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Study of the reactions of fluorinated α,β-unsaturated carbonyl compounds with nitrogen and sulfur dinucleophiles

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Abstract

The fluorinated α,β-unsaturated ketone 1,1,1-trifluoro-4-ethoxy-3-butene-2-one reacted with dinucleophiles such as 2-aminothiophenol and 2-amino-ethanethiol to give trifluoroacetyl substituted 4H-1,4-benzothiazine, or 4H-1,4-thiazine, while the reaction of 5-trifluoroacetyl-3,4-dihydro-2H-pyran or 4-trifluoroacetyl-2,3-dihydro-furan with 2-amino-phenthiol gave 3-(2,2,3-2H-benzothiazolyl)-2-(trifluoromethyl)-tetrahydrofuran-2-ol or 3-(2-2,3-2H-benzothiazolyl)-2-(trifluoromethyl)- tetrahydro-2H-pyran-2-ol, respectively. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Fluorinated α,β-unsaturated carbonyl compounds; Fluorinated heterocycles; Dinucleophiles; Nucleophilic reactions; Thiazine

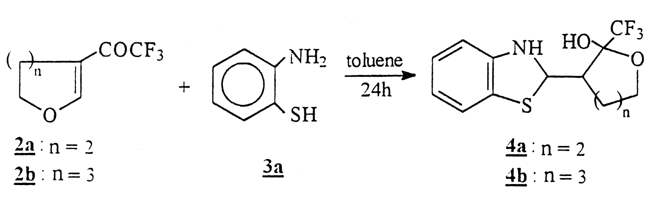
1. Introduction

In recent years, considerable attention has been concentrated on the development of new methodologies for the synthesis of various fluorine-containing heterocycles, since these compounds are now widely recognized as important organic materials showing interesting biological activities for their potential use in medicinal and agricultural scientific fields [1–4]. α,β-Unsaturated carbonyl compounds are of wide interest because of their possible applications in the preparation of various heterocycles [5,6]. In our previous work, we have reported the reactions of α,β-unsaturated ketones $EtOCH{=}CHCOCF_3$ (**1**) and $\overline{(CH_2)_nOCH{=}C}COCF_3$ (**2**) with some nitrogen nucleophiles [7,8]. Herein, we wish to report a convenient synthesis of fluorinated 4H-1,4-benzothiazine, 4H-1,4-thiazine and some other heterocycles from the reaction of the fluorinated α,β-unsaturated ketones with dinucleophiles such as 2-amino-phenthiol or 2-aminoethanethiol.

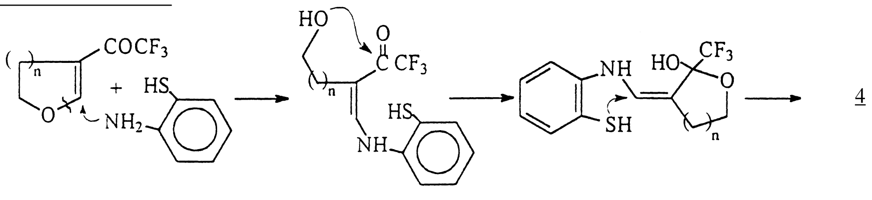
2. Result and discussion

5-Trifluoroacetyl-3,4-dihydro-2H-pyran (**2a**) or 4-trifluoroacetyl-2,3-dihydro-furan (**2b**) reacts with 2-aminothiophenol (**3a**) to give the corresponding fluorinated heterocycles **4a** and **4b**, respectively. Compound **4b** was easily recrystalized from petroleum ether:acetic ether (10:1) to give fine crystals for analysis. Their structures were fully characterized by spectroscopic methods and further confirmed by X-ray diffraction (XRD) analysis.

Fig. 1 shows the molecular structure of **4b**. Selected bond lengths and bond angles are listed in Table 1.



The possible reaction mechanism is as follows:



Under the same reaction conditions, 1,1,1-trifluoro-4-ethoxy-3-butene-2-one $EtOCH{=}CHCOCF_3$ (**1**) reacted smoothly with 2-aminothiophenol (**3a**) or 2-aminophenol

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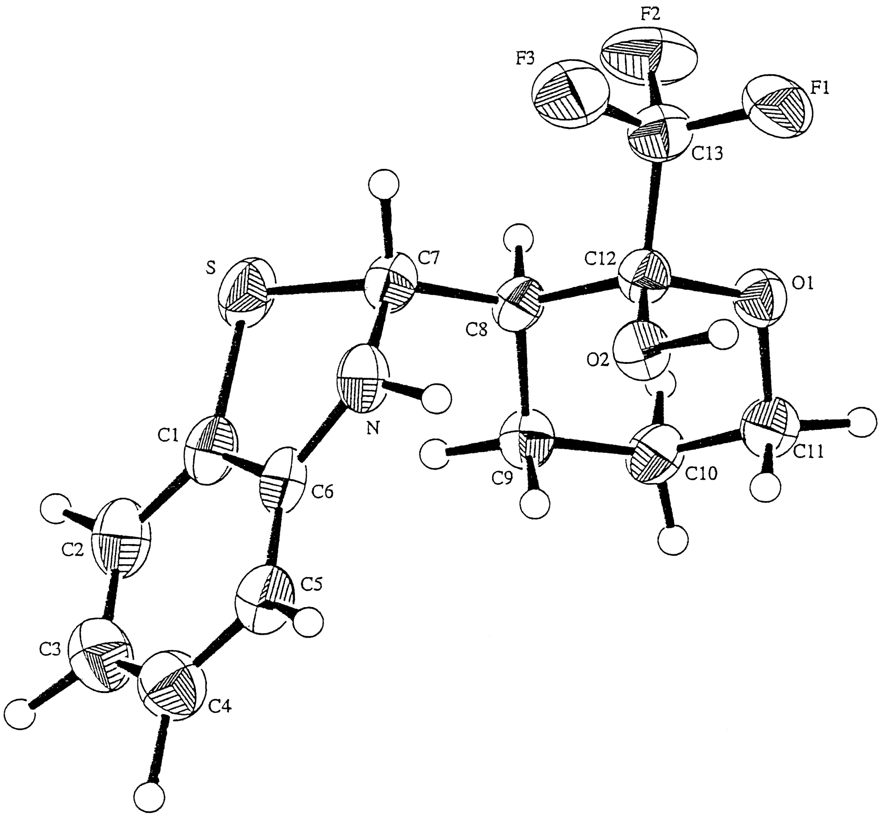


Fig. 1. The molecular structure of **4b**.

(**3b**) to form 4-(2-mercaptophenyl)amino-1,1,1-trifluoromethyl-3-butene-2-one (**5a**) or 4-(2-hydroxy-phenyl)amino-1,1,1-trifluoromethyl-3-butene-2-one (**5b**), respectively [8].

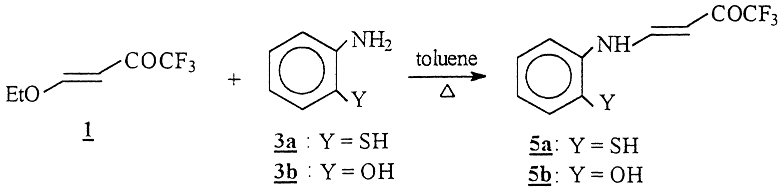
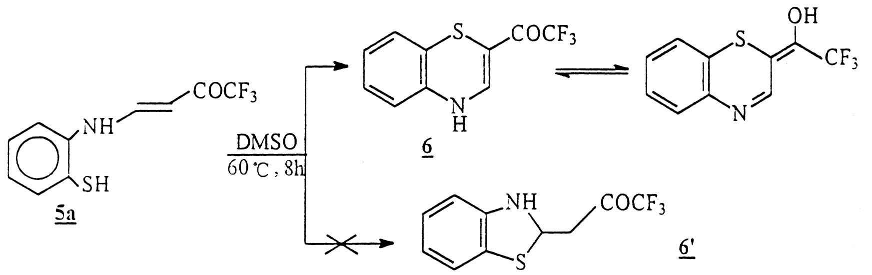


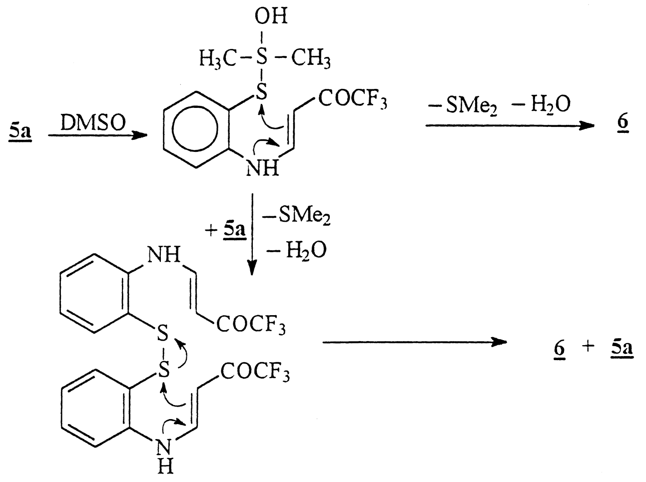
Table 1
Selected bond lengths and bond angles of compound **4b**

Bond length (Å)	Bond angle (°)
S–C(1) 1.755(5)	C(1)–S–C(7) 91.2(2)
S–C(7) 1.841(4)	C(6)–N–C(7) 111.9(3)
O(1)–C(11) 1.450(5)	S–C(7)–N 104.3(3)
O(1)–C(12) 1.410(4)	S–C(7)–C(8) 110.9(3)
O(2)–C(12) 1.399(4)	N–C(7)–C(8) 115.4(3)
N–C(6) 1.413(5)	C(9)–C(8)–C(12) 109.6(3)
N–C(7) 1.482(5)	C(7)–C(8)–C(12) 113.9(3)
C(12)–C(13) 1.534(6)	C(7)–C(8)–C(9) 113.4(3)
C(8)–C(12) 1.554(5)	C(11)–O(1)–C(12) 114.5(3)
C(7)–C(8) 1.540(5)	O(2)–C(12)–C(13) 109.3(3)

It is interesting that a further reaction occurred when **5a** was heated in DMSO. The product is a red solid 2-trifluoroacetyl-4H-1,4-benzothiazine (**6**) instead of the expected product 1,1,1-trifluoro-3-(2-2,3-2H-benzothiazolyl)-2-propanone (**6′**). Recrystalization of **6** from CH_3CN gave a good crystal for XRD analysis. The bond lengths of N(1)–C(4) 1.332(5), C(3)–C(4) 1.357(5), C(2)–C(3) 1.403(5) and C(2)–O(1) 1.226(4) showed that they are between single bond and double bond. This should be attributed to the delocalized conjugation π-bond system and the N–H–O intermolecular hydrogen bond (N–H, 0.95 Å; H–O, 1.80 Å). The molecular structure is shown in Fig. 2 and selected bond lengths and bond angels are summarized in Table 2.

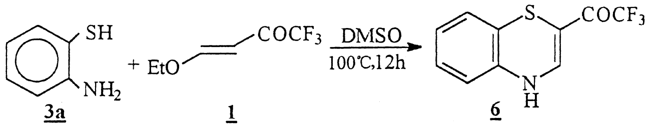


The conversion of the **5a** into **6** presumably proceeds by a mechanism analogous to that reported previously [9,10].



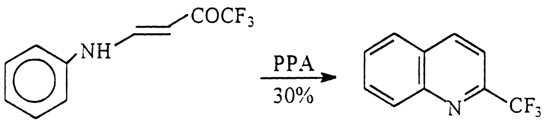
The conversion also takes place in DMF in good yield, however, no reaction occurred in CCl_4.

1,1,1-Trifluoro-4-ethoxy-3-butene-2-one (**1**) reacts directly with 2-aminothio-phenol (**3a**) to give **6** in 32% yield.

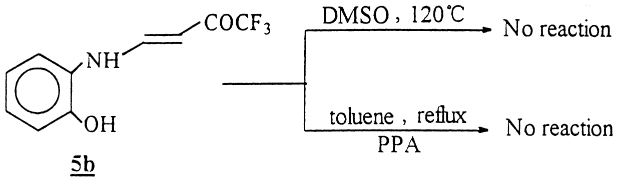


But 4-(2-hydroxyphenyl)amino-1,1,1-trifluoromethyl-3-butene-2-one (**5b**) is stable, i.e. does not cyclize in DMSO or HOAc at 120°C or in the presence of K_2CO_3.

Gorbunova [11] has reported that 2-trifluoromethylquinoline was the product of heating $C_6H_5NHCH{=}CHC(O)CF_3$ with polyphosphoric acid (PPA).



In our case, no analogous reaction is found.



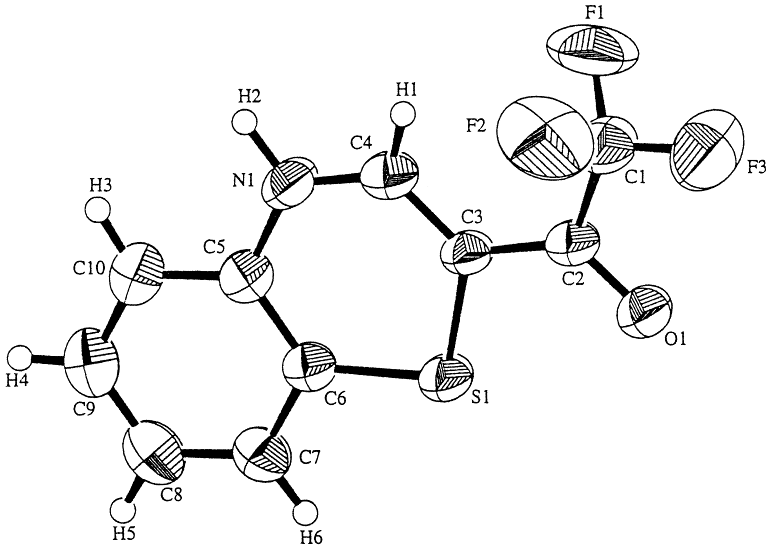
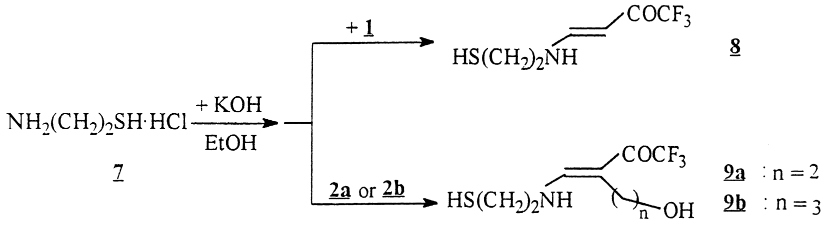


Fig. 2. The molecular structure of **6**.

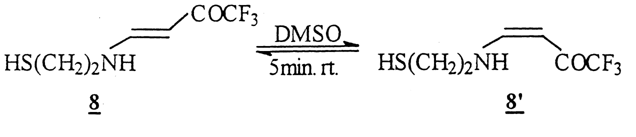
Table 2
Selected bond lengths and bond angles of compound **6**

Bond length (Å)	Bond angle (°)
S(1)–C(3) 1.774(3)	C(6)–S(1)–C(3) 101.4(2)
S(1)–C(6) 1.758(4)	C(5)–N(1)–C(4) 125.4(3)
N(1)–C(4) 1.332(5)	N(1)–C(4)–C(3) 126.0(3)
N(1)–C(5) 1.410(4)	S(1)–C(3)–C(4) 121.8(3)
C(4)–C(3) 1.357(4)	C(4)–C(3)–C(2) 125.2(3)
C(3)–C(2) 1.403(5)	S(1)–C(3)–C(2) 113.0(2)
C(2)–C(1) 1.530(5)	C(3)–C(2)–O(1) 123.5(3)
O(1)–C(2) 1.226(4)	C(3)–C(2)–C(1) 121.1(3)
C(5)–C(6) 1.383(5)	C(1)–C(2)–O(1) 115.3(3)

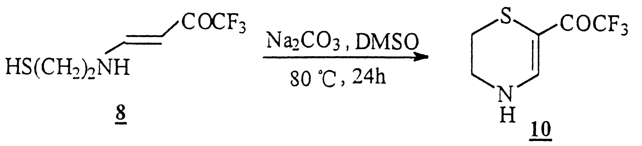
When α,β-unsaturated ketones (**1** and **2**) were treated with 2-amino-ethanethiol (**7**), only the *trans*-form of 1,1,1-trifluoro-4-(2-mercaptoethylamino)-3-buten-2-one (**8**), 1,1,1-trifluoro-5-hydroxy-3-((2-mercaptoethylamino)methylene)-2-pentanone (**9a**) or 1,1,1-trifluoro-6-hydroxy-3-((2-mercaptoethylamino)methylene)-2-hexanone (**9b**) were obtained in high yield, respectively.



It is worth noting that compound **8** is partially converted from the all *trans* isomer into the *cis*-form in DMSO but the same conversion did not occur in CCl$_4$. The ratio of *trans*- and *cis*-form is nearly 4:5. But the *cis*-form could not be isolated since it reverted to the *trans*-form when poured into water. The same conversion was not observed when we treated the compounds **9a** and **9b** in DMSO.



It is interesting that only in the presence of an inorganic base in DMSO, was 1,1,1-trifluoro-4-(2-mercaptoethylamino)-3-buten-2-one (**8**) converted to the 2-trifluoroacetyl-4H-1,4-thiazine (**10**). The reaction was carried out under several different conditions and the results were summarized in Table 3.



As mentioned above, compound **6** could be synthesized directly from the reaction of **1** with **3a**, however, under the same reaction conditions, **1** treated with 2-amino-ethanethiol (**7**) failed to give **10**. Similarly, treatment of compounds **9a** and **9b** in DMSO or HOAc led to decomposition.

In summary, we synthesized a series of fluorinated heterocycles such as trifluoroacetyl-substituted 4H-1,4-benzothiazine, 4H-1,4-thiazine, furan and pyran from the reactions of fluorinated α,β-unsaturated carbonyl compounds with some dinucleophiles.

3. Experimental

Melting points are measured on a Temp-Melt. apparatus and are uncorrected. ^{1}H NMR (90 MHz) and ^{19}F NMR (54.6 MHz) spectra were recorded on a Varian-360L instrument or Bruker AM-300 spectrometer with TMS and TFA (δCFCl$_3$ = δTFA — 76.8 ppm) as the internal and external

Table 3
Conversion of compound **8** in different conditions

Entry	Catalyst (mol%)	Condition			Yield of 10 (%)
		Solvent	Temperature (°C)	Time (h)	
1	HOAc (20)	DMSO	100	8	No reaction
2	- - - -	HOAc	120	8	No reaction
3	Et$_3$N (20)	DMSO	100	8	No reaction
4	Pyridine (20)	DMSO	100	8	No reaction
5	KOH (20)	DMSO	80	24	23
6	K$_2$CO$_3$ (100)	DMSO	80	24	38
7	K$_2$CO$_3$ (20)	DMSO	80	24	42
8	Na$_2$CO$_3$ (20)	DMSO	80	24	64

standard, respectively. IR spectra were obtained with an IR-440 Shimadzu spectrophotometer on KBr disks. Low resolution mass spectra was obtained on a Finnigan GC–MS 4021, an instrument. X-ray structure analysis was performed with Rigaku AFC 7R diffractometer. Elemental analyses were performed by this institute. Compound **1** was prepared according to the literature method [12].

*3.1. Reaction of **2** with 2-amino-phenthiol (**3a**)*

2-Aminothiophenol (**3a**, 5 mmol) was added to a 50 ml flask containing a solution of 5-trifluoroacetyl-3,4-di-hydro-2H-pyran (**2a**, 5 mmol) and toluene (30 ml). This reaction mixture was stirred for 24 h at room temperature, and then TLC was used to show that the reaction had finished. The solvent was evaporated in vacuum. The crude product was separated by column chromatography to give the pure product **4a** in a yield of 74% and by-product 2,2′-dithio-bis-benzenamine **11** which was identified by its spectral data [13] in a yield of 8%.

At room temperature, similar treatment of 2-amino-phenthiol (**3a**) with **2b** gave **4b** in a yield of 70% and **11** in a yield of 7%.

*3.1.1. 3-(2-2,3-2H-Benzothiazolyl)-2-(trifluoromethyl)-tetrahydro-furan-2-ol (**4a**)*

Mp: 120–122°C. Anal for $C_{12}H_{12}F_3NSO_2$ Calc.: N, 4.81%; H, 4.12%; C, 49.48%; found: N, 4.72%; H, 3.84%; C, 49.15%; IR (ν_{max}, cm^{-1}): 3310 (s, O–H), 3100 (s, N–H), 2948 (m, C–H), 1195, 1162 (vs, C–F); δH: (90 MHz, $D_3CC(O)CD_3$), 6.7 (m, 4H), 5.5 (d, 1H), 4.0 (m, 2H), 3.2 (br, 2H), 2.7 (m, 1H), 2.0 (m, 2H); δF: (60 MHz, $D_3CC(O)CD_3$), −83.8 (s, CF_3); MS (*m/z*, %): 291 (M^+, 6.94), 149 ($C_8H_7NS^+$, 4.17), 136 ($C_7H_6NS^+$, 100), 108 ($C_6H_4S^+$, 12.30).

*3.1.2. 3-(2-2,3-2H-Benzothiazolyl)-2-(trifluoromethyl)-tetrahydro-2H-pyran-2-ol (**4b**)*

Mp: 106–108°C. Anal for $C_{13}H_{14}F_3NSO_2$ Calc.: N, 4.59%; H, 4.59%; C, 51.15%; found: N, 4.65%; H, 4.63%; C, 51.32%; IR (ν_{max}, cm^{-1}): 3315 (s, O–H), 3102 (s, N–H), 2954 (m, C–H), 1274, 1142 (vs, C–F); δH: (90 MHz, $CDCl_3$), 6.9 (m, 4H), 5.8 (d, 1H), 4.8 (br, 2H), 3.8 (m, 2H), 2.0 (m, 1H), 1.7 (m, 4H); δF: (60 MHz $CDCl_3$), −80.8 (s, CF_3); MS (*m/z*, %): 306 (M^+–H, 2.42), 305 (M^+, 6.97), 136 ($C_7H_6NS^+$, 100), 108 ($C_6H_4S^+$, 13.18).

XRD analysis data: Crystal system: trigonal, lattice type: *R*-centered, no. of reflections used for unit cell determination (2θ range): 25 (18.3–21.6°), space group: $R\bar{3}$(#148), $Z = 18$, $a = 22.094(4)$ Å, $c = 15.235(3)$ Å, $V = 6435(2)$ Å^3, λ (Mo Kα) = 0.71069 Å, Dc = 1.553 g/cm^3. F(0 0 0) = 3114.00, $R = 0.052$, Rw = 0.061, μ (Mo Kα) = 2.72 cm^{-1}. Crystal dimensions: 0.20 mm × 0.20 mm × 0.30 mm. (Data were measured at 293 K on a Rigaku AFC 7R diffractometer with graphite monochromated Mo Kα radiation and a 12 kW rotating anode generator.)

*3.2. Reaction of **1** and **2** with 2-amino-ethanethiol*

2-Amino-ethanethiol hydrochloride **7** (10 mmol) was added into a 100 ml flask containing a solution of KOH (10 mmol) and EtOH (60 ml). After the mixture was stirred for 0.5 h at room temperature, 1,1,1-trifluoro-4-ethoxy-3-butene-2-one (**1**, 10 mmol) was added to the solution. After this reaction mixture was stirred for 8 h at room temperature, the solvent was evaporated in vacuum. The crude product was separated by column chromatography to give the pure product **8** in a yield of 94%.

Similar treatment of **2a** and **2b** gave the corresponding products **9a** in a yield of 88% and **9b** in a yield of 89%, respectively.

*3.2.1. trans-1,1,1-Trifluoro-4-(2-mercaptoethylamino)-3-buten-2-one (**8**)*

Mp: 109–110°C. Anal for $C_6H_8F_3NSO$ Calc.: N, 7.04%; H, 4.02%; C, 36.18%; found: N, 6.88%; H, 3.72%; C, 36.41%; IR (ν_{max}, cm^{-1}): 3235 (m, N–H), 1647 (s, C=O), 1593 (s, C=C), 1200, 1136 (vs, C–F); δH: (90 MHz, $CDCl_3$), 10.3 (br, NH), 7.2 (m, 1H), 5.4 (d, 1H), 3.7 (br, SH), 3.6 (m, 2H), 2.9 (m, 2H); δF: (60 MHz $CDCl_3$), −76.7 (s, CF_3); δH: (90 MHz, DMSO-d_6), 10.3 (br, NH), 7.5 (m, 1H), 5.4 (d, 1H), 3.7 (m, 2H), 3.0 (m, 2H), 3.0 (br, 1H); δF: (60 MHz, DMSO-d_6), −76.7 (s, CF_3); MS (*m/z*, %): 199 (M^+, 11.34), 152 (M^+–CH_2SH, 73.07), 130 (M^+–CF_3, 11.75), 102 (M^+–$COCF_3$, 30.86), 43 (CH_2CH_2NH, 100).

*3.2.2. cis-1,1,1-Trifluoro-4-(2-mercaptoethylamino)-3-buten-2-one (**8′**)*

δH: (90 MHz, DMSO-d_6), 8.7 (br, NH), 7.9 (m, 1H), 5.5 (d, 1H), 3.7 (m, 2H), 3.0 (m, 2H), 3.0 (br, 1H); δF: (60 MHz, DMSO-d_6), −76.65 (s, CF_3).

*3.2.3. 1,1,1-Trifluoro-5-hydroxy-3-((2-mercaptoethyl-amino)methylene)-2-pentanone (**9a**)*

Mp: 112–114°C. Anal for $C_8H_{12}F_3NSO_2$ Calc.: N, 5.76%; H, 4.94%; C, 39.51%; found: N, 5.68%; H, 4.81%; C, 39.34%; IR (ν_{max}, cm^{-1}): 3380 (s, O–H), 3210 (s, N–H), 2946 (m, C–H), 1648 (m, C=O), 1583 (s, C=C), 1182, 1136 (vs, C–F); δH: (90 MHz, $D_3CC(O)CD_3$), 7.3 (s, 1H), 3.4 (m, 2H), 3.2 (m, 2H), 3.2 (br, 3H), 2.6 (m, 2H), 2.1 (m, 2H); δF: (60 MHz, $D_3CC(O)CD_3$), −75.8 (s, CF_3); MS (*m/z*, %): 243 (M^+, 2.58), 242 (M^+–H, 3.04), 196 (M^+–CH_2SH, 6.90), 167 (M^+–$NHCH_2CH_2SH$, 2.22), 88 ($HSCH_2CH_2NHC^+$, 100), 69 ($CF_3{}^+$, 89.28).

*3.2.4. 1,1,1-Trifluoro-6-hydroxy-3-((2-mercaptoethyl-amino)methylene)-2-hexanone (**9b**)*

Mp: 118–120°C. Anal for $C_9H_{14}F_3NSO_2$ Calc.: N, 5.45%; H, 5.45%; C, 42.02%; found: N, 5.42%; H, 5.34%; C, 42.23%; IR (ν_{max}, cm^{-1}): 3380 (s, O–H), 3212 (s, N–H), 2948 (m, C–H), 1648 (m, C=O), 1585 (s, C=C), 1194, 1124 (vs, C–F); δH: (90 MHz, $D_3CC(O)CD_3$), 7.3 (s, 1H), 3.4 (m, 2H), 3.1 (m, 2H), 3.1 (br, 1H), 2.9 (br, 2H), 2.6 (m, 2H), 2.0

(m, 2H), 1.2 (m, 2H); δF: (60 MHz, $D_3CC(O)CD_3$), −75.8 (s, CF_3); MS (*m*/*z*, %): 257(M^+, 12.88), 256 (M^+–H, 37.10), 239 (M^+–H_2O, 8.51), 210 (M^+–CH_2SH, 75.91), 88 ($HSCH_2CH_2NHC^+$, 100), 69 ($CF_3{}^+$, 98.93).

3.3. Conversion of **5a** or **8** in DMSO

At room temperature, 4-(2-mercaptophenyl)amino-1,1,1-trifluoromethyl-3-butene-2-one (**5a**, 5 mmol) was added to a 25 ml flask containing 15 ml DMSO. After being stirred for about 8 h at 60°C, ^{19}F NMR indicated that the reaction was finished, the reaction mixture was poured into water. The crude product was separated by filtration and it was purified by column chromatography to give the pure red product 2-trifluoroacetyl-4H-1,4-benzothiazines (**6**) in a yield of 90%.

Similar treatment of 1,1,1-trifluoro-4-(2-mercaptoethylamino)-3-buten-2-one (**8**) in the presence of inorganic base o give the corresponding product 2-trifluoroacetyl-4H-1,4-thiazine (**10**).

3.3.1. 4-Trifluoroacetyl-4H-1,4-benzothiazine (**6**)

Mp: 182°C (decomposition temperature). Anal for $C_{10}H_6F_3NSO$ Calc.: N, 5.71%; H, 2.45%; C, 48.98%; found: N, 4.70%; H, 2.21%; C, 48.82%; IR (ν_{max}, cm^{-1}): 3238 (s, N–H), 1587 (m, C=O), 1560 (m, C=C), 1192, 1129 (vs, C–F); δH: (90 MHz, DMSO-d_6), 10.1 (br, NH), 7.5 (d, 1H), 7.0 (m, 4H); δF: (60 MHz, DMSO-d_6), −75.9 (s, CF_3); MS (*m*/*z*, %): 245 (M^+, 28.83), 244 (M^+–H, 100), 176 (M^+–CF_3, 21.39), 148 (M^+–$COCF_3$, 73.78), 69 ($CF_3{}^+$, 7.12).

XRD analysis data: Crystal system: triclinic, lattice type: primitive, no. of reflections used for unit cell determination (2θ range): 21(13.6–25.6°), space group: $P\bar{1}$(#2), $Z = 2$, $a = 10.688(2)$ Å, $b = 10.976(4)$ Å, $c = 10.272(3)$ Å, $\alpha = 111.12(3)$, $\beta = 95.54(2)$, $\gamma = 111.00(2)$, $V = 1013.5(6)$ Å^3, λ(Mo Kα) = 0.71069 Å, Dc = 1.607 g/cm^3. F(0 0 0) = 496.00, $R = 0.047$, Rw = 0.060, μ(Mo Kα) = 3.38 cm^{-1}. Crystal dimensions: 0.20 mm × 0.20 mm × 0.30 mm. (Data were measured at 293 K on a Rigaku AFC 7R diffractometer with graphite monochromated Mo Kα radiation and a 12 kW rotating anode generator.)

3.3.2. 4-Trifluoroacetyl-4H-1,4-thiazines (**10**)

Mp: 162–164°C. Anal for $C_6H_6F_3NSO$ Calc.: N, 7.11%; H, 3.05%; C, 36.55%; found: N, 7.06%; H, 2.80 1%; C, 36.22%; IR (ν_{max}, cm^{-1}): 3210 (s, N–H), 3020 (m, C–H), 1585 (m, C=O), 1560 (m, C=C), 1150, 1125 (vs, C–F); δH: (90 MHz, $D_3CC(O)CD_3$), 8.1 (s, 1H), 3.7 (m, 2H), 3.0 (m, 2H); δF: (60 MHz, $D_3CC(O)CD_3$), −75.9 (s, CF_3); MS (*m*/*z*, %): 198 (M^+–H, 19.26), 197 (M^+, 100), 128 (M^+–CF_3, 80.01), 100 (M^+–$COCF_3$, 42.45).

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