8	18.38±0.41	31.12±1.55
	R(400 mL)	57.65±1.93	19.13±5.98	35.41±2.78
	R(800 mL)	63.42±0.13	21.79±1.78	42.17±0.13
	R(1200 mL)	72.74±7.11	22.32±0.86	51.26±1.34
	R(1600 mL)	93.891±5.45	38.71±0.81	64.18±3.22
	R(2000 mL)	125.72±1.22	46.91±2.68	85.94±3.87

Table 3: Total phenolic compounds, flavonoids, and antioxidant activity in crudes and retentates of NF-270 type nanofiltration membrane separation

Plant	Sample	TPC (mg GAE/g dw)	TFC (mg QUE/g dw)	AA (mg ASE/g dw)
Anise	Initial	28.12±1.93	7.5±4.68	8.15± 0.44
	R(400 mL)	29.87±1.58	7.8±0.47	10.12± 1.01
	R(800 mL)	31.19±2.67	8.18±0.82	10.87±0.21
	R(1200 mL)	32.27±2.56	9.26±1.85	11.16±0.98
	R(1600 mL)	38.14±0.56	11.47±1.22	12.23±0.81
	R(2000 mL)	41.7±1.67	12.54±1.23	13.56±0.99
Hawthorn	Initial	45.31±0.8	18.38±0.41	31.12±1.55
	R(400 mL)	55.23±1.92	18.7±1.23	33.74±1.34
	R(800 mL)	63.12±0.81	19.56±0.98	37.41±2.02
	R(1200 mL)	70.17±0.45	22.46±0.34	42.81±2.6
	R(1600 mL)	83.19±1.73	28.87±0.93	56.29±1.9
	R(2000 mL)	114.61±2.99	31.45±1.23	78.35±1.45

We note in Tables 1, 2, and 3 that:

- The (TPC), and (TFC) increased during the concentration process. To reach the maximum using (X20) membrane. This membrane was superior to the other two membranes.
- The total phenolic and flavonoid retentions in both anise and hawthorn extracts were > 99%.
- With RO99 membrane the phenolic compounds and flavonoids retentions was (96.80%, 95.54%) respectively for anise extracts and (96.81%, 96.95) for hawthorn.
- The NF270 membrane was the least rejected to these compounds with retentions (77.93%, 78.16%) of phenolic compounds, and (75.25%, 82,65%) of flavonoids for anise and hawthorn extracts respectively.
- The final concentrated extract obtained using (X20) membrane contains the highest amount of antioxidants in final retentates.

Total soluble solids (TSS):

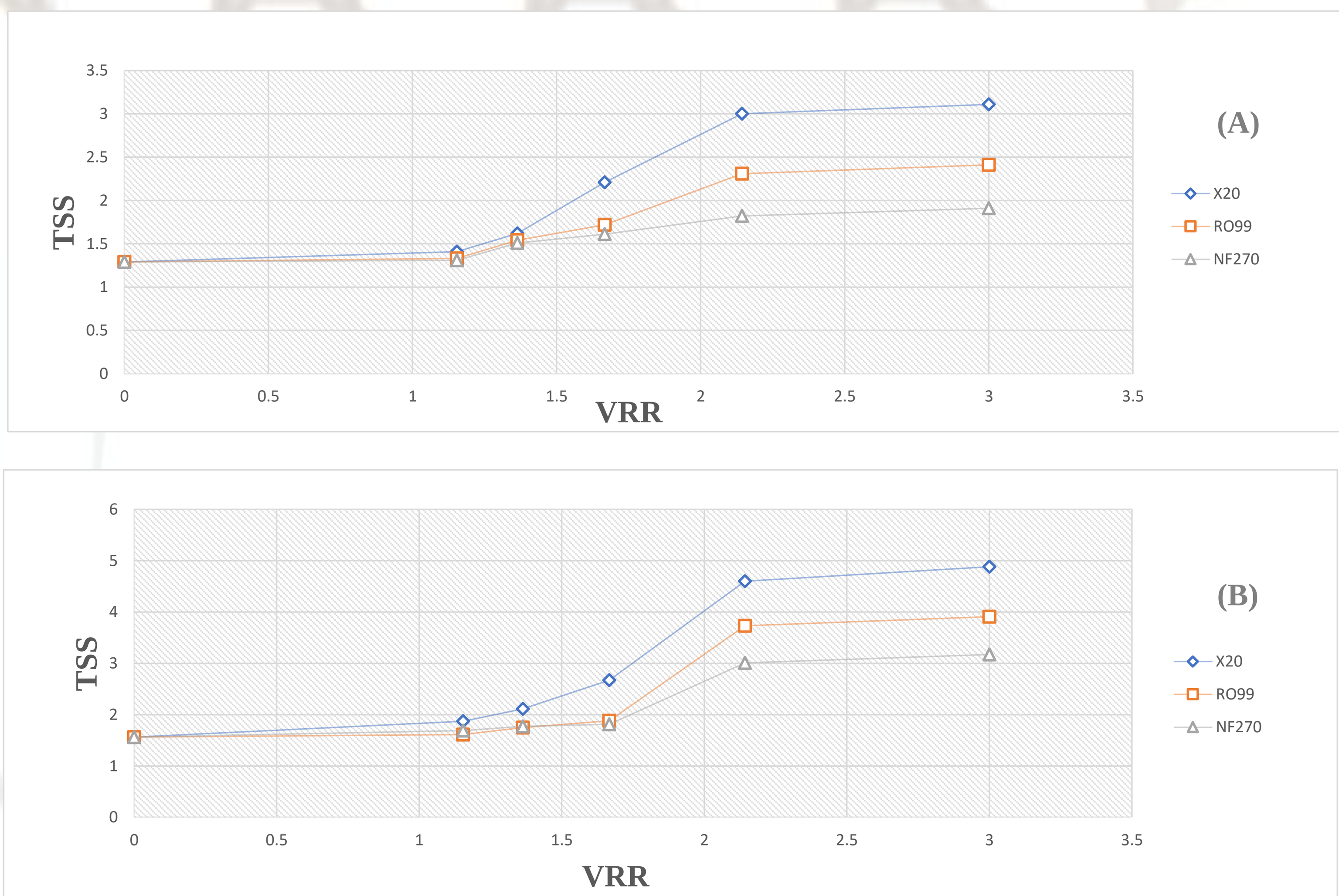

Figure 1: Change of TSS during the concentration (A) anise, (B) hawthorn.

The results of some studies compared membrane efficiencies in the concentration of plant extracts:

- (Nunes, et al., 2019): found that the reverse osmosis (BW30) membrane was the most effective for extracts concentration, and the total phenolics and flavonoid contents of the concentrate from BW30 were significantly higher(around 15%) than those achieved with NF270 and NF90.
- (Li et al. 2010): reported a comparative study using nanofiltration and reverse osmosis membranes for phenol removal from synthetic wastewater. Was observed that nanofiltration showed low rejection (0.41–0.72)with maximum flux (180 L m-2 h-1), while reverse osmosis recorded high rejection (0.81) with minimum flux (60 L m-2 h-1).
- (Zin et al.,2020): indicated that the antioxidant activity increased around 4.5 times using (X20) membrane, vs around 4 times using RO99 for ethanol-water extracts of beetroot peel.
- (Li et al., 2018): reported that using a nanofiltration membrane to concentrate grape seed extract increased the antioxidant activity around 2.24 times.

CONCLUSIONS:

Bioactive compounds with health and economic added value were recovered and concentrated from anise seed and hawthorn extracts using membrane technology. Two reverse osmosis (X20, RO-99), and one nanofiltration (NF270) membrane were evaluated in terms of efficiency in rejecting phenolic compounds and flavonoids. For the future scale-up of the process, the X20 membrane was found to be the most appropriate to be used since the rejection of X20 toward phenolic and flavonoid compounds was >99% which is the highest rejection value obtained, and concentrate presented the highest antioxidant activity. While the nanofiltration membrane was the least efficient with the rejection of phenolic compounds and flavonoids < 80%.

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