Homo- and copolymerization of 2-benzothiazoylacrylamide with α-methyl styrene and methyl acrylate: synthesis, characterization and reactivity

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DOI: https://doi.org/10.14314/polimery.2022.10.3

Abstract: 2-benzothiazolyl acrylamide (BTA) monomer was synthesized by reacting of 2-aminobenzothiazole with acryloyl chloride. Homo- and copolymerization of this monomer with α-methylstyrene (MSt) and methyl acrylate (MA) was carried out in dimethylsulfoxide (DMSO) solution. The prepared homo-and copolymers were characterized by Fourier Transform Infrared (FTIR) and their thermal stability was studied by thermogravimetric analysis (TGA). The composition of copolymers and the reactivity coefficients (r_1, r_2) were determined on the basis of sulfur analysis using linearization methods proposed by Fineman-Ross and Kelen-Tudos and the Mayo-Lewis intersection method. The derived (r_1, r_2) for BTA-*co*-MSt and BTA-*co*-MA are: (0.197, 0.230) and (0.294, 4.314), respectively. The microstructure of the copolymers and the distribution of the monomer sequences in the copolymers were calculated statistically, the results obtained are consistent with the values (r_1, r_2). BTA with MSt form alternative copolymer whilst BTA-*co*-MA tends to block with MA.

Keywords: 2-benzothiazoylacrylamide, α-methyl styrene, methyl acrylate, homocopolymerization, reactivity.

Homo- i kopolimeryzacja 2-benzotiazoliloakrylamidu z α-metylostyrenem i akrylanem metylu: synteza, charakterystyka i reaktywność

Streszczenie: Monomer 2-benzotiazoliloakryloamidowy (BTA) otrzymano w reakcji 2-aminobenzotiazolu z chlorkiem akryloilu. Homo- i kopolimeryzację tego monomeru z α-metylostyrenem (MSt) i akrylanem metylu (MA) przeprowadzono w roztworze dimetylosulfotlenku (DMSO). Otrzymane homo- i kopolimery scharakteryzowano przy użyciu spektroskopii w podczerwieni z transformacją Fouriera (FTIR), a ich stabilność termiczną metodą analizy termograwimetrycznej (TGA). Skład kopolimerów i współczynniki reaktywności (r_1, r_2) określono na podstawie analizy siarki metodami linearyzacji zaproponowanymi przez Finemana-Rossa i Kelena-Tudosa oraz metodą przecięcia Mayo-Lewisa. Pochodne (r_1, r_2) dla BTA-*co*-MSt i BTA-*co*-MA wynoszą odpowiednio (0,197, 0,230) i (0,294, 4,314). Mikrostrukturę kopolimerów i rozkład sekwencji monomerów w kopolimerach obliczono statystycznie, otrzymane wyniki są zgodne z wartościami (r_1, r_2). BTA z MSt tworzą alternatywny kopolimer, podczas gdy BTA-*co*-MA ma tendencję do blokowania z MA.

Słowa kluczowe: 2-benzotiazoiloakrylamid, α-metylostyren, akrylan metylu, homokopolimeryzacja, reaktywność.

Copolymerization is a process of chain polymerization, where a combination of two monomers can be used to undertake copolymerization, which results in the formation of polymeric products with different compositions within the polymer chain that are called copolymers [1, 2]. The relative concentrations and reactivity of the two monomers establish the proportions in which they are introduced into the copolymer [3]. In order to determine r_1 and r_2, in principle , it is only necessary to take two monomer mixtures of known composition, polymerize them to a conversion less than 15% and isolate, purify and analyze the products. These results will give two correlations, in the form of the following equation between the two unknowns r_1 and r_2, which can be solved to give r_1 and r_2 [4, 5]:

$$\frac{d[M_1]}{d[M2]} = \frac{[M_1](r_1[M_1]+[M_2])}{[M_2]([M_1]+r_2[M_2])} \quad (1)$$

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In practice, such a procedure is not reliable, because of experimental errors which arise in the copolymers analyses, and it is necessary to prepare copolymers from monomer mixtures covering a wide, and preferably the whole, range of possible composition. Such copolymerization yields a series of the mole fractions values [M_1] and [M_2] of the two monomers in the monomer mixture together with the corresponding values of the mole fractions d[M_1] and d[M_2], in the copolymer. Several methods have been proposed to reach a best fitting r_1, r_2 pair from a set of [M_1], [M_2] and d[M_1], d[M_2] pairs.

Thionyl and its derivatives [6, 7] show an essential role in the making of pharmaceutical compounds [8] and disperse dyes [9–11]. Aminothiazoles' biological activity has been widely documented [12–14]. They have wide applications in the treatment of hypertension [15], bacterial infections [16], schizophrenia [17], HIV [18], inflammation [19], and allergies [20]. The amide group in their derivatives is highly polar, which confirms it's hydrophilic properties and has high affinity for many large and small molecules that are known to be good hydrogen-bond acceptors. Many studies have reported on the copolymerization and reactivity relationships of acrylamide derivatives with other monomers such as acrylic acid, 2-acrylamido-2-methylpropane sulfonic acid, itaconic acid, acrylonitrile, methacryloxyethyltrimethyl ammonium chloride, and methyl methacrylate [21–26]. On the other hand, methacrylate derivatives are attracting much attention and have been copolymerized with other monomers including *N*-2-anisylmaleimidem, naphthylacrylamide, butyl acrylate, maleic anhydride, and *N*-(4-carboxyphenyl) maleimide [27–31].

In this study, hydrophobic monomers (MSt and MA) and hydrophilic synthesized monomer (BTA) were selected to prepare copolymers at low conversion. This study also calculates the reactivity ratios of BTA, MSt, and MA. So far there has been no investigation on the preparation and study on the reactivity correlations of 2-benzothiazoylacrylamide with α-methyl styrene and methyl acrylate.

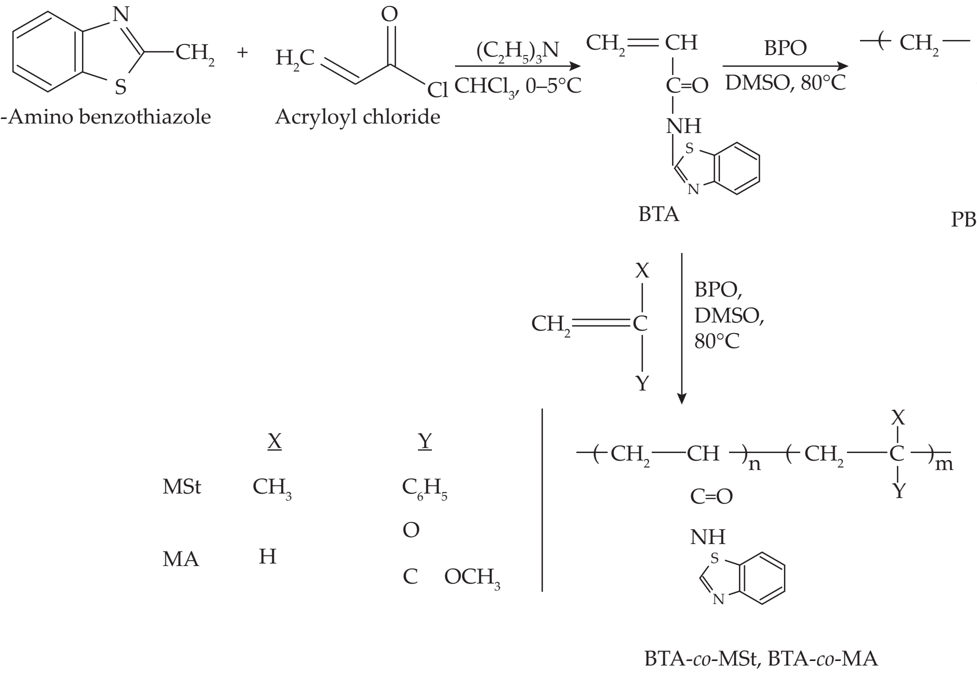
EXPERIMENTAL PART

Materials

2-aminobenzothiazole and acryloyl chloride were provided by (Aldrich-oma chemical Co). α- methyl styrene and methacrylate (Merck chemical Co) were shaken 2–3 times with 10% NaOH to eliminate hydroquinone inhibitor, then dried over anhydrous $CaCl_2$ for 10 h. Benzoyl peroxide (Aldrich-oma chemical Co) was recrystallized twice from methanol before use. Chloroform, dimethylsufoxide, triethyl amine, carbon tetrachloride, and anhydrous magnesium sulfate, were all obtained from (Aldrich-oma chemical Co) and used as received.

Methods

Perken Elmer-1650 spectrophotometer completed with data processing facilities was used to determine the functional groups in the BTA, PBTA, and copolymers using a KBr disk method at wavenumber range of 400 to 4000 cm^{-1}. Sulfur analysis (S %) were performed ~~by~~ using Elemental Analyzer CHNS-932 instrument. Thermal sta-



Scheme 1. Schematic reactions of BTA, PBTA synthesis and their copolymers with MSt and MA

bility of PBTA and copolymers was studied by TGA using Perkin Elmer. Ostwald viscometer was used to determine intrinsic viscosity [ŋ] according to the Solomon Gotessman relationship [32].

Synthesis of 2-benzothiazoyl acrylamide (BTA) monomer

In a three necked round bottom flask equipped with thermometer, dropping funnel, and stirrer, 15 g of 2 amino benzothiazole (0.1 mol) dissolved in 50 mL of chloroform and 15 mL of triethyl amine were introduced. Then the flask was cooled to 0–5°C and the acryloyl chloride (8.12 mL, 0.1 mol) was added dropwise with continuous stirring. The bath was removed once all of the acryloyl chloride had been added, and stirring was continued at room temperature for about 6 hours. The BTA hydrochloride salt was removed and rinsed with carbon tetrachloride. The solution was shaken with water three times and the separated organic layer was dried with anhydrous magnesium sulfate. After separating the solution from magnesium sulfate, carbon tetrachloride was partially distilled out under vacuum. The product was purified by recrystallization from 98% methanol as a brown powder (melting point 90–92°C, yield 81%).

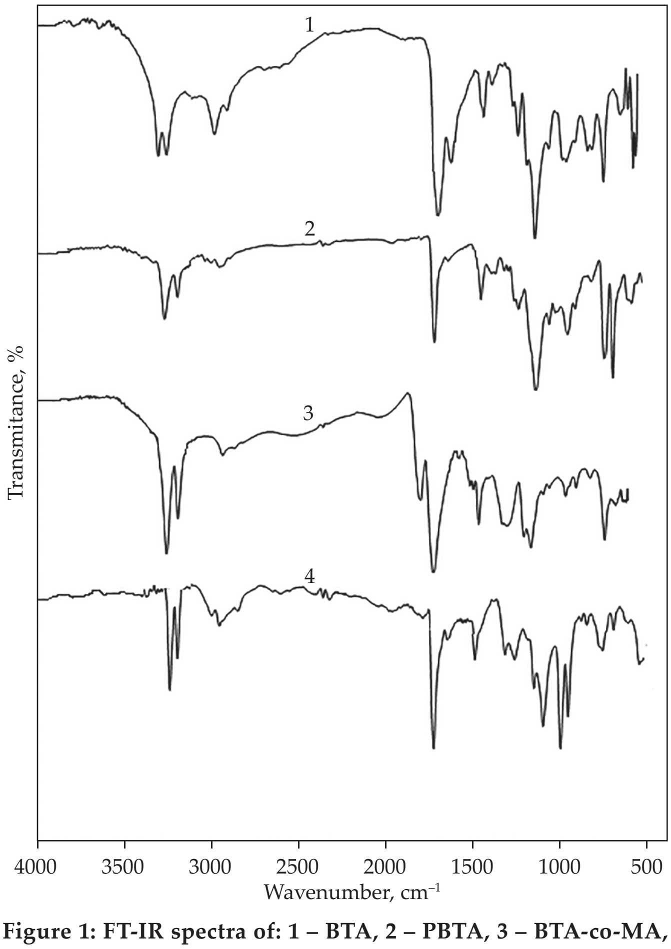


Figure 1: FT-IR spectra of: 1 – BTA, 2 – PBTA, 3 – BTA-co-MA, 4 – BTA-co-MSt

Polymerization process

The copolymerization were carried out in glass quick fit test tubes at 80°C using BPO at an overall concentration of 0.001 mol dm^{-3} as initiator and dimethylsufoxide (DMSO) as solvent. As shown in Table 1, five different feed compositions were used, the initial mole fractions of all the monomers (BTA, MSt and MA) are lying within the range (0.1–0.9). In order to remove all oxygen which may inhibit polymerization, nitrogen gas was bubbled through the mixture for 15 minutes before the reaction. The reaction was initiated by placing the tubes in a thermostatic water bath at 80°C. The copolymers were isolated by precipitation in methanol. After polymerization, the prepared copolymers were washed with methanol three times to remove impurities left from the preparation process. These include unreacted monomers, initiator and other products produced by side reactions. Copolymers were dried in vacuum at 40°C until constant weight. Calculations were then performed in order to determine the percent conversion of the copolymers. All the copolymers that were prepared in this study were produced during the first 15% monomer conversion and the produced copolymers that had a higher value than 15% conversion were set aside and not used in the current study on reactivity ratios. Poly BTA was synthesized following the same procedure mentioned above. To confirm the obtained results, the preparation and characterization by FTIR and nitrogen analysis, of each composition, were repeated three times. The reactions of synthesizing BTA, PBTA, and its copolymers with MSt and MA are shown in Scheme 1.

RESULTS AND DISCUSSION

The structures of BTA, PBTA, BTA-*co*-MSt, and BTA-*co*-MA are confirmed by FITR as shown in Figure 1. The absorption bands which appear in the FTIR spectra of BTA and PBTA are near symmetrical, and as follow: 3340 cm^{-1} (amide N-H), 3118 cm^{-1} (aromatic C-H), 2905 cm^{-1} (alkane C-H), 1681 cm^{-1} (secondary amide C=O), 1453 cm^{-1} (aromatic C=C), and 1200 cm^{-1} (amide C-N). FTIR spectrum of PBTA was characterized by absence of the band that appears in BTA spectrum at 1610 cm^{-1} (related to aliphatic C=C) indicating a complete homopolymerization. In both BTA-*co*-MSt, and BTA-*co*-MA, the complete copolymerization is confirmed by the absence of band at 1610 cm^{-1} corresponding to aliphatic C=C. For BTA-*co*-MA, there is a band at 1760 cm^{-1}corresponding to ester C=O, 3351 cm^{-1} (amide N-H), 3119 cm^{-1} (aromatic C-H), 2901 cm^{-1} (alkane C-H), 1675 cm^{-1} (secondary amide C=O), 1456 cm^{-1} (aromatic C=C), and 1198 cm^{-1} (amide C-N).

Copolymer composition

Characterization of copolymers samples requires determining the mole fractions (F_1) of comonomer in the copo-

T a b l e 1. Feed and copolymer compositions, conversion, sulfur analysis and intrinsic viscosity of BTA/MSt and BTA/MA copolymers

Sample	f_1 (feed)	Conversion %	S %	F_1 (copolymer)	Intrinsic viscosity dL/g
BTA/MSt-1	0.10	11.2	6.71	0.294	0.87
BTA/MSt-2	0.30	12.5	9.75	0.488	0.95
BTA/MSt-3	0.50	13.3	10.03	0.510	1.17
BTA/MSt-4	0.70	14.1	11.52	0.615	1.25
BTA/MSt-5	0.90	14.6	13.98	0.826	1.33
BTA/MA-1	0.10	11.3	1.98	0.055	0.96
BTA/MA-2	0.30	13.4	4.97	0.157	0.92
BTA/MA-3	0.50	11.8	8.03	0.305	0.89
BTA/MA-4	0.70	12.7	10.95	0.492	1.08
BTA/MA-5	0.90	13.1	13.66	0.759	1.02

f_1 – is the mole fraction of monomer-1 (BTA) in the initial feed; $f_2 = 1 - f_1$
F_1 – is the mole fraction of monomer-1 (BTA) in the copolymer; $F_2 = 1 - F_1$

lymer composition, which were obtained using elemental analysis to determine the (S %), in order to determine the BTA incorporated into the copolymer as follow [33]:

$$\frac{S\%\text{Copolymer}}{S\%M_1} = \frac{M.wt \text{ of } M_1}{M.wt \text{ of } M_1 + M.wt \text{ of } M_2/F} \qquad (2)$$

Where M_1 is BTA and $F=F_1/F_2$ is the molar ratio in the copolymer. The mole fraction of BTA can be calculated as: $F_1 = F/1 + F$. Results of (S %) analysis, the copolymer composition, and the values of intrinsic viscosity are listed in Table 1. Figure 2 shows the plots of BTA mole fraction in the feed (f_1) versus that of BTA mole fraction in the copolymer (F_1). In BTA/MSt system, BTA forms alternative copolymer with MSt. On the other hand, BTA/MA copolymer tends to block with MA units. Although M_w of BTA (204 g/mol) is much higher than M_w of MA (85 g/mol), the viscosity of all compositions is not affected because their copolymer is rich in MA units. In case of BTA/MSt copolymer, since this copolymer tend to be alternative, viscosity values increase as BTA increases in the feed.

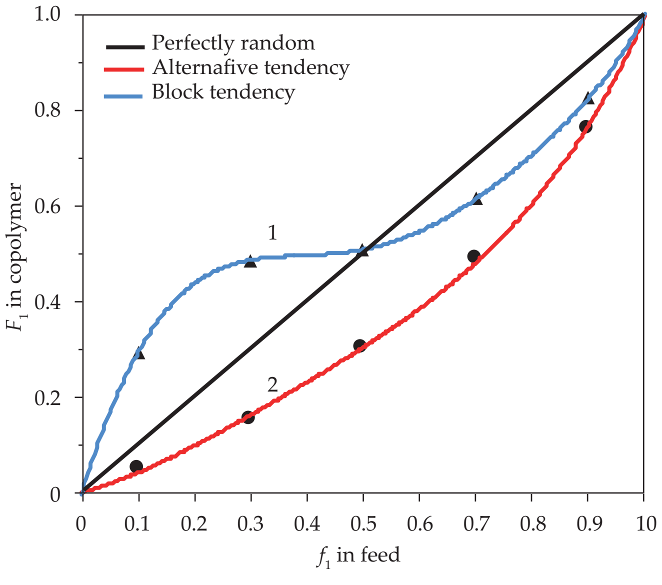


Figure 2: Variation of feed composition f_1 (BTA) with copolymer composition F_1 (BTA) of: 1 – BTA/MSt, 2 – BTA/MA copolymers

Reactivity ratios

Reactivity ratios are an important parameter for describing copolymerization characteristics. The monomer reactivity ratios for the copolymers BTA/MSt and BTA/MA were determined from the amount of comonomer in the feed ratios and the amount of comonomer incorporated into the copolymer composition. The weight of different monomer feed for BTA, MSt, and MA was converted into molar fractions for use in the least square method. The mole fraction of the comonomer-1 incorporated into the monomer feed is represented by (f_1) whilst the mole fraction of the comonomer-1 incorporated into the copolymer is represented by (F_1). For the best fitting of (r_1 & r_2) pair from a set of $[M_1]$, $[M_2]$, $d[M_1]$ and $d[M_2]$ pair, three procedures have been employed: linearization methods repre-

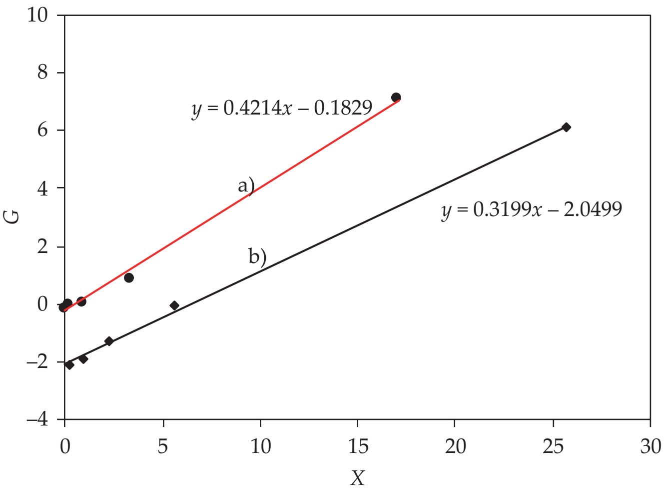


Fig. 3. Fineman-Ross plots of: a) BTA/MSt, b) BTA/MA copolymers

T a b l e 2. Fineman-Ross, Kelen-Tudos, and Mayo-Lewis parameters of BTA/MSt, BTA/MA copolymers

Sample	G $f(F-1)/F$	X f^2/F	η $G/(\alpha + X)$	ζ $X/(\alpha + X)$	f	$1/F$
BTA/MSt-1	-0.155	0.028	-0.215	0.038	0.111	2.403
BTA/MSt-2	-0.021	0.192	-0.023	0.217	0.428	1.049
BTA/MSt-3	0.038	0.961	0.023	0.581	1.000	0.961
BTA/MSt-4	0.864	3.391	0.211	0.830	2.300	0.628
BTA/MSt-5	7.106	17.08	0.401	0.964	9.000	0.210
BTA/MA-1	-2.111	0.249	-0.772	0.088	0.111	20.00
BTA/MA-2	-1.872	0.981	-0.540	0.283	0.428	5.376
BTA/MA-3	-1.272	2.270	-0.267	0.477	1.000	2.272
BTA/MA-4	-0.071	5.571	-0.132	0.691	2.300	1.033
BTA/MA-5	6.131	25.72	0.217	0.910	9.000	0.318

$\alpha = (X\text{min} \times X\text{max})^{0.5} = 0.691$ and 2.48 for BTA-*co*-MSt and BTA-*co*-MA, respectively, $f = f_1/f_2$, $F = F_1/F_2$

sented by Kelen-Tudos and Fineman-Ross, and intersection method proposed by Mayo-Lewis. For more details about the mathematical processes, the references [34-36] of the three methods should be consulted ,all the data are tabulated in Tables 2 and plotted in Figures 1–3.

The values of reactivity ratios calculated by various methods are listed in Table 3, the values are very close. In case of BTA/MSt copolymer, the reactivity of BTA (0.413) is of the same order as reactivity of MSt (0.183) due to the presence of the amide and aromatic groups for each BTA and MSt units. These active groups cause a significant attraction of free electron in the double bond and generate a positive charge in the growing polymer chain and stabilization of the corresponding macroradicals. Since both monomers are electron rich, they form the bond easily with electron deficient species, thus they are easily involved in polymerization. For this reason attendance to alternative copolymer forming can be postulated; that is, both r_1 and r_2 are between 0 and 1 in addition to the value of r_1r_2 which is less than 1. On the other hand, in case of BTA/MA copolymer, a high amount of incorporation of MA monomer is observed, which is in accordance with the previous report of Hegazy, et al., [33] who showed high incorporation of methyl methacrylate into the *N*-(2-thiazolyl)methacrylamide-methyl methacrylate copolymer chain. This can be interpreted in terms of steric hindrance effect of benzothiazoyl group in BTA which is higher than the effect of steric hindrance of the methyl group in MA unit.

Azeotropic copolymerization ($f_1(az.)$) often occurs, if feed and copolymer compositions are identical at the same point, which can be expressed by the following equation:

$$f_1(az) = (1 - r_2) / (2 - r_1 - r_2) \qquad (3)$$

If values of r_1, r_2 are both more or less than 1, the possibility of an azeotropic composition increases. For BTA-*co*-MSt, this condition is fulfilled since r_1 and r_2 are less than 1. Figure 2 proves this fact, in which a value of 0.5 for f_1 (*az.*) can be clearly observed. On the other hand, there is no azeotropic composition for BTA-*co*-MA because r_1< 1 and r_2> 1.

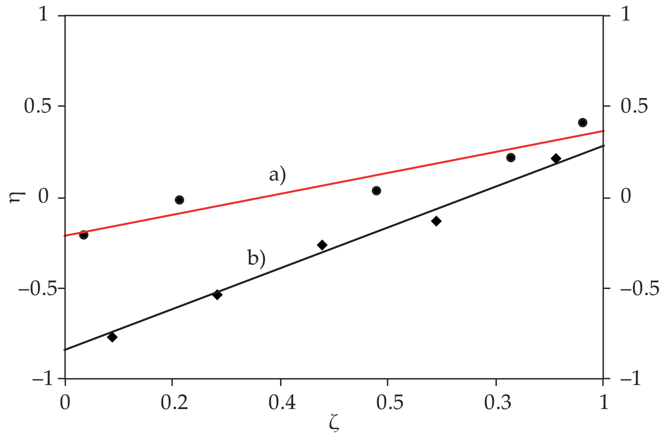


Fig. 4. Kelen-Tudos plots of: a) BTA/MSt, b) BTA/MA copolymers

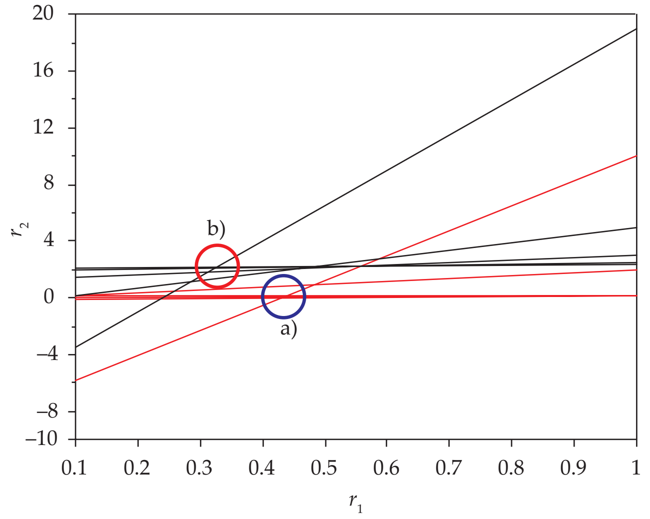


Fig. 5. Mayo-Lewis plots of: a) BTA/MSt, b) BTA/MA copolymers

T a b l e 3. Monomer reactivity ratios values of BTA/MSt, BTA/MA copolymers

Copolymer	Procedure	r_1	r_2
BTA/MSt	Fineman-Ross	0.421	0.182
	Kelen-Tudos	0.392	0.193
	Mayo-Lewis	0.427	0.174
	Average value	**0.413**	**0.183**
BTA/MA	Fineman-Ross	0.319	2.049
	Kelen-Tudos	0.287	1.989
	Mayo-Lewis	0.345	2.066
	Average value	**0.317**	**2.034**

Copolymer microstructure

The statistical distribution of the monomers (BTA, MSt and MA) in the formation of the copolymers BTA/MSt and BTA/MA is calculated using equation (4) [37], and listed in Table 4:

$$\begin{cases} S_{1-1} = F_1 - \dfrac{2F_1F_2}{1+\sqrt{(2F_1-1)^2+4r_1r_2F_1F_2}} \\ S_{2-2} = F_2 - \dfrac{2F_1F_2}{1+\sqrt{(2F_1-1)^2+4r_1r_2F_1F_2}} \\ S_{1-2} = \dfrac{4F_1F_2}{1+\sqrt{(2F_1-1)^2+4r_1r_2F_1F_2}} \end{cases} \tag{4}$$

Where S_{1-1}, S_{2-2}, and S_{1-2} represent the mole fractions of 1-1, 2-2, and 1-2 sequences, respectively.

The probabilities of finding the sequence of monomer-1 (BTA) and monomer-2 (MSt or MA) units are calculated by applying equation (5) [38, 39], and listed in Table 4:

$$\begin{cases} P_{11} = r_1[f_1]/(r_1[f_1]+[f_2]) \\ P_{22} = r_2[f_2]/(r_2[f_2]+[f_1]) \\ P_{12} = [f_2]/(r_1[f_1]+[f_2]) \\ P_{21} = [f_1]/(r_2[f_2]+[f_1]) \end{cases} \tag{5}$$

Where P_{11}, P_{12}, P_{21}, and P_{22} are the probabilities for forming the dyads, M_1M_1, M_1M_2, M_2M_1, and M_2M_2, respectively.

The average length sequences of monomer-1 (BTA) and monomer-2 (MSt or MA) are calculated using equation (6) [40], and tabulated in Table 4:

$$\begin{cases} \mu_1 = \dfrac{1}{P_{12}} \\ \mu_2 = \dfrac{1}{P_{21}} \end{cases} \tag{6}$$

In general, the results of both systems are in accordance with the values of r_1, r_2. In case of BTA/MSt copolymer, S_{1-1}, P_{11}, P_{21}, and μ_1 increase as BTA increases in the feed while S_{2-2}, P_{22}, P_{12}, and μ_2 increase as MSt increases. At the same time, values of S_{1-2} ranging from 0.329 to 0.784. From these results, it can be seen that both BTA and MSt have a tendency to react with the other monomer (BTA or MSt) in addition to itself in the growing chain to form alternative-block copolymer. On the other hand, for BTA/MA copolymer, the values S_{2-2} (0.937), P_{22} (0.948), P_{12} (0.965) and μ_2 (19) for BTA/MA-1 are much higher than S_{1-1} (0.562), P_{11} (0.740), P_{21} (0.815), and μ_1 (3.861) for the corresponding composition (BTA/MA-5). Moreover, S_{1-2} values are between 0.062 and 0.393. The results clearly indicate that MA prefers to react with itself rather than BTA to form block (MA) units distributed in the growing chain.

Thermal properties

Figure 6 shows the results of TGA of poly BTA, BTA-*co*-MSt, and BTA-*co*-MA. PBTA is more stable than both copolymers with 75% weight loss of PBTA at about 500 °C which is higher than 470°C and 450°C of BTA-*co*-MSt and BTA-*co*-MA, respectively. This result can be attributed to the presence of rigid amide group in the backbone of BTA side chain. In addition, it is found that increased amount of MSt and MA content in copolymers results in decreased thermal stability~~,~~ .This may be due to the pres-

T a b l e 4. Statistical data of BTA/MSt, BTA/MA copolymers

Sample		Blockness		Alternation	Sequence probability		Sequence length		
	S_{1-1}	S_{2-2}	S_{1-2}	P_{11}	P_{22}	P_{12}	P_{21}	μ_1	μ_2
BTA/MSt-1	0.015	0.425	0.560	0.043	0.622	0.956	0.377	1.046	2.652
BTA/MSt-2	0.096	0.121	0.783	0.150	0.299	0.849	0.700	1.177	1.428
BTA/MSt-3	0.118	0.098	0.784	0.292	0.154	0.707	0.845	1.414	1.183
BTA/MSt-4	0.262	0.039	0.699	0.490	0.072	0.509	0.849	1.964	1.177
BTA/MSt-5	0.659	0.012	0.329	0.788	0.019	0.211	0.956	4.739	1.046
BTA/MA-1	0.001	0.937	0.062	0.034	0.948	0.965	0.051	1.036	19.00
BTA/MA-2	0.018	0.703	0.279	0.119	0.825	0.880	0.174	1.136	5.740
BTA/MA-3	0.074	0.464	0.462	0.240	0.670	0.759	0.329	1.317	3.039
BTA/MA-4	0.214	0.230	0.556	0.426	0.465	0.574	0.406	1.742	2.463
BTA/MA-5	0.562	0.045	0.393	0.740	0.184	0.259	0.815	3.861	1.226

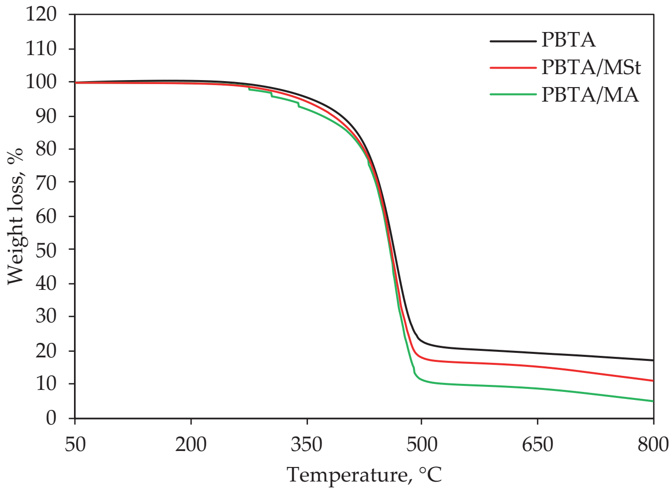


Fig. 6. TGA thermograms of PBTA, BTA-*co*-MSt and BTA-*co*-MA

ence of methyl groups in the backbone of MSt and MA side chain which significantly lowers the thermal stability of both copolymers.

CONCLUSIONS

The monomers BTA, PBTA, BTA-*co*-MSt, and BTA-*co*-MA were successfully prepared. FT-IR technique confirmed formation of the synthesized monomer and copolymers. Sulphur analysis test was employed to determine the copolymer compositions and the reactivity ratios by various procedures: Fineman-Ross, Kelen-Tudos, and Mayo-Lewis, an accordance was observed between the three methods. BTA-*co*-MSt tends to be alternative while BTA-*co*-MA form block copolymer with MA units. Copolymer microstructure results were calculated by statistical method and showed an accordance with the obtained reactivity ratios values. PBTA showed more thermal stability than its copolymers with MSt and MA.

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Received 13 VII 2022.