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New approach to the utilisation of concentrates obtained during geothermal water desalination

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ABSTRACT

The paper presents the results of research on the efficient use of concentrates obtained during the desalination of different types of geothermal water using membrane processes. The analysis is based on results for 18 concentrates which included macroelements and certain therapeutic ingredients, potentially toxic elements (heavy metals) and radioactive elements. The research demonstrated that concentrations of potentially toxic metals in most concentrates do not exceed the limits recognised as safe for human health, irrespective of the manner in which the human body comes into contact with them. These concentrates can be used: (1) for inhalation and rinsing the nose and mouth for the purpose of loosening and removing mucus and relieving inflammation; (2) for cosmetic purposes in the form of cleansing and moisturising liquids; (3) for bathing, both individual and in recreational or therapeutic pools. The second innovative approach is related to harvesting energy from the salinity gradient. The purpose is to purify geothermal water while also extracting the energy generated by mixing effluents with different salinity levels. The RO process results in the discharge of concentrated brine which can be considered a source of salinity gradient energy. In this respect, two methods are considered: reverse electrodialysis and capacitive mixing.

Keywords: Concentrate; Membrane processes; Human health; Salinity gradient; Energy

1. Introduction

The use of geothermal energy is seen as important part of renewable energy utilisation. Worldwide, geothermal water plays significant role as a source for electricity generation in conventional and binary cycle power plants [1,2], for heating purposes (district heating and hot water supply), in greenhouses, agriculture, for therapeutic and recreational purposes, etc. [3–5]. In physical and chemical terms, the water present in geological structures exhibits various properties. The types of water encountered include fresh waters, in which total dissolved substances (TDS) are below 1.0 g/L, brackish waters (TDS from 1 to 10 g/L), saline waters (TDS from 10 to 30 g/L) and brines (TDS more than 30 g/L). In many cases, the manner in which they have used results from their physical and chemical properties. These properties play a particular role when water is used in swimming pools or for balneological and therapeutic procedures where special attention is paid to the presence in geothermal water of elevated concentrations of minerals – therapeutic ingredients such as metasilicic acid, iodides, radon or hydrogen sulphide as these ingredients have a positive effect on the skin and on the respiratory system or play a role in the rehabilitation of the musculoskeletal system.

The objective of this article is to present both the traditional manner in which waste geothermal water is used and an innovative concept for its multidirectional use. The author's previous research, which was conducted on a pilot

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plant scale, demonstrated that waste geothermal water could be purified using membrane processes (ultrafiltration/nanofiltration/reverse osmosis) and subsequently reused as drinking water and/or water suitable for other household purposes [6–8]. In these processes, concentrate is usually perceived as a by-product/waste product resulting from the separation of minerals from the desalinated feedwater [9–12]. However, it is perhaps worth looking at the properties of the concentrate and consider whether its use could bring some economic benefits as well.

The paper presents two innovative approaches, both of them based on geothermal water treatment processes. First, results of research on the use of concentrate as a new product for therapeutic and balneological purposes and/or a source of medicinal raw materials are presented. The second innovative approach is related to the concept of harvesting energy from the salinity gradient by using the concentrate obtained during water treatment processes.

2. Methods

Research into the use of concentrates was conducted in parallel with the analysis of the quality of the permeate obtained as a result of treating geothermal waters. The scope of the research was very broad and included the determination of both physical (T, pH and EC) and chemical (macro- and microelements) properties, taking into account ingredients of therapeutic importance as well as potential toxic ingredients (heavy metals) and radioactive elements. The geothermal water treatment process was conducted using membrane techniques, with the technological setup adapted to the peculiar properties of raw water – so-called feed, that is, cooled geothermal water previously used for energy generation purposes. In each variant, this included pretreatment, which involved prefiltration in order to remove any suspensions present in the raw water, and ultrafiltration as a process which allowed the removal of colloids and fine suspensions (>0.05 μm). As geothermal waters often contain elevated iron concentrations (of up to several mg/L), the water pretreatment process also included iron removal using a catalyst bed. The pretreated water was fed to nanofiltration and/or reverse osmosis installations. The choice of treatment techniques was dependent on water hardness, its salinity and the content of ingredients which could potentially result in membrane scaling. The research was implemented on a pilot plant scale, with installation capacity amounting to approximately 0.5–1 m^3/h of treated water (permeate), with essentially two variants related to the configuration of the technological process:

- variant I: prefiltration, iron removal using a catalyst bed, ultrafiltration and reverse osmosis;
- variant II: prefiltration, iron removal using a catalyst bed, ultrafiltration, nanofiltration and reverse osmosis.

Installation diagrams are shown in Figs. 1 and 2.

In total, 18 tests were carried out, using five different geothermal waters. The tests were carried out on geothermal waters extracted from the following polish geothermal wells: GT-1, GT-2/3 (mixed water from two geothermal intakes, which were simultaneously operated in a continuous manner, which prevents long-term tests from being carried out in a selective manner for technological reasons), GT-4, GT-5 and GT-6. In total, 18 tests were conducted in a variety of process setups, which enabled the impact of changes in key factors on the quality of the products obtained to be identified. The pH and chemical properties of the feed, transmembrane pressure, recovery levels of both permeate and concentrate were all modified, and concentrate was also recirculated in the water treatment process.

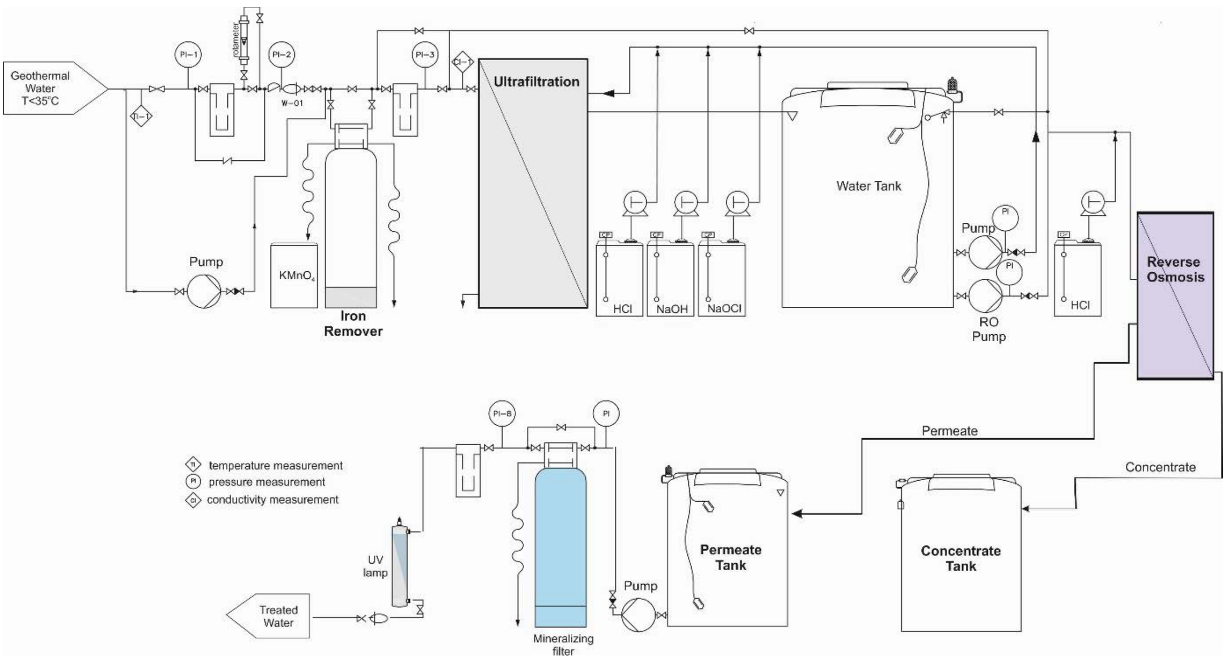

Fig. 1. Process diagram for variant I.

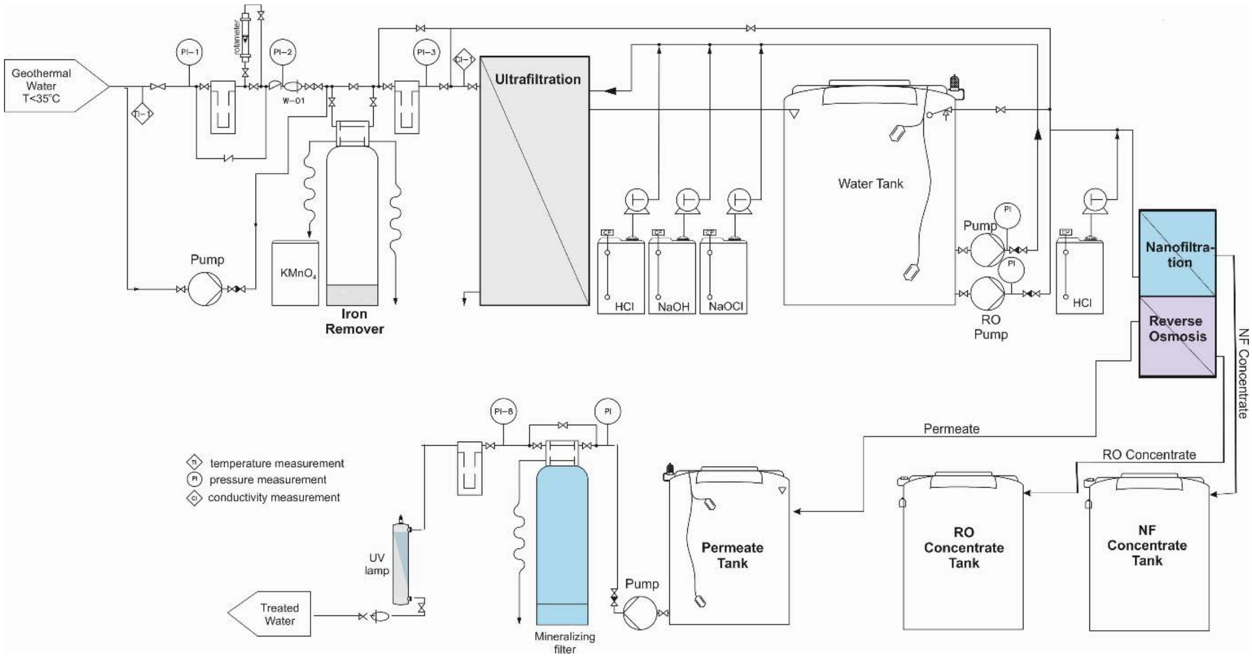

Fig. 2. Process diagram for variant II.

Basic data concerning the configuration of the water treatment installation depending on specific feed characteristics are presented in Table 1.

3. Study results and discussion

Apart from energy consumption, the key criterion for the implementation of RO technology is the volume of concentrate produced during the process. The volume of concentrate generated is even more critical for inland RO plants located in areas far away from the ocean [13]. The concentrate is highly saline with constituent concentrations found to be double or higher than those in feed water [14]. Sea and ocean waters, which are commonly used as feed in water treatment installations, generate concentrates with concentrations ranging from 50 up to 80 g/dm^3 [11], which results from the high feed TDS (from around a dozen to 35 g/dm^3). The considerations presented related to the use of geothermal water concentrates concern incomparably lower concentrations of minerals, which are dissolved in geothermal waters (feed water, which is subsequently desalinated). The desalination of geothermal waters with low mineral content (with TDS levels typically below 10 g/dm^3) may be a method of diversifying water supply for household purposes or technological ones, for example, in secondary circuits of district heat distribution systems. Capacities of water desalination installations in these cases are much lower than of those used on a large scale in areas where there are drinking water shortfalls. Since such small flows of geothermal water (up to several dozen m^3/h) are desalinated in areas located far from the sea, the manner of use/disposal of the concentrate is a serious challenge. For this reason, according to the author, the solutions proposed in this article may provide interesting options enabling the production of new products from geothermal waters.

3.1. Considerations related to the direct use of concentrate

For each of the waters tested, a series of tests aimed at obtaining new useful products (drinking water, technological water and useful concentrates) were conducted. These included desalination/concentration tests at varying permeate and concentrate recovery levels, but also experiments involving the recirculation of the concentrate obtained as a result of nanofiltration and reverse osmosis. This enabled concentrate to be more highly concentrated and physiochemically useful solutions (concentrates) to be obtained. During the tests, no antiscalants, biocides or other chemicals were used. Before RO-1, feed reaction was lowered to about 5.5 by dosing minuscule amounts of hydrochloric acid, which effectively prevented membrane scaling. Positive results of such measures have been presented in the author's other works [6].

Results of that research were described in the following opinion published by Zakład Tworzyw Uzdrowiskowych (Department of Materials Used in Spa Treatments) in Poznań of Państwowy Zakład Higieny (National Institute of Hygiene) in Warsaw [15]: *Ocena właściwości biochemicznych koncentratów pozyskanych w wyniku uzdatniania wód geotermalnych w aspekcie możliwości ich wykorzystania do celów kosmetycznych lub profilaktycznych* ('Assessment of biochemical properties of concentrates obtained as a result of the treatment of geothermal waters with respect to the possibility of using them for cosmetic or prophylactic purposes').

Using all geothermal waters studied, new interesting aqueous solutions were obtained, which were highly concentrated, with increased mineral content and high content of therapeutic ingredients (Table 2).

Of particular interest were the results of concentrating the waste fresh geothermal water extracted from the GT-5 well. In its natural state, geothermal water from the well analysed

Table 1
Configuration of the water treatment installation depending on specific feed geothermal water characteristics

Geothermal water	Feed water type	Process configuration	Permeate recovery level (%)	Recirculation of the concentrate (%)	Concentrate number	Concentrate type
GT-1	0.25% SO_4-Cl-Na-Ca	Variant I (Fig. 1)	90	100	1	1.36% SO_4-Cl-Na-Ca
			75	100	2	1.0% SO_4-Cl-Na-Ca
			83	100	3	0.98% SO_4-Cl-Na-Ca
			65	–	4	0.7% SO_4-Cl-Na-Ca
			84	–	5	0.97% SO_4-Cl-Na-Ca
GT-2/3	0.23% SO_4-Cl-Na-Ca	Variant I (Fig. 1)	50	–	1	0.97% Cl-SO_4- Na-Ca
			50	–	2	0.97% Cl-SO_4- Na-Ca
GT-4	0.62% Cl-Na	Variant II (NF concentrate) (Fig. 2)	70	–	1	1.2% Cl-Na
		Variant II (RO concentrate)	62	66	2	0.7% Cl-Na
		(Fig. 2)	66	–	3	0.63% Cl-Na
		Variant II (NF concentrate) (Fig. 2)	75	–	4	1.3% Cl-Na
GT-5	0.05% HCO_3-Ca-Na	Variant I (Fig. 1)	67	40	1	0.17% Cl-Ca-Na
			58	50	2	0.11% Cl-Ca-Na,
			80	–	3	0.21% Cl-SO_4-Na-Ca
			90	–	4	0.21% Cl-SO_4-Na-Ca
			83	–	5	0.35% Cl-SO_4-Na-Ca
GT-6	0.47% Cl-Na	Variant II (NF concentrate) (Fig. 2)	76	–	1	1.05% Cl-Na
			71	100	2	0.85% Cl-Na

exhibits a low TDS of 0.5 g/dm^3 and an increased content of therapeutic ingredients: iron ions (0.32 mg/dm^3) and metasilicic acid (26.57 mg/dm^3). As concerns its ionic composition, it is dominated by bicarbonates (319.2 mg/dm^3), calcium (58.69 mg/dm^3) and sodium (34.71 mg/dm^3), which results in the water being of the bicarbonate-calcium-sodium hydrogeochemical type. The water from the well-examined does not contain potentially toxic natural ingredients in concentrations considered harmful to health. Its heavy metal content is below the limit of quantification of the test apparatus. Additional radiological tests of the water analysed, which were conducted by Narodowy Instytut Zdrowia Publicznego – Państwowy Zakład Higieny (National Institute of Public Health – National Institute of Hygiene) in Warsaw [16], have demonstrated that radionuclide activity of this water is low and meets the requirements for water intended for human consumption. Its total alpha activity is 128.2 mBq/dm^3 while the WHO reference value [17] is 500 mBq/dm^3, and its total beta activity is 351.1 mBq/dm^3 while the WHO reference value [17] is 1,000 mBq/dm^3.

Concentration tests were successful both in terms of their efficiency and RO process stability. As a result of the tested water being concentrated in the RO installation at a transmembrane pressure of around 1.0 MPa, high-quality concentrate of the HCO_3-Ca-Na hydrogeochemical type and with a mineral content of 1.21 g/dm^3 was obtained. It exhibited an increased content of valuable minerals: metasilicic acid (65 mg/dm^3), calcium (145 mg/dm^3), magnesium (32 mg/dm^3) and potassium (32 mg/dm^3). Just as in the case of natural water, the concentration of heavy metals in the concentrate was found to be low, below the limit of quantification of the test apparatus. The factors which determine the possible uses of water include mainly the overall content of dissolved minerals, but also the types of dominant ingredients and their biochemical properties (Cl^-, Na^+, Ca^{2+}, Mg^{2+}, SO_4^{2-} and HCO_3^-) as well as the content of therapeutic ingredients (I^-, F^-, H_2SiO_3, S^{2-}, Rn and Fe^{2+}). According to the National Institute of Hygiene [15], the production of concentrates for human consumption purposes may be appropriate in the case of waters which contain increased levels of the aforementioned inorganic ingredients. The above issues were analysed in detail in relation to the solutions obtained. It was found that the calcium, magnesium or potassium content found in the concentrates obtained on the basis of water from the GT-5 well meets these requirements. In addition, at varying degrees of concentration, it was possible to obtain concentrates (0.11%–0.35%) of various hydrogeochemical types:

- Cl-Ca-Na (<0.2%) with I^-<0.2 mg/dm^3;
- Cl-SO_4-Na-Ca (>0.20%–0.35%) with iodide concentrations ranging from 0.14 to 0.55 mg/dm^3.

Due to the biochemically significant content of silica (metasilicic acid) and their reaction (pH = 8.21), in the opinion of the National Institute of Hygiene [15] these solutions can be used for cosmetic purposes as liquid cleansers and moisturisers.

Table 2
Concentrate characteristic obtained during geothermal water treatment

Element (mg/dm^3)	GT-1					GT-2/3		GT-4				GT-5					GT-6	
	1	2	3	4	5	1	2	1	2	3	4	1	2	3	4	5	1	2
TDS	13,743	10,147	9,880.2	6,827.5	9,761.6	5,545.4	4,359.2	12,460.9	6,981.9	6,289.7	12,741.7	1,767.9	1,212.9	2,238.6	2,108.5	3,763.2	10,550.1	8,504.9
Na	2,835	2,025	2,019	1,340	1,997	1,183.09	901.2	4,185	2,678	2,349	4,327	177.32	113.5	332.1	508.86	585.3	3,644.5	2,950.7
K	295.4	186.4	167.64	134	181	111.21	84.66	30.9	32.98	37.57	41.46	56.49	38.73	64.5	62.71	106.8	61.0	38.1
Ca	1,161	860	854.4	560.8	823.3	485.2	371.24	367.6	8.85	16.27	333.8	279.9	200.68	261.57	124.64	435.39	299.9	201.78
Mg	221	151.1	136.6	111.1	140.9	99.1	76.72	68.97	1.378	2.75	60.98	60.64	40.79	58.29	25.67	93.45	71.3	58.95
Cl	3,337	2,468	2,399	1,648	2,372	1,433.7	1,085	7,416	4,189	3,752	7,490	840.2	656.5	909.7	851.5	1,515.1	5,665	4,954
SO_4	4,973	3,723	3,630	2,518	3,521	1,744.7	1,443.05	256.7	10.77	41.03	316.6	64.14	18.22	391.83	391.4	723.6	607.5	218.07
I	2.52	2.89	2.65	3.73	4.11	1.07	0.68	–	–	–	–	0.14	0.08	0.34	0.38	0.55	0.16	0.05
As	0.014	0.017	0.021	0.021	0.025	0.012	0.01	0.027	0.019	0.02	0.029	0.025	0.026	0.018	0.009	0.029	0.023	0.018
F	4.7	–	–	2.4	–	3.2	–	–	–	–	–	2.1	–	–	–	–	0.95	0.63
Cr	0.006	0.013	0.019	0.022	0.021	0.012	0.009	0.018	0.014	0.011	0.019	0.018	0.016	0.019	0.017	0.031	0.023	0.023
Cd	<0.0003	<0.0003	<0.0003	<0.0003	<0.0003	<0.0003	<0.0003	<0.0003	<0.0003	<0.0003	<0.0003	0.0007	0.0005	0.0004	0.0004	0.0003	<0.0003	<0.0003
Ni	0.004	0.007	0.009	0.011	0.009	0.003	0.002	0.003	0.002	0.002	0.003	0.003	0.005	0.004	0.001	0.008	0.003	0.003
Pb	0.0024	0.0028	0.0033	0.0039	0.0031	0.0005	<0.0001	0.0092	0.006	0.0018	0.0085	0.0016	0.0069	0.0019	0.0008	0.0018	0.0105	0.0281
Hg	0.0001	0.0001	0.0002	<0.0001	0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	0.0025	0.001	0.0018	0.0026	0.0019	0.0006	0.0004
Al	0.006	<0.005	0.005	0.007	<0.005	<0.005	<0.005	<0.005	<0.005	0.009	<0.005	<0.005	<0.005	<0.005	<0.005	0.02	<0.005	<0.005
H_2SiO_3	386.5	316.56	283.17	226.99	296.58	187.2	144.05	81.92	22.30	26.12	93.73	149.55	98.75	151.97	80.95	245.77	91.21	58.11
HBO_2	101.23	75.16	72.40	72.60	77.03	62.95	52.35	5.11	8.64	12.95	6.77	1.95	0.88	13.12	11.30	19.05	2.58	0.91
Alpha (mBq/dm^3)	610	520	500	520	590	975	950	1,870	99	106	1,680	270	220	238	225	240	1,065	1,100
Beta (mBq/dm^3)	910	850	820	800	900	1,610	1,620	2,940	<300	310	2,850	1,350	1,320	1,410	1,380	1,350	1,215	1,200

A broad spectrum of possibilities was opened by tests concerning the concentration of waters from the G-1 and GT-2/3 wells, whose TDS levels in the natural state range from 2.5 to 3.0 mg/dm^3. The overall concentration level of concentrates and the fact that they contain significant amounts of sodium chloride as well as calcium, iodide and metasilicic acid compounds indicate that they could have beneficial biochemical effects when applied externally to skin and mucous membranes. The following concentrates classified as (Tables 1 and 2):

- 0.7% SO_4-Cl-Na-Ca solution with a pH of 6.75 containing 3.73 mg/dm^3 of iodides, 227 mg/dm^3 of metasilicic acid as well as 16.95 mg/dm^3 of manganese, 0.021 mg/dm^3 of arsenic and 73 mg/dm^3 of metaboric acid;
- 0.55% Cl-SO_4-Na-Ca solution with a pH of 6.93 containing 1.07 mg/dm^3 of iodides, 187 mg/dm^3 of metasilicic acid and 62 mg/dm^3 of metaboric acid;
- 0.43% Cl-SO_4-Na-Ca solution with a pH of 6.42 containing 0.68 mg/dm^3 of iodides, 144 mg/dm^3 of metasilicic acid and 52.36 mg/dm^3 of metaboric acid.

can be used undiluted for inhalation and for rinsing the nose and mouth in order to loosen and remove mucus and relieve inflammation.

On the other hand, concentrates with concentration levels >0.9% and iodide content >1 mg/dm^3 are suitable for use when bathing in a bathtub. However, it should be noted that the concentration of iodides in these concentrates, which is beneficial due to their significant impact on the human body, may make it difficult to use them in swimming pools. This is due to the fact that water in these installations is treated by chlorination, which results in the production of trihalomethanes – toxic volatile compounds formed by free chlorine in contact with iodine and bromine [18].

In turn, the concentrate which is a 1.37% SO_4-Cl-Na-Ca solution with a pH of 7.07, containing 2.52 mg/dm^3 of iodides, 386.5 mg/dm^3 of metasilicic acid and 4.7 mg/dm^3 of fluoride can be used as a solution suitable for gargling and rinsing the mouth, for example, in order to prevent tooth decay.

The overall mineral content of all concentrates, with the predominant ingredient being sodium chloride, indicates their suitability mainly for bathing – both individual and in recreational or therapeutic pools. Used in the bath, salt solutions (mainly NaCl) with concentrations of >0.5% stimulate the human body; when combined with physical factors (temperature, hydrostatic pressure), the systematic use of such baths strengthens the body's immune functions as well as improving motor functions. The silicon and iodine compounds also present in the solution may enhance these effects, especially on the skin.

The tests conducted have demonstrated that the concentrations of potentially toxic metals measured in most concentrates do not exceed the limits recognised as safe for human health irrespective of the manner in which the concentrates come into contact with the human body (Table 2). Elevated concentrations of toxic substances always limit the possibility of administering concentrates via the oral route. In the case of the waters tested, several concentrates, especially after they had been recirculated during the concentration process, were found to contain arsenic in concentrations ranging from <0.01 to 0.029 mg/dm^3 and their overall radioactivity amounted to $\alpha > 0.1$ Bq/dm^3 and $\beta > 1$ Bq/dm^3, which makes them suitable for external use only.

3.2. Considerations related to the indirect use of concentrate

There is also alternative, but equally innovative, possibilities for the indirect use of geothermal water concentrates: for the generation of electricity from the salinity gradient. Salinity gradient energy is a form of energy available whenever two solutions with different salinity levels are mixed, which occurs in nature when a river discharges into the sea [19]. Of course, the spontaneous mixing of river water into the sea results in the complete dissipation of the energy associated with the mixing process. The harnessing of this energy would require a suitable device capable of performing a 'controlled mixing' of the two streams with different salinity levels (e.g., river water and seawater). Pattle [20] was the first to describe this form of energy. He concluded: *'The osmotic pressure of sea-water is about 20 atmospheres, so that when a river mixes with sea, free energy equal to that obtainable from a waterfall 680 ft high is lost'*. Studies on the application of two methods: reverse electrodialysis (RED) and capacitive mixing (CAPMIX) are currently being carried out jointly by researchers from Poland and Turkey under a bilateral international project. Both mentioned methods are based on transportation of ions and can extract 50%–85% of total energy from the salinity gradient. In both methods, the issue of mixing the two solutions with low and high salinity (e.g., concentrate) is crucial, allowing the Gibbs mixing energy to be converted into electricity [21]. In the case of the RED system, the point is to use alternately organised ion-exchange membranes and pump both solutions between them. The RED system is composed of a repetitive assembly of cation and anion exchange membranes, between which salty and fresh water are pumped [22]. The ionic current generated is converted to electrical current by a redox couple at the electrodes. In the case of the CAPMIX system, water is pumped sequentially between two carbon super-capacitors: saline and nonsaline solutions [23]. The CAPMIX system, described by Brogioli [24], performs the mixing of the two solutions in a controlled way, generating an electrical current by periodically switching between the high-salinity and low-salinity feed solution. A detailed description of the technology for energy recovery from the salinity gradient can be found in the following works: Micale et al. [19], Kempener and Neuman [21], Vermaas et al. [22], Brogioli et al. [25], Fernandez et al. [26], Iglesias et al. [27] and Jia et al. [28]. To date, these solutions have only been discussed in relation to mixing ordinary water, mainly river and sea/ocean water. The use of geothermal water concentrates in order to generate electricity from the salinity gradient is an innovation which could contribute to a more rational use of water and geothermal energy resources. Test results and the feasibility of energy recovery from concentrates obtained with membrane techniques will be the subject of further scientific research.

4. Summary

The determination of the fashion in which the concentrate obtained during water treatment processes could be used is among the key factors determining the feasibility of the implementation of such processes in inland areas. The tests conducted using membrane techniques have demonstrated that the concentrate in question may be a useful product in cosmetology, for inhalation and for rinsing the nose and mouth in order to loosen and remove mucus and relieve inflammation as well as for bathing and therapeutic purposes. However, the possibility of obtaining and developing new useful products will be crucially determined by physical and chemical properties of the feed (natural geothermal water), the desalination/concentration techniques used and the use or nonuse of chemicals.

In addition to the proposed direct use of concentrates, research is being conducted on their use for the generation of electricity from the salinity gradient. This innovative solution could prove interesting in the context of rational use of geothermal energy with support from modern technological processes.

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