ISSN 1061-9348, Journal of Analytical Chemistry, 2018, Vol. 73, No. 9, pp. 855–861. © Pleiades Publishing, Ltd., 2018.

ARTICLES

Molecularly Imprinted Polymer for Selective Extraction of Tartaric Acid[1]

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Received August 10, 2016; in final form, October 15, 2017

Abstract—A molecularly imprinted polymer (**MIP**) was synthetized through precipitation polymerization method for the detection of tartaric acid. Acrylamide or methacrylic acid as functional monomers as well as amount of cross-linker were regarded as polymerization variables. The reactants were dissolved in acetonitrile-ethanol (8 : 2) solution. The synthetized polymers were characterized by HPLC, fourier transform infrared spectrometry (**FTIR**), thermogravimetric and dynamic light scattering techniques. Batch adsorption experiments with control by HPLC revealed better binding capacity of acrylamide-based polymers compared to methacrylic acid polymers in phosphate buffer saline solution as binding medium. Highest binding capacity was obtained at 30 : 1 cross-linker to template ratio. Tartaric acid complexation with acrylamide was affirmed by FTIR. Tartaric acid is the dominant acid in red grape and can be utilized as red grape juice "fingerprint" in other juices like pomegranate juice which is known to be adulterated in beverage industry. Based on the obtained results, this easily synthesized polymer for tartaric acid can be used in the detection of the presence of red grape juice in pomegranate juice. The synthesized MIP can potentially be applied in sensors for tartaric acid.

Keywords: acrylamide, molecularly imprinted polymer, tartaric acid

DOI: 10.1134/S1061934818090046

Adulteration in beverage industry is one of the important issues in food quality control and safety programs. A common method of fruit juice adulteration is the use of other juices to comply to the juice requirements. Pomegranate juice is a reach source of antioxidant compounds, higher than most other fruit juices, including anthocyanins (delphinidin, cyanidin, and pelargonidin) and other phenolic compounds (hydrolysable tannins such as punicalin, pedunculagin, punicalagin, gallagic and ellagic acid) which give it many health promoting effects [1]. As a result, pomegranate juice consumption is growing fast, and accordingly adulteration such as addition of red grape juice to valuable juices like pomegranate juice for reducing the cost of juice production has been increased.

Tartaric acid (**TA**) is the dominant and strongest organic acid existing in grapes. It can be used as a ‘‘fingerprint” indicator for detecting grape juice presence [2, 3], as tartaric acid is not a part of pomegranate organic acids profile [4]. There are differences in the specifications of organic acids in juices. Common techniques used in the field of food chemistry are enzymatic method, chromatography [5, 6], spectrophotometry [2, 3], electroanalysis and capillary electrophoresis [7, 8]. These techniques have disadvantages such as cost ineffectiveness, insufficient selectivity, time consuming and hard sample preparation [9].

A new method capable of obviating the mentioned disadvantages, called molecular imprinting, is a technology providing synthetic polymer (MIP) with specific cavity matching with a target molecule. This method involved radical polymerization of functional monomers with the target molecule via covalent or non-covalent interactions parallel with monomer arrangements using a cross-linking agent. Once the template was eluted by chemical reaction or extraction, specific binding sites were created which were complementary to the template in size, shape, and position of the functional groups [9, 10]. MIPs are stable at extreme pH values, organic solvents and temperature, which allows for more flexibility in the analytical methods. High affinity, selectivity, stability and simplicity of preparation are among other several advantages of MIPs [10, 11]. One of the facile and most widely used methods in the synthesis of molecu-

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larly imprinted polymer is precipitation polymerization which is performed in a more dilute polymerization mixture and results in sub-micron sized polymers in comparison with other techniques [12, 13].

Tan et al. [14] have developed a titania-based fluorescent indicator for the discrimination of fruit juices. Citric, malic, succinic and tartaric acids were investigated. They reported the use of titania as the imprinted matrix and only one fluorophore as the indicator. They found that a small array showed enough ability for discerning carboxylic acids. Tadi and Motghare [9] studied potentiometric determination of oxalic acid in spinach using polyvinyl chloride–oxalic acid imprinted polymer coated graphite electrode. He et al. [15] investigated the adsorption of D- and L-tartaric acids by β-cyclodextrin derivative modified with L-tryptophan. They reported that L-tryptophan–cyclodextrin had significantly higher adsorption capacity for L-TA than D-TA.

Several variables, including monomer type and amount, cross-linker, initiator and solvent amount, affected the final characteristics of MIPs including capacity, affinity and selectivity for the target molecule. In case of precipitation polymerization, aiming in this study, optimizing the cross-linker amount was of critical importance, and it was reported that by reducing the concentration of the template, the polymer binding properties were improved and the level of non-specific interactions was decreased [16]. In addition, according to Tadi and Motghare [9] and Subrahmanyam and Piletsky [11], when the template had hydrogen bonding functional groups, acrylamide served as the best functional monomer in polar solvents, as could be seen in the calculations of binding energies of template-monomer complex.

Considering the mentioned studies, this work aimed at fabricating a facile molecularly imprinted polymer for separating tartaric acid from red grape juice medium through precipitation polymerization and evaluating the potential of this method in tartaric acid separation and detection via applying HPLC, FTIR, thermogravimetric and particle size analysis methods. According to the obtained results, this MIP could be further applied in sensors for tartaric acid separation or in solid phase extraction for improvement of sample preparation via selectivity increment.

EXPERIMENTAL

Reagents, solutions and apparatus. All polymerization reagents including methacrylic acid (**MAA**), acrylamide (**AA**), ethylene glycol dimethacrylate (**EGDMA**), 2,2'-azobisisobutyronitrile (**AIBN**), the template L(+)-tartaric acid, $C_4H_6O_6$), potassium dihydrogen phosphate (KH_2PO_4) and HPLC grade acetonitrile were purchased from Merck (Darmstadt, Germany). Methanol (CH_3OH) and glacial acetic acid (CH_3COOH) as washing solvent and ethanol (C_2H_5OH) were purchased from Scharlau (Barcelona, Spain) and CDH (New Delhi, India), respectively. Double distilled water and deionized water were used in all solutions and experiments.

Devices applied in this study were: centrifuge (Hettich, EBA 270, Germany), ultrasonic equipment (Ultrasonic cleaner, Parasonic 2600s, Iran), micro centrifuge (Eppendorf, MiniSpin, Germany), laboratory balance (Acculab Sartorius group, Atilon, USA), magnetic heater-stirrer (Heidolph, MR Hei-Standard, Germany), HPLC (Cecil, CE4900, UK), FTIR spectrometer (Perkin Elmer, Spectrum Two, USA), oven (Memmert, EFB 400, Germany), deionizer (Millipore, Direct Q UV-3, France), horizontal tube shaker (Behdad, Iran), particle size analyzer (Malvern Zetasizer, Nano-ZS, UK), scanning electron microscope (Zeiss, LEO 1430VP, Germany–UK), thermogravimetric analyzer (Linseis, STA PT-1000, Germany) and Brunauer–Emmett–Teller (**BET**) surface analyzer (Belsorp mini II, BEL, Japan).

The synthesis of tartaric acid molecularly imprinted polymer through precipitation polymerization. The template, 0.3 mmol tartaric acid, was added into 100 mL flask containing 20 mL acetonitrile–ethanol (80 : 20) as porogen and solvent, respectively, and mixed by sonication until complete dissolution. Monomers (MAA, AA), cross-linker (EGDMA: at 6 and 9 mmol) and initiator (AIBN) were then added. After sonication for 5 min, degassing of the mixture was done by purging N_2 for 10 min and then the container was sealed completely. Polymerization was launched by heating the mixture at 60°C in a water bath. After 24 h, the obtained solid polymer was washed by methanol–acetic acid (90 : 10) until no tartaric acid was detected in the supernatant by HPLC; then it was washed three times with deionized water to elute the methanol and acetic acid from the polymer. This stage had crucial importance in MIP synthesis because incomplete rinsing of the template makes difficult its rebinding into the polymer interaction sites. Drying of the solid polymers was performed at 55°C in an oven for 24 h; then the dried polymer was ground carefully and sieved. Non-imprinted polymer (**NIP**) as a control polymer was also prepared following the same procedure as MIP but in the absence of tartaric acid.

Adsorption study of the synthesized polymers by HPLC. To obtain a proper adsorption solution, three tartaric acid solvents including ethanol–water (50 : 50), water and 0.01 M KH_2PO_4 were tested as binding medium. Tartaric acid stock solution (10 mg/L) was used in preparing each solvent. Buffer solution gave the best results. Through varying buffer solution pH and ratios of polymers to binding solution the optimum solution condition was obtained.

Batch adsorption experiment has been developed to evaluate the binding capacity of the imprinted polymers toward tartaric acid. Considering the elementary studies mentioned before, 30 mg of polymer (MIP and NIP) and 2 mL of 10 ppm tartaric acid KH_2PO_4 buffer solution (0.01 M) were mixed in a 10 mL tube and horizontally stirred at 200 rpm for 1 h to achieve an equilibrium between polymer and template. Each solution was prepared 3 times. The mixture was then centrifuged at 10.000 rpm for 10 min. After syringe filtering (PTFE, 0.22 μm) of the supernatant, the concentration of free tartaric acid was determined by HPLC–UV. The amount of tartaric acid adsorbed onto the binding sites of the polymer was calculated from subtraction of free concentration from the initial concentration in supernatant. Selectivity test for imprinted polymer was performed in the presence of malic acid through the way mentioned in binding test. HPLC method details are represented below [17]:

C18 Hichrom 250 mm × 4.6 mm × 5 μm	Column
40 μL	Injection volume
0.5 mL/min	Flow rate
0.01 M KH_2PO_4	Mobile phase
7 min	Run time
25°C	Temperature
UV, 210 nm	Detector, wavelength
Water–methanol	Washing solution

Tartaric acid peaks were evaluated by their retention times by comparing the sample and standard spectra which obtained at 5.36 min. Quantification and linearity range of tartaric acid were obtained using a standard curve with 8 points. The linearity range was evaluated by plotting the peak area corresponding to each concentration in the range of 0.2 to 100 ppm which fitted the following equation: $A = 90.796c + 100.19$ with $R^2 = 0.9994$.

Spectroscopic analysis. Fourier transform infrared spectrometry was utilized to analyze the molecular structure of the MIP. Therefore, spectra of MIP after washing, relevant NIP, template and monomer were recorded by FTIR spectrometer. Before any sample scanning, KBr spectrum was recorded as control. For preparation of the samples for FTIR spectroscopic analysis, 1 mg of completely dried sample was mixed with about 150 mg of dry KBr [18]. A thin pellet of each sample was prepared by compressing the mixture under about 60 kPa within 10 min in a compress instrument. FTIR spectroscopy was run within a wavenumber range from 400 to 4000 cm^{-1} with a resolution of 0.5 cm^{-1}.

Particle size and zeta potential analysis. Particle size of the synthesized polymers (diameter and width) was determined by dynamic light scattering (**DLS**) technique which measures random changes in the intensity of light scattered from a suspension or solution. Both particle size and zeta potential experiments were performed in Zetasizer. Deionized water was used for dispersing the polymer particles. About 10 mg of the dried powder was diluted with 1 mL of water. After sonication and filtration, the suspension was poured in a cuvette for analysis.

Surface morphologic analysis of the polymer. Scanning electron microscopy (**SEM**) was applied for the investigation of shape and surface morphology of the polymer. The particles were sputter-coated with gold for a few minutes before SEM analysis. SEM measurement was conducted at an accelerating voltage of 15 kV.

Surface area and porosity measurements of the polymers. The analysis of surface area, mean pore diameter and pore volume of the polymers (MIP and NIP of the AA-based polymer) were determined from the nitrogen adsorption–desorption analysis at 77 K, using the BET method. Before the measurements, about 0.1 g of powdered samples were degassed at 393 K for 24 h to remove adsorbed gases and moisture.

Thermogravimetric analysis (TGA). Approximately 10 mg of sample was heated at a constant rate of 10 grad/min in the temperature range of 0–1000°C. Before scanning the changes, the atmosphere was made inert by purging with nitrogen gas.

Swelling test. The swelling ratio of MIP was performed in three solvents: 0.01 M KH_2PO_4, water–ethanol (50 : 50), and acetonitrile–ethanol (80 : 20). With this purpose, 30 mg polymer was incubated in 3 mL of each solvent for 2 h while stirring. After centrifuging at 11.000 rpm for 10 min, the supernatant was removed and the weight of swollen polymer was quickly measured [19, 20]. The swelling ratio was calculated as follows:

$$\begin{aligned} &\text{Swelling ratio} = [\text{weight (swollen polymer)} \\ &- \text{weight (drypolymer)}]/\text{weight (dry polymer)}. \end{aligned} \qquad (1)$$

Preparation of grape juice. Keeping in mind the high concentration of tartaric acid in grape juice (about 2000 ppm) [4], fruit juice sample was diluted 200 times with 0.01 M KH_2PO_4 solution. An aliquot of 2 mL diluted red grape juice was added to 30 mg polymer and the mixture was stirred horizontally at 200 rpm for 1 h. The mixture was then centrifuged at 10000 rpm for 10 min, the supernatant was filtered through a 0.22 μm membrane to remove any particles and was immediately injected into the separation system.

RESULTS AND DISCUSSION

HPLC analysis was used for evaluating the adsorption capacity indicating the effectiveness of imprinting process. Primary experiments revealed that 0.01 M buffer solution with pH 4.7 as adsorption medium improved the binding ability of polymers in comparison with ethanol–water (50 : 50) and water. HPLC results showed better adsorption of tartaric acid on

Table 1. Adsorption amount of different synthesized polymers toward tartaric acid

Code	Binding amount (%) ± SD	Initiator, g	Cross-linker, mmol	Monomer (1.2 mmol)	Template, mmol
MIP 1	26 ± 1	0.05	6	AA	0.3
MIP 2	37 ± 1	0.05	9	AA	0.3
MIP 3	14 ± 1	0.05	6	MAA	0.3
MIP 4	12 ± 2	0.05	9	MAA	0.3
NIP	Trace	0.05	9	AA	0

AA-based polymers compared to MAA-based polymers (Table 1).

According to Tadi and Motghare [9] and Subrahmanyam and Piletsky [11] when the template had hydrogen-bonding functional groups, acrylamide served as the best functional monomer in polar solvents, which was confirmed by the calculations of binding energies of template-monomer complex.

Considering cross-linker amount (Table 1), (1 : 4 : 30) TA–AA–EGDMA ratio gave the best results regarding to binding capacity. Quantitative parameters including the limit of detection, and limit of quantification, correlation coefficient (R^2) and relative standard deviation were determined for the extraction of tartaric acid in 10 ppm stock solution to be 0.07 mg/L, 0.2 mg/L, 0.9994 and 4.82% respectively.

Adsorption capacity of AA MIP toward TA, calculated from equation (2), was obtained to be 2.46 mg TA adsorbed per gram of synthesized MIP:

$$Q = (c_\mathrm{i} - c_\mathrm{f})V/m, \tag{2}$$

where V is the solution volume, L; m is the polymer mass, g; c_i and c_f are the initial and final (supernatant) TA concentrations in the binding medium, ppm [21].

Vasapollo et al. [16] also reported that by reducing the concentration of the template against cross-linker, the polymer binding properties were improved and the level of non-specific interactions was decreased in precipitation method. The NIP of the mentioned polymer showed no affinity to the template. Binding capacity was obtained from dividing the peak area of each sample to peak area of the relevant NIP.

Selectivity test was applied to evaluate the AA based polymer ability in selective separation of tartaric acid. For this purpose, an organic acid which is analogue in the structure named malic acid was chosen. Synthetized AA MIP gave the best binding capacity to tartaric acid (37%) compared to malic acid (5%). This could be due to structural resemblance of malic acid to tartaric acid. Selectivity (k) coefficient was obtained 11.1 which was calculated by the following equation:

$$k = k_{\mathrm{d\ (tartaric\ acid)}}/k_{\mathrm{d\ (malic\ acid)}}. \tag{3}$$

Distribution (k_d) coefficient was calculated by the following equation:

$$k_\mathrm{d} = (c_\mathrm{i} - c_\mathrm{f})/c_\mathrm{f} \times V/m, \tag{4}$$

where V is the solution volume, mL; m is the polymer mass, mg; c_i and c_f are the initial and final (supernatant) tartaric acid concentrations in the binding medium, ppm, respectively [21].

Rebinding test data showed low concentration of tartaric acid in desorption medium. It can be attributed to strong binding of TA to binding sites, which illustrates the importance of washing tartaric acid from the primary MIP. Schematic representation of TA and AA complexation via polymerization is shown in Fig. 1.

Spectrometric analysis. FTIR spectroscopy was applied to analyze the molecular structure of the imprinted polymer. The FTIR spectra of MIP (after washing), relevant NIP, functional monomer and the template are shown in Fig. 2. Both MIP and NIP had similar IR spectra indicating the similarity in the backbone structure. However, template and monomer complexation caused differences in MIP spectrum. The sharp adsorption peaks at 1610–1740 cm^{-1} were assigned to the carbonyl groups which included both carboxylic and amide groups. It can be seen that hydrogen bonded C=O in MIP had higher frequencies than that of acrylamide. The broad band at 3300–3500 cm^{-1} corresponded to the stretching vibration of hydrogen bonded –OH and –NH groups, while free –OH and –NH groups gave sharp peaks in this region, as seen in tartaric and acrylamide spectra. The peak at 2817 cm^{-1} corresponded to CH stretching of acrylamide which was transferred to higher frequencies in MIP. Therefore FTIR results indicated that tartaric acid interacted successfully with the functional monomer.

Particle size and zeta potential of the polymer. According to *ISO 14887-2000*, deionized water was utilized as dispersant solvent. The median particle sizes of TA-imprinted and non-imprinted particles were determined by DLS. The particle sizes were about 850 and 201 nm in diameter and 131 and 23 nm in width, respectively, for MIP and NIP, which was expected in precipitation polymerization. Polydispersity index which shows the particle size distribution and ranges from 0 to 1 in Zetasizer, was about 0.5. Based on zeta potential data, the polymer was negatively charged (–13.1 mV) which indicated the poor tendency of polymer particles to aggregation [22].

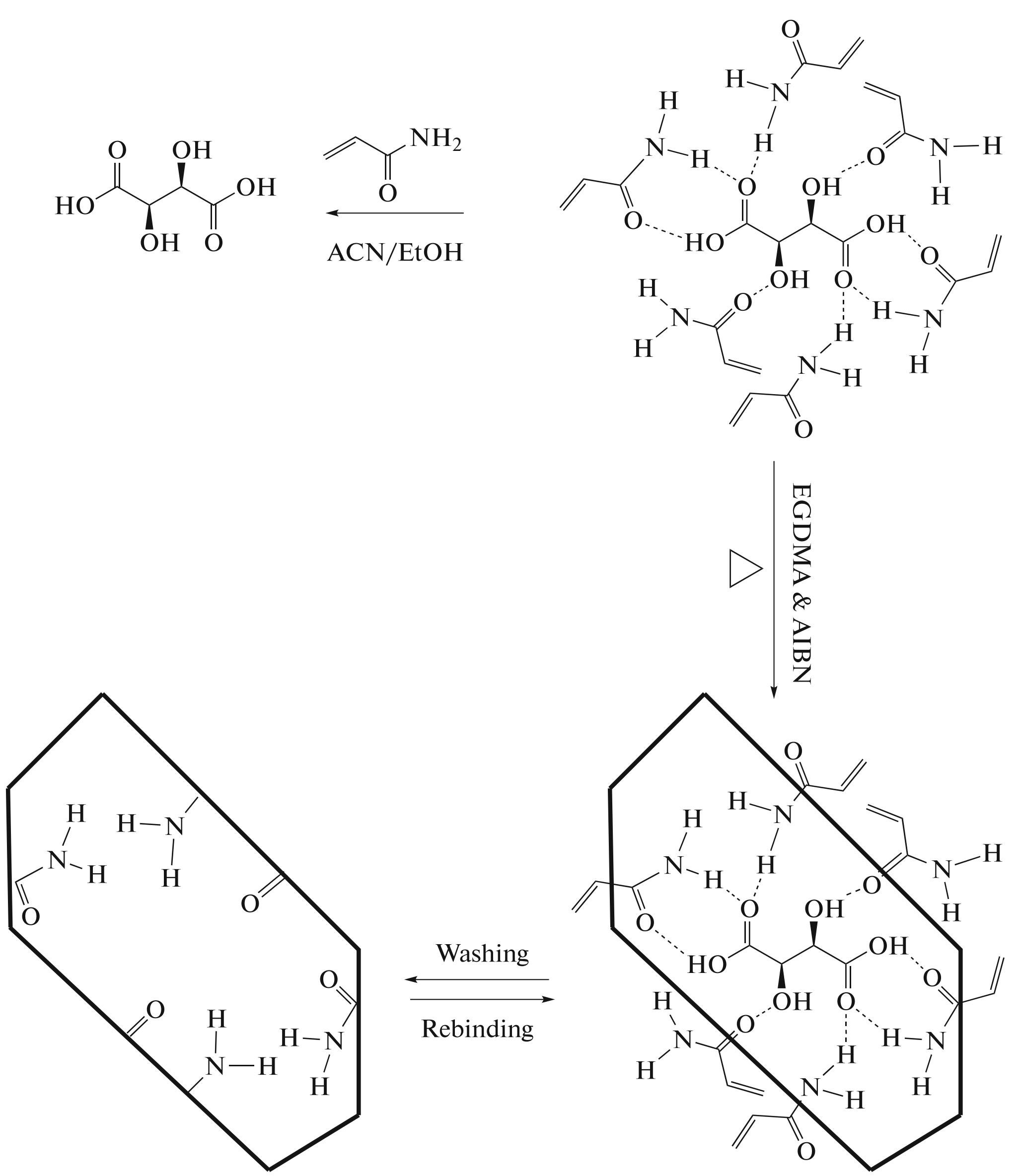

Fig. 1. Schematic representation of tartaric acid imprinting procedure with acrylamide.

Scanning electron microscopy of the imprinted polymer. SEM image of the imprinted polymer (Fig. 3) was taken at a magnification of 50000×. The polymer was obtained using AA and EGDMA at the ratios of 4 : 1 and 30 : 1 to template in a diluted acetonitrile–ethanol medium. As it can be seen in Fig. 3, the particles were spherically shaped.

Porosity properties. Nitrogen sorption porosimetry by using BET analysis is the main technique used to determine specific surface area and porosity properties of the polymer particles. Template adsorption to binding sites is proportional to the surface area of the polymer. Table 2 presents the surface area, mean pore diameter and pore volume of the AA based MIP and NIP, as calculated from the adsorption isotherms. As it can be seen, higher surface area, which is in the nor-

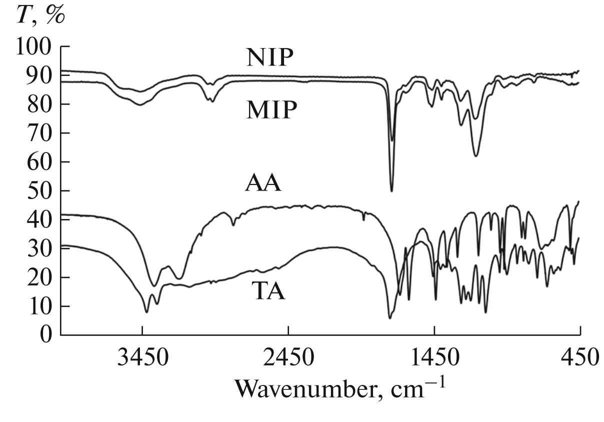

Fig. 2. FTIR spectra of MIP, NIP, tartaric acid (TA) and acrylamide (AA).

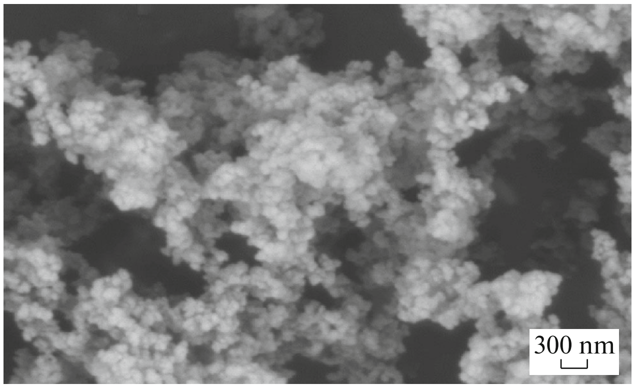

Fig. 3. Scanning electron microscopic image of the tartaric acid imprinted polymer obtained with TA–AA–EGDMA at the ratio of 1 : 4 : 30 in acetonitrile–ethanol (80 : 20).

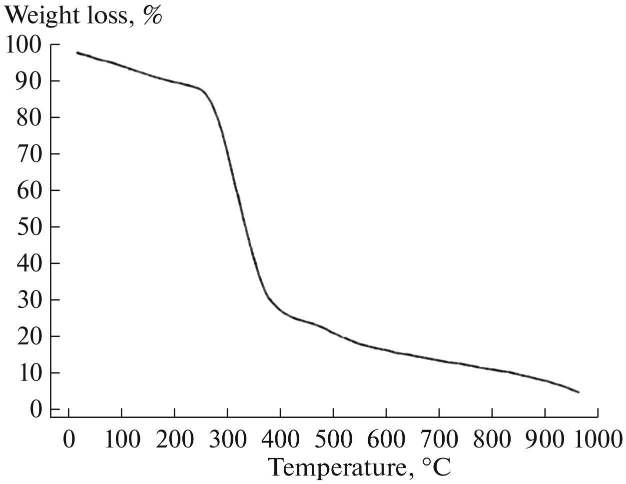

Fig. 4. Thermogravigram of TA-imprinted polymer.

mal range of 100–400 m^2/g was obtained for MIP particles in comparison with NIP. Furthermore, the pore volume of the MIP sample was higher than of the NIP one. These results showed that imprinting of tartaric acid have made the surface area larger and developed the porosity related to almost non-porous nature of non-imprinted polymer. The mean pore diameters of the MIP and NIP polymers were in the range of 2–100 nm, which place them as mesoporous particles [23].

Thermal analysis of the polymer. TGA is a thermal analysis technique used to characterize properties of materials like thermal stability and mass loss. Thermal analysis data depend on molecular weight, polymeric architecture, synthetic route and moisture content [24]. The TGA results showed that the tartaric acid imprinted polymer underwent thermal degradation beginning at 250°C and with a total mass loss of about 10%, which could be attributed to the release of solvent residues. Maximum polymer degradation was about 75%, as it can be seen in Fig. 4, which was achieved at 400°C [25].

Swelling ratio. Swelling test was conducted in three solvents: 0.01 M KH_2PO_4 solution (binding medium), water–ethanol (50 : 50) and acetonitrile–ethanol (80 : 20) (polymer synthesis media). Configuration of template cavities in the MIP structure introduced hydrodynamic volume into the polymer chain, which enhanced water penetration and resulted in higher water uptake [19, 20]. There was a significant difference in swelling ratios of polymer in different solvents; the swelling ratio in the binding medium (17%) was much higher than in the porogenic solvents (10%). According to Ye and Mosbach [26], the solvent which was applied in non-covalent MIP synthesis should be a non-solvent or non-swelling solvent. In this study, acetonitrile as porogen and ethanol as tartaric acid solvent were considered as porogenic solvent in MIP synthesis.

Table 2. Surface area, pore volume and mean pore diameter of the MIP and NIP

Polymer	Diameter, nm	Pore volume, m^3/g	Surface area, m^2/g
MIP	8.1	61	267
NIP	7.2	46	200

Application of the synthesized polymer in the separation of tartaric acid from grape juice medium. To investigate the separation potential of the imprinted polymer in a real medium, the prepared acrylamide MIP was incubated in diluted natural grape juice. The results showed 29% adsorption of tartaric acid with the prepared polymer from juice medium, which could be due to competing compounds (such as other organic acids, especially malic acid, amino acids and sugars). Binding capacity can be enhanced by optimizing the variables such as dilution level and pH of real medium.

CONCLUSIONS

According to experiments conducted in this study, acrylamide as functional monomer and ethylene glycol dimethacrylate as cross-linker in amounts higher than the template gave the best binding capacity. Acrylamide as the template having hydrogen-bonding functional groups served as the best functional monomer, which was also reported in literature. According to reports, by reducing the concentration of template against cross-linker in precipitation syntheses of MIPs, the polymer binding properties were improved and the level of non-specific interactions decreased. The critical point of this polymerization method was the elution stage of template from polymer. The prepared MIP can be utilized for the separation of tartaric acid in sensors or in solid phase extraction for improving the selectivity of sample preparation. Synthesizing the MIP with new surface imprinting techniques was proposed to facilitate the adsorption and desorption stages.

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