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EXTRACTION OF THE FOOD ADDITIVE TARTARIC ACID USING OCTANOL, METHYL ISOBUTYL KETONE, KEROSENE, MUSTARD OIL, AND GROUNDNUT OIL

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Tartaric acid (TA) is a dicarboxylic acid found in bananas, grapes, apples, papaya, cherries, pineapple, pears, mangoes, and tamarind. Due to its widespread use in the food, cosmetic and pharmaceutical industries, it is an essential carboxylic acid. Tartaric acid is produced commercially from wine-industry byproducts and is also present in the industry's effluent. Separating tartaric acid from wastewater is challenging. In this research, tartaric acid was separated from the aqueous phase using chemical and organic solvents such as groundnut oil, mustard oil, kerosene, octanol, and methyl isobutyl ketone (MIBK). Experiments were conducted at 298 K to determine the extraction efficiency ($E\%$) and distribution coefficient (K_D). The maximum extraction efficiencies of tartaric acid were found to be 49.01, 25.62, 16.73, 15.89 and 14.29% when using MIBK, octanol, kerosene, mustard oil and groundnut oil, respectively. The results demonstrate the significance of solvent choice in the extraction of tartaric acid with solvents such as MIBK and octanol being more effective at extracting TA from aqueous solutions. On the other hand, the sustainability of the method for separating tartaric acid was highlighted when organic solvents were applied.

Keywords: tartaric acid, separation, solvent, sustainable process, extraction efficiency

1. Introduction

Tartaric acid (TA), also known as 2,3-dihydroxy-butanedioic acid, is a white, crystalline organic acid found naturally in tamarind, grapes, citruses, and bananas. Winemakers have been aware of tartaric acid for centuries. It is one of the least antimicrobially active organic acids, inhibiting microbial growth and inactivating fewer microbes than malic, ascorbic, formic, citric, lactic, acetic, levulinic, fumaric, propionic and benzoic acids. In the food and wine industries, tartaric acid is primarily used as a preservative, pH regulator, chelating agent, emulsifier, antioxidant, acidifier, firming agent and acidulant [1]-[3]. It can be utilized to separate and purify metals and tan leather in addition to as an oil and lubricant additive. Its salts are also utilized in the production of pharmaceuticals, pigments, textiles, mirrors, cigarettes, and cosmetics. In the wine industry, tartaric acid - which is found naturally in grapes as well as contributes to the acidity and flavor profiles of grape juice and wine - plays an important role. The wine industry utilizes tartaric acid primarily for acidification, stabilization, pH adjustment, flavor enhancement and tartrate removal.

Tartaric acid is derived from potassium acid tartrate, a byproduct of the wine industry consisting of fermented and partially fermented press cakes of grape juice [4]. It can be produced through a variety of processes, including chemical synthesis and natural extraction from plant sources such as grapes. The basic stages of each technique are as follows: the precursor chemicals, including glucose and fructose, are derived from renewable resources, or synthesized from other compounds. Using a catalyst, typically a metal catalyst, and other chemical reactions, the precursor compounds are oxidized to produce tartaric acid. The resultant tartaric acid is then purified through crystallization, filtration, or chromatography to remove impurities and obtain high-quality tartaric acid.

Germany, Spain, France, and Italy are among the leading nations worldwide in terms of TA production. From 2018 to 2026, the demand for TA is projected to increase at a compound annual growth rate of 5.54% [5].

The wine industry generates a substantial amount of environmentally damaging waste streams during production where tartaric acid is readily available. Before being released, their environmental impact must be reduced [6]. TA is separated by precipitation in the form of calcium or potassium tartrate, which can be recovered

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Table 1: List of the chemicals used and their properties without any pretreatment in the present study

Chemicals	*MW* (g/mol)	S_{water} (g/L)	*Density* (kg/m^3)	Supplier
Tartaric acid	150	1.33	1760	HiMedia Laboratories
Groundnut oil	_	Immiscible	912	Patanjali Ayurved Ltd.
Mustard oil	99.15	Immiscible	920	Patanjali Ayurved Ltd.
Kerosene	170	Immiscible	820	Local store
Octanol	130.23	Immiscible	824	Loba Chemie Pvt. Ltd.
MIBK	100.16	0.19	802	Spectrochem Pvt. Ltd.

through filtration and sulfuric acid treatment. Although this method is effective, it is extremely complicated, costly, laborious and environmentally damaging as large quantities of sulfate residue are generated [7]-[8].

Tartaric acid can be extracted from winery waste and effluent, which are generated by the winemaking industry. It is one of the primary acids present, moreover, is used as a sour flavor enhancer and antioxidant in other foods. Tartaric acid is found in three different chiral structures: dextro tartaric acid or L(+) tartaric acid, levo tartaric acid or D(-) tartaric acid and meso tartaric acid.

Kontogiannopoulos et al. (2017) recovered 42.6 g/L of TA from wine residue using a nanofiltration membrane [9]. Several authors [10]-[12] have also utilized ion exchange and adsorption to recover TA. Although liquid membranes are a new promising method for the separation of carboxylic acids [13], they also exhibit fouling problems. However, since the membrane process is expensive and plagued with contamination issues, industries require a simple and cost-effective method for extracting TA from winery wastewater.

The literature indicates that liquid-liquid extraction is a proven method for extracting carboxylic acids from fermentation fluids and water streams. In this procedure, a water-immiscible extractant is introduced into a solution and the solute is extracted based on the relative solubility of the solute molecules in the two media. Solvent selection is central in the liquid-liquid extraction method [14]. Several authors [15]-[18] have studied the solvent extraction method for extracting TA using kerosene, 1-decanol, 1-octanol and n-heptane as chemical solvents.

In this study, TA was separated from the aqueous phase using chemical and organic solvents such as groundnut oil, petroleum, mustard oil, octanol and MIBK (methyl isobutyl ketone). To reduce toxicity and create an environmentally favorable method for separating TA from the aqueous phase, organic solvents were utilized. This experimental study was defined using several extraction parameters such as extraction efficiency (*E%*) and distribution coefficient (K_D).

2. Experimental

2.1. Samples and measurements

Tartaric acid was purchased from HiMedia Laboratories, while solvents like groundnut oil, mustard oil, octanol, MIBK and kerosene were procured from Patanjali Ayurved Ltd., Loba Chemie, Spectrochem and a local store (*Table 1*). Sodium hydroxide pellets were procured from Merck India Private Limited for the titration, while oxalic acid was acquired from S D Fine-Chem Ltd. (India). To normalize the NaOH solution, oxalic acid was utilized. Every time a new 0.01 M solution of NaOH was prepared for titration in the study, all the chemicals used were not purified or pre-treated beforehand.

2.2. Experimental procedure

Aqueous solutions of varying concentrations (0.0406 mol/L–0.1960 mol/L) of TA were prepared by dissolving different amounts of TA in distilled water. The initial concentrations of the samples were measured between pH 2.91 and 3.85 using an IKA® ETS-D pH meter (*Table 2*). The range of initial concentrations of the acid was in line with its industrial concentrations in winery waste streams [19]. Various solvents like groundnut oil, mustard oil, kerosene, octanol and MIBK were used individually as solvents in the organic phase.

Table 2: List of instruments used in the present study

Instruments	Conditions	Model
Orbital Shaking Incubator	T=25°C, rpm=200	S-24, REMI India
Bench Top Centrifuge	T=25°C, rpm=4000	R-4C, REMI India
pH meter	T=25°C	IKA® ETS-D6, India

Both the organic and aqueous phases were prepared in a conical flask at a volumetric ratio of 1:1. Until equilibrium was reached, the samples were kept in a REMI Orbital Shaking Incubator at 298 ± 1 K for 5 hours and shaken at 200 rpm before the samples were centrifuged for 5 mins at 4000 rpm (REMI R-4C, India) to clearly separate the aqueous and organic phases (*Table 2*). Having been separated, the pH was measured in the aqueous samples and titrated against a freshly prepared 0.1 M NaOH solution to determine the concentration of TA in the aqueous phase.

Mass balance equations were used to calculate the concentration of the TA organic phase. The change in volume of both phases after separation was checked and found to be negligible.

3. Results and Analysis

The separation of TA from the aqueous phase using various solvents such as groundnut oil, mustard oil, kerosene, octanol and MIBK was experimentally investigated (*Table 3*).

The following equations define the experiments:

$$[TA]_{in} + [S]_{org} \leftrightarrow [TA]_{org}.[S]_{org} \qquad (1)$$

where $[TA]_{in}$ denotes the initial concentration of tartaric acid; $[TA]_{org}$ stands for the concentration of tartaric acid in the organic phase and $[S]_{org}$ represents the concentration of the solvent in the organic phase.

Using the concentrations in the aqueous phase, mass balancing was used to determine those in the organic phase:

$$[TA]_{in} - [TA]_{org} = [TA]_{aq} \qquad (2)$$

To achieve accurate results, each experiment was repeated three times.

The experimental data concerning the separation of TA was defined by the distribution coefficient (K_D) and extraction efficiency ($E\%$).

The ratio of the organic phase to the aqueous one under equilibrium conditions is known as the distribution coefficient and can be defined as follows:

$$K_D = \frac{[TA]_{org}}{[TA]_{aq}} \qquad (3)$$

where $[TA]_{org}$ and $[TA]_{aq}$ denote the tartaric acid concentrations in the organic and aqueous phases, respectively, in mol/L.

In terms of the distribution coefficient, the extraction efficiency can be defined as:

$$E\% = \frac{K_D}{1 + K_D} \times 100 \qquad (4)$$

Table 3: Separation of tartaric acid using various organic and conventional solvents at 298±1K

Solvents	$[TA]_{in}$ (mol/L)	$[TA]_{aq}$ (mol/L)	$[TA]_{org}$ (mol/L)	K_D	E%
Groundnut oil	0.0406	0.0400	0.0006	0.015	1.48
	0.0798	0.0768	0.0030	0.039	3.76
	0.1212	0.1130	0.0082	0.073	6.77
	0.1540	0.1366	0.0174	0.127	11.30
	0.1960	0.1680	0.0280	0.167	14.29
Mustard oil	0.0406	0.0384	0.0022	0.057	5.42
	0.0798	0.0720	0.0078	0.108	9.77
	0.1212	0.1050	0.0162	0.154	13.37
	0.1540	0.1300	0.0240	0.185	15.58
	0.1960	0.1650	0.0310	0.188	15.82
Kerosene	0.0406	0.0392	0.0015	0.037	3.57
	0.0798	0.0740	0.0058	0.078	7.27
	0.1212	0.1056	0.0156	0.148	12.87
	0.1540	0.1296	0.0244	0.188	15.84
	0.1960	0.1632	0.0328	0.201	16.73
Octanol	0.0406	0.0318	0.0088	0.276	21.63
	0.0798	0.0625	0.0173	0.276	21.64
	0.1212	0.0945	0.0267	0.283	22.03
	0.1540	0.1154	0.0386	0.334	25.04
	0.1960	0.1458	0.0502	0.344	25.62
MIBK	0.0406	0.0330	0.0076	0.230	18.71
	0.0798	0.0592	0.0206	0.348	25.81
	0.1212	0.0735	0.0477	0.649	39.34
	0.1540	0.0867	0.0673	0.776	43.68
	0.1960	0.0999	0.0961	0.961	49.01

4. Discussion

The extraction data under equilibrium conditions indicate that molecules of tartaric acid are perpetually relatively soluble in the organic phase, while the concentration of TA in the aqueous phase increases (*Figure 1*).

The experimental results depicted the highest distribution coefficient values of 0.961, 0.344, 0.201, 0.188 and 0.167 with MIBK, octanol, kerosene, mustard oil and groundnut oil, respectively, at a concentration of 0.1960 mol/L of TA (*Figure 2*). The distribution coefficient was influenced by the initial concentrations of the TA. As the initial concentration increased, the distribution coefficient also rose when all the solvents were used. A higher distribution coefficient was observed when MIBK was used with 0.1960 mol/L of TA.

According to *Figure 2*, conventional solvents like MIBK and octanol exhibit higher distribution coefficients compared to organic solvents like mustard oil, groundnut oil and kerosene because these solvents are immiscible in the aqueous phase, moreover, cannot form free hydrogen bonds due to their high viscosity and density [20]. These solvents have low K_D values. Since the organic phase cannot dissolve in the aqueous one, these solvents form a barrier between aqueous and organic phases, resulting in oils yielding lower distribution coefficients [21]. The distribution coefficient of an acid rises as its molecular weight increases because the distribution coefficient is influenced by the molecular weight and solubility [22].

The extraction efficiency also increased by increasing the initial concentration of TA using all solvents (*Figure 3*). According to the experimental study, the maximum extraction efficiency of 49.01% was observed using MIBK, while the lowest of 14.29% was measured in groundnut oil. Due to the non-polar nature and long fatty acids in organic solvents, their extraction efficiency is poorer than chemical solvents. Since the organic solvents have no dipole moment, they were unable to form complexes with the acid in the organic phase with free electrons nor serve as a hydrogen donor in solvents other than octanol and MIBK. The extraction efficiency when using MIBK was higher than with octanol because MIBK is a positively charged polar solvent. Therefore, it dissolves more easily in tartaric acid in the organic phase, resulting in a higher extraction efficiency. In contrast, octanol is a non-polar solvent due to the large hydrophobic group it contains, limiting mass transfer from the aqueous to the organic phase when compared to MIBK [23]. The maximum extraction efficiencies were 49.01, 25.62, 16.73, 15.82 and 14.29% when MIBK, octanol, kerosene, mustard oil, and groundnut oil were used, respectively, with 0.1960 mol/L of tartaric acid. Since the extraction efficiencies of organic solvents were less than conventional ones, they can be used with various extractants like tributylphosphate, trioctylamine, Aliquat 336, etc. to increase the extraction efficiency of TA.

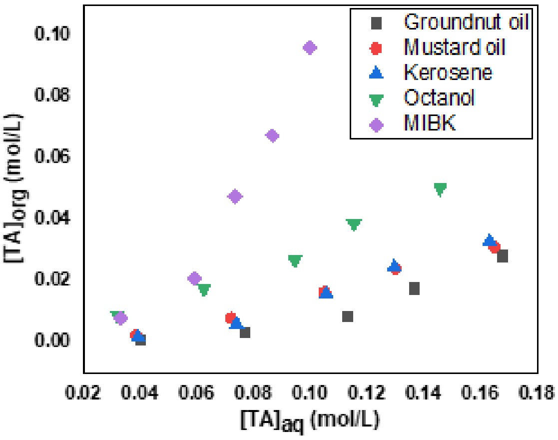

Figure 1: Equilibrium data for the separation of tartaric acid using various solvents at 298±1 K

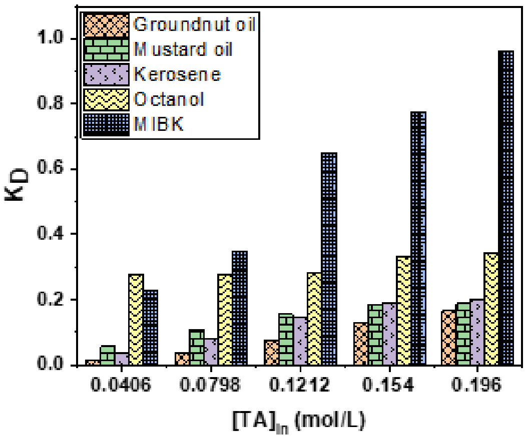

Figure 2: Variation in the distribution coefficient against the initial concentrations of tartaric acid at 298±1 K

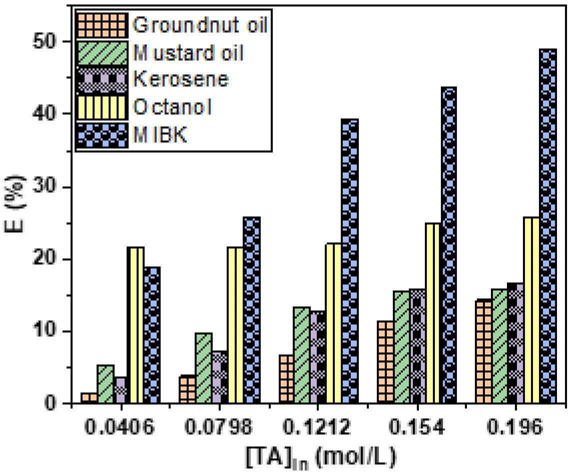

Figure 3: Variation in the extraction efficiency against the initial concentrations of tartaric acid at 298±1K

5. Conclusion

Tartaric acid was separated by an extraction process from the aqueous phase using groundnut oil, mustard oil, kerosene, octanol and MIBK as solvents. When MIBK was used, higher values for the distribution coefficient (0.961) and extraction efficiency (49.01%) were recorded, the latter was measured at a TA concentration of 0.1960 mol/L in the case of all the solvents. Since the extraction efficiencies of the organic solvents were lower than their chemical counterparts, the former can be used with extractants and in a closed loop with the mixer-settler reactor during the next step to improve the extraction efficiency of TA. When organic solvents are applied, the toxic nature of using an extractant can be avoided, thereby preventing the organic phase coming into contact with the organism and providing an environmentally-friendly process for extracting the TA.

SYMBOLS

MW	molecular weight
S_{water}	solubility in water
K_D	distribution coefficient
$E\%$	extraction efficiency
$[TA]_{in}$	initial concentration of tartaric acid
$[TA]_{aq}$	concentration of tartaric acid in the aqueous phase
$[TA]_{org}$	concentration of tartaric acid in the organic phase
$[S]_{org}$	concentration of the solvent in the organic phase

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