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Spectral LFER in some aryl azine derivatives

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ABSTRACT

About ten (*Z*)-1-(1-(substitutedphenyl)ethylidene)-2-(diphenylmethylene)hydrazines were prepared and their purities were examined by literature method. The infrared and NMR spectra of these hydrazines were recorded. The characteristics C=N and N-N stretches (ν, cm^{-1}) and the chemical shifts (δ, ppm) of methyl protons and carbons were assigned. These spectral have been correlated with Hammett substituent constants, F, R and Swain-Lupton's parameters using single and multi-regression analysis. From the results of statistical analyses, the effects of substituent on the spectral data have been studied.

***Keywords*:** Aryl hydrazines, IR spectra, NMR spectra, Hammett correlations, Swain-Lupton's parameters

1. INTRODUCTION

Azine derivatives containing –N–N– moieties in their structures [1]. These are good synthones for synthesis of heterocyclic compounds such as pyrimidines, pyrazolines and purines [2]. These classical azines possess numerous pharmaceutical [3] and biological activities [4] such as antibacterial [5, 6], antifungal [5, 6] anti-tuberculur [7] and EMT6 tumor [8]. Generally azine derivatives have been synthesized by reaction between carbonyl compounds and hydrazine hydrate in the presence of acetic acid in ethanol medium [9].

Many conventional methods have been reported for synthesis of azines [10, 11]. However, these conventional methods have some disadvantages such as huge amount of solvent consuming, long reaction time, low percentage of yields, formation of side products, difficulty of separation and handling.

Now-a-days greener methods employed for synthesis of azine derivatives with green catalysts [12-14]. Khouzani et al., have synthesized more than 90% yields of some azines from α, β-unsaturated ketones by solid phase reaction using alumina catalyst [12]. Shah and Chudgar have prepared some azines by thermolysis of semicarbazones. Malkin and Kuzmin have studied the photochemical properties of azines [3].

Carmeli and Rozen [15] have investigated the deprotection of azines by oxygen isotope labeling on carbonyl compounds. Kaladya et. al., have synthesized some metal complexes of azines and studied their anticancer activity [16]. Nekrasov reported the synthesis of aza heterocyclic compounds containing azine moieties [9]. These azines used as colorimetric chemosensors for detection of metal ions [17].

The physiochemical properties [18] such as dipole moments, potentiometric titration, vibrational spectroscopy of flat *s-trans* configuration of azines and the pKa values of symmetric and non-symmetrical benzalazines have been reported by Kitae et. al., the redox properties of ferrocenyl substituted azine derivatives was studied by Saure and Workentin [19]. Spectroscopic data were useful for prediction of geometry of organic compounds such as, *s-cis* and *s-trans* conformers of unsaturated carbonyl compounds, *E* and *Z* configuration of pyrazolines [20, 21].

These data also applied for studying QSAR, QPR, QSPR and SAR through Hammett equation [22]. Recently, Vijayakumar et. al., and Rajarajan et. al., have investigated the effect of substituents on some azine derivatives and they observed satisfactory correlations [5, 6].

Within the above view, there is no report available for spectral correlation on (*Z*)-1-(1-(substitutedphenyl)ethylidene)-2-(diphenylmethylene) hydrazines in the past. Therefore the author taken efforts for studying the effect of substituents on the spectral data of (*Z*)-1-(1-(substituted phenyl)ethylidene)-2-(diphenylmethylene) hydrazines by synthesis an recorded their infrared and nuclear magnetic resonance spectra.

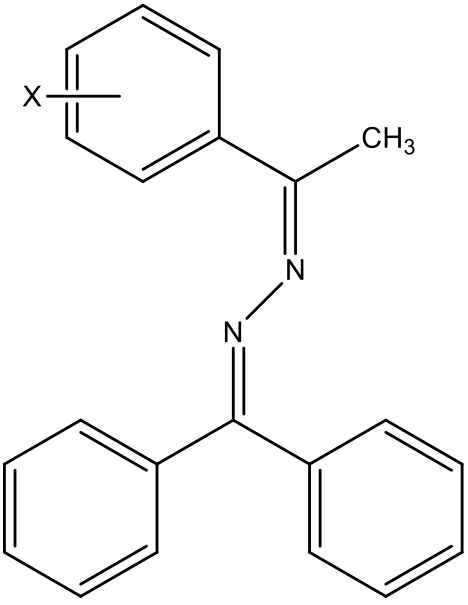
2. EXPERIMENTAL

2. 1. Genral procedure for synthesis of (*Z*)-1-(1-(substituted phenyl)ethylidene)-2-(diphenylmethylene) hydrazines [23]

An equi-molar quantities of benzophenone hydrazone (1 mmol) and substituted acetophenones (1 mmol) in dry media, 0.1 g of TiO_2–SO_4^{2-} was added and the mixture was ground in a mortar with a pestle at room temperature for 1 min. Completion of the reaction was tested by Thin Layer Chromatography (TLC).

After completion of the reaction, the reaction mixture was extracted with ethanol. The solidified mixture and the insoluble catalyst were separated by filtration. The filtrate was dried over anhydrous Na_2SO_4.

Evaporation of solvent afforded the product. Then it was subjected to GC and GC–MS analysis for the determination of the product yield. The purities of the synthesized hydrazines were examined with their physical constants and spectral data. The general structure of the synthesized hydrazines are shown in Figure 1.



No.	1	2	3	4	5	6	7	8	9	10
X	H	4-Br	4-Cl	4-N(CH_3)$_2$	4-F	4-OH	4-OCH_3	4-CH_3	3-NO_2	4-NO_2

Fig. 1. General structure of (*Z*)-1-(1-(substituted phenyl)ethylidene)-2(diphenylmethylene)hydrazines.

3. RESULTS AND DISCUSSION

3. 1. Infrared spectral correlations

In the present study, the author studied the effect of substituents on the spectral data of azine derivatives. In infrared spectral correlations the Hammett equation was employed (1) as

$$\nu = \nu_o + \rho\sigma \qquad \ldots(1)$$

where ν is the absorption group frequencies of substituent system and ν_o absorption group frequency of unsubstituted system, ρ is the reaction constant and σ is the substituent constant.

Table 1. The infrared and MNR spectral data of (*Z*)-1-(1-(substituted phenyl)ethylidene)-2-(diphenylmethylene)hydrazines

Sl. No.	**X**	**IR (ν, cm^{-1})**		**NMR (δ, ppm)**			
1	H	CN	N-N	^{1}H CH_3	^{13}C CH_3	^{13}C C-CH_3	^{13}C C-Ph_2
2	4-Br	1585.62	1116.27	2.33	16.01	127.84	159.61

3	4-Cl	1596.81	1113.96	2.34	15.94	127.85	159.62
4	4-$N(CH_3)_2$	1597.72	1115.76	2.32	15.91	127.86	159.66
5	4-F	1555.36	1112.68	2.16	15.78	127.32	159.41
6	4-OH	1574.26	1116.32	2.24	15.86	127.44	159.48
7	4-CH_3	1573.11	1116.48	2.26	15.83	127.41	159.43
8	4-OCH_3	1571.21	1112.12	2.17	15.72	127.22	159.21
9	3-NO_2	1572.36	1112.84	2.22	15.74	127.26	159.26
10	4-NO_2	1605.72	1118.26	2.48	16.24	127.96	159.72

The assigned νCN and –N-N- stretches (cm^{-1}) of prepared hydrazines are tabulated in Table 1. These stretches were correlated [5,6,20,22,24] with Hammett substituent constants, F and R parameters using single and multi-regression analysis. The results of statistical analysis the single parameter correlations are presented in the form of regression equations (2-13). The νCN (cm^{-1}) stretches of hydrazines gave good and satisfactory correlations with Hammett σ, σ^+, σ_I, σ_R and R parameters (eq. 2-7). The filed components of the substituents were failing in correlations. This is due to the inability of the effect of substituents on the stretches and along with resonance-conjugative structure as shown in Figure 2. All correlations gave positive ρ values. This may mean that the normal substituent effects operated in all system.

$$\nu CN(cm^{-1}) = 47.581(\pm 3.813)\sigma + 1605.73(\pm 16.942) \quad \ldots(2)$$
$$(r = 0.994, s = 5.31, n = 10)$$

$$\nu CN(cm^{-1}) = 20.460(\pm 2.825)\sigma^+ + 1587.33(\pm 2.148) \quad \ldots(3)$$
$$(r = 0.993, s = 6.61, n = 10)$$

$$\nu CN(cm^{-1}) = 47.119(\pm 16.208)\sigma_I + 1568.12(\pm 6.722) \quad \ldots(4)$$
$$(r = 0.971, s = 12.67, n = 10)$$

$$\nu CN(cm^{-1}) = 55.234(\pm 12.898)\sigma_R + 1595.02(\pm 4.109) \quad \ldots(5)$$
$$(r = 0.983, s = 10.01, n = 10)$$

$$\nu CN(cm^{-1}) = 35.185(\pm 12.193)F + 1570.83(\pm 8.610) \quad \ldots(6)$$
$$(r = 0.854, s = 15.27, n = 10)$$

$$\nu CN(cm^{-1}) = 39.388(\pm 8.129)R + 1596.29(\pm 3.877) \quad \ldots(7)$$
$$(r = 0.986, s = 9.16, n = 10)$$

The correlation of νN-N(cm^{-1}) stretches of hydrazine derivatives gave good and satisfactory correlations with Hammett σ, F and R parameters (eq. 8-13). The Hammett σ^+, σ_I and σ_R constants were failing in correlations. This is due to the reasons stated earlier and with

resonance-conjugative structure as shown in Figure 2. All correlations gave positive ρ values. This may mean that the normal substituent effects operated in all system.

$$\nu N\text{-}N(cm^{-1}) = 3.609(\pm 1.162)\sigma + 1115.11(\pm 0.516) \quad \ldots(8)$$
$$(r = 0.973, s = 1.66, n = 10)$$

$$\nu N\text{-}N(cm^{-1}) = 1.877(\pm 0.372)\sigma^{+} + 1115.63(\pm 0.598) \quad \ldots(9)$$
$$(r = 0.864, s = 2.84, n = 10)$$

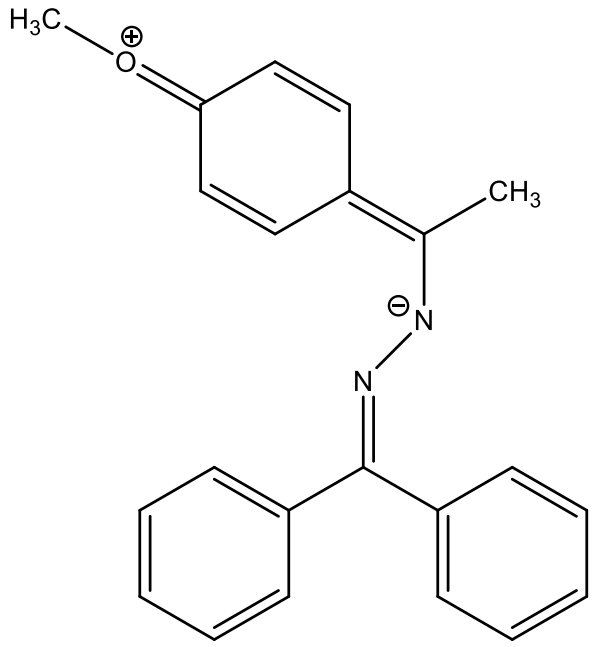


Fig. 2. The resonance-conjugative structure in (*Z*)-1-(1-(4-methoxyphenyl)ethylidene)-2-(diphenyl methylene) hydrazine.

$$\nu N\text{-}N(cm^{-1}) = 5.966(\pm 2.245)\sigma_I + 1113.31(\pm 0.936) \quad \ldots(10)$$
$$(r = 0.868, s = 2.75, n = 10)$$

$$\nu N\text{-}N(cm^{-1}) = 4.710(\pm 2.617)\sigma_R + 1116.25(\pm 0.881) \quad \ldots(11)$$
$$(r = 0.853, s = 2.83, n = 10)$$

$$\nu N\text{-}N(cm^{-1}) = 5.571(\pm 3.322)F + 1113.24(\pm 1.035) \quad \ldots(12)$$
$$(r = 0.964, s = 1.83, n = 10)$$

$$\nu N\text{-}N(cm^{-1}) = 3.210(\pm 1.811)R + 1116.32(\pm 0.863) \quad \ldots(13)$$
$$(r = 0.953, s = 2.04, n = 10)$$

Some of the single parameter correlation plots are shown in Figures 3-6.

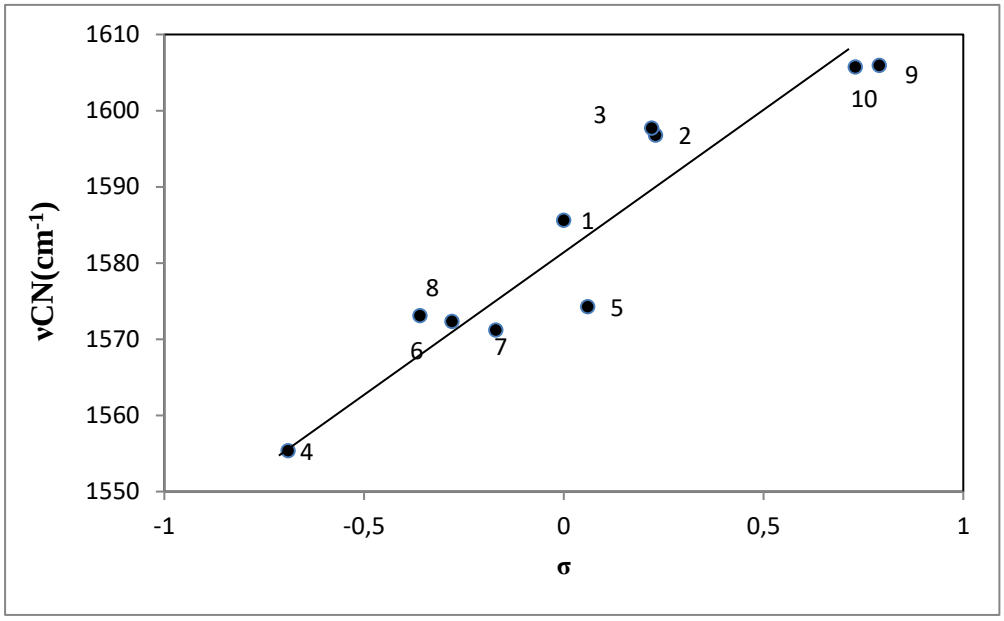


Fig. 3. Plot of νCN(cm^{-1}) of (*Z*)-1-(1-substitutedphenylethylidene)-2-(diphenyl methylene) hydrazines versus Hammett σ constant.

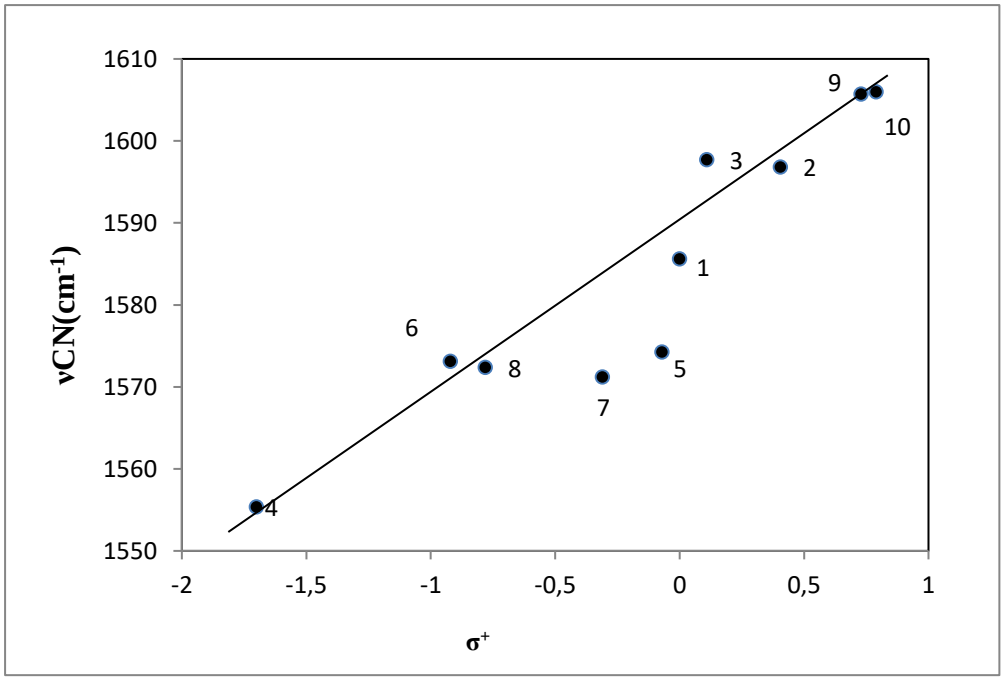


Fig. 4. Plot of νCN(cm^{-1}) of (*Z*)-1-(1-substitutedphenylethylidene)-2-(diphenyl methylene) hydrazines versus Hammett σ^{+} constant.

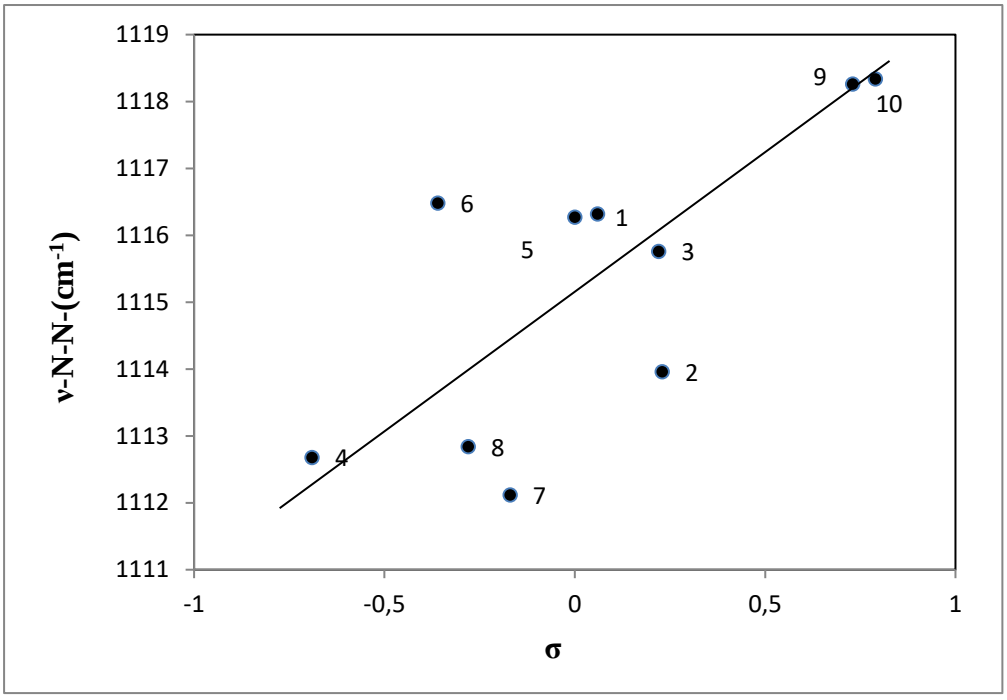


Fig. 5. Plot of νN-N(cm^{-1}) of (*Z*)-1-(1-substitutedphenylethylidene)-2-(diphenyl methylene) hydrazines versus Hammett σ constant.

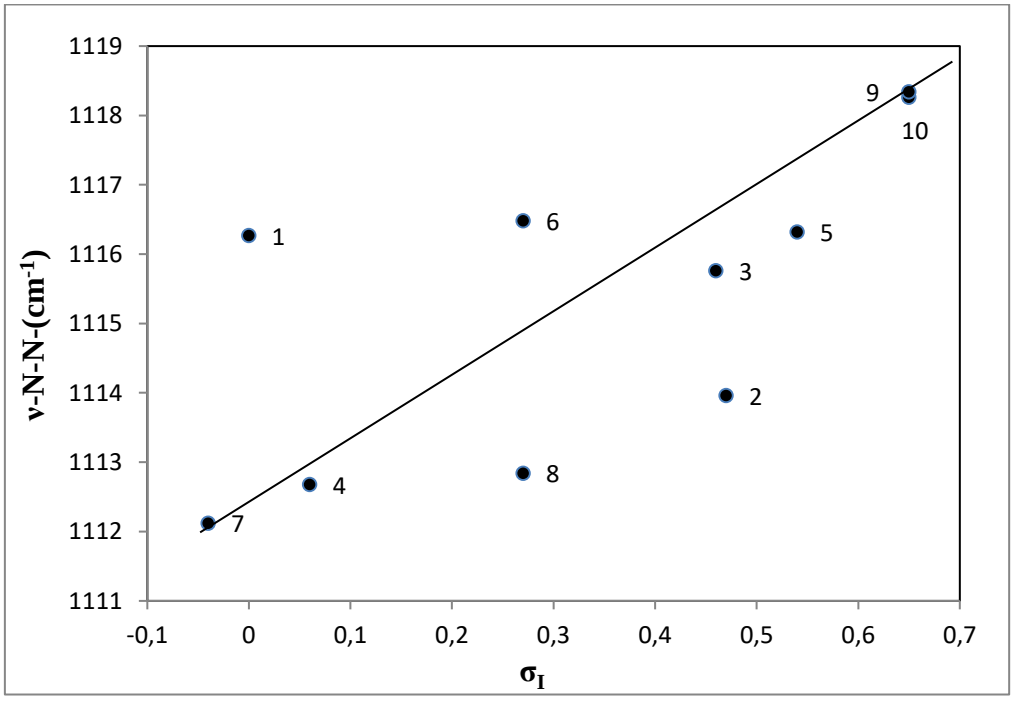


Fig. 6. Plot of νN-N(cm^{-1}) of (*Z*)-1-(1-substitutedphenylethylidene)-2-(diphenyl methylene) hydrazines versus Hammett σ_I constant.

In the infrared spectral single parameter correlations, some of the correlations were failed with Hammett substituent constants and F parameters. This is worthwhile when these are seeking in multi-parameter correlation involving σ_I, σ_R constants and Swain-Lupton's F and R parameters [25]. They produced good and satisfactory correlations. The multi-regression equations are shown in (14-17).

$$\nu CN(cm^{-1}) = 1581.95(\pm3.781) + 32.720(\pm7.538)\sigma_I + 44.503(\pm7.590)\sigma_R \quad \ldots(14)$$
$$(R = 0.995, P > 95\%, n = 10)$$

$$\nu CN(cm^{-1}) = 1581.90(\pm4.367) + 25.203(\pm8.285)F + 35.939(\pm5.815)R \quad \ldots(15)$$
$$(R = 0.994, P > 95\%, n = 10)$$

$$\nu N\text{-}N(cm^{-1}) = 1114.14(\pm1.264) + 0.983(\pm0.217)\sigma_I + 3.997(\pm1.313)\sigma_R \quad \ldots(16)$$
$$(R = 0.972, P > 95\%, n = 10)$$

$$\nu N\text{-}N(cm^{-1}) = 1114.27(\pm1.133) + 4.970(\pm2.260)F + 3.080(\pm1.213)R \quad \ldots(17)$$
$$(R = 0.976, P > 95\%, n = 10)$$

3. 2. ^{1}H NMR spectral correlations

The assigned CH_3 proton chemical shifts (δ, ppm) of hydrazine derivatives are presented in Table 1. These chemical shifts (δ, ppm) were correlated with Hammett substituent constants, F and R parameters using single and multi-regression analysis. In NMR chemical shift correlations, the Hammett equation was taken in the form of (18).

$$\delta = \delta_o + \rho\sigma \quad \ldots(18)$$

where δ is the absorption group frequencies of substituent system and δ_o absorption group frequency of unsubstituted system, ρ is the reaction constant and σ is the substituent constant.

The results of statistical analysis [5,6,20,22,24] the single parameter correlations are presented in the form of regression equations (19-24). The CH_3 proton chemical shifts (δ, ppm) of hydrazines gave good and satisfactory correlations with Hammett σ, σ^+, σ_I, σ_R and R parameters (eq. 19-24).

The filed components of the substituents were failing in correlations. This is due to the inability of the effect of substituents on the stretches and along with resonance-conjugative structure as shown in Figure 2. All correlations gave positive ρ values. This may mean that the normal substituent effects operated in all system.

$$\delta CH_3(ppm) = 0.221(\pm0.032)\sigma + 2.287(\pm0.014) \quad \ldots(19)$$
$$(r = 0.992, s = 0.04, n = 10)$$

$$\delta CH_3(ppm) = 0.121(\pm0.026)\sigma^+ + 2.320(\pm0.020) \quad \ldots(20)$$
$$(r = 0.984, s = 0.06, n = 10)$$

$$\delta CH_3(ppm) = 0.321(\pm0.103)\sigma_I + 2.195(\pm0.043) \quad \ldots(21)$$
$$(r = 0.972, s = 0.08, n = 10)$$

$$\delta CH_3(ppm) = 0.351(\pm0.088)\sigma_R + 2.370(\pm0.028) \quad \ldots(22)$$
$$(r = 0.981, s = 0.06, n = 10)$$

$$\delta CH_3(ppm) = 0.246(\pm0.212)F + 2.207(\pm0.057) \quad \ldots(23)$$
$$(r = 0.958, s = 0.09, n = 10)$$

$$\delta CH_3(ppm) = 0.235(\pm0.06)R + 2.373(\pm0.030) \quad (24)$$
$$(r = 0.979, s = 0.07, n = 10)$$

3. 3. ^{1}H NMR spectral correlations

The assigned ^{13}C chemical shifts (δ, ppm) of CH_3, δC-CH_3 and C-Ph_2 carbons of hydrazine derivatives are presented in Table 1. These chemical shifts (δ, ppm) were correlated with Hammett substituent constants, F and R parameters using single and multi-regression analysis. From the statistical analyses the single parameter correlation equations are given in (25-42).

The correlation of ^{13}C chemical shifts (δ, ppm) of CH_3 and C-CH_3 carbons of hydrazine derivatives gave good and satisfactory correlations with Hammett σ, σ^+, σ_I, σ_R, F and R parameters (eq.25-36).

$$\delta CH_3(ppm) = 0.360(\pm0.066)\sigma + 15.908(\pm0.029) \quad \ldots(25)$$
$$(r = 0.988, s = 0.09, n = 10)$$

$$\delta CH_3(ppm) = 0.190(\pm0.053)\sigma^+ + 15.961(\pm0.040) \quad \ldots(26)$$
$$(r = 0.978, s = 0.12, n = 10)$$

$$\delta CH_3(ppm) = 0.482(\pm0.192)\sigma_I + 15.763(\pm0.079) \quad \ldots(27)$$
$$(r = 0.966, s = 0.15, n = 10)$$

$$\delta CH_3(ppm) = 0.397(\pm0.211)\sigma_R + 15.784(\pm0.094) \quad \ldots(28)$$
$$(r = 0.955, s = 0.16, n = 10)$$

$$\delta CH_3(ppm) = 0.378(\pm0.117)F + 16.047(\pm0.056) \quad \ldots(29)$$
$$(r = 0.975, s = 0.13, n = 10)$$

$$\delta CH_3(ppm) = 0.362(\pm0.021)R + 16.324(\pm0.065) \quad \ldots(30)$$
$$(r = 0.965, s = 0.15, n = 10)$$

$$\delta C\text{-}CH_3(ppm) = 0.555(\pm0.128)\sigma + 127.584(\pm0.057) \quad \ldots(31)$$
$$(r = 0.983, s = 0.17, n = 10)$$

$$\delta C\text{-}CH_3(ppm) = 0.321(\pm0.081)\sigma^+ + 127.670(\pm0.062) \quad \ldots(32)$$
$$(r = 0.981, s = 0.19, n = 10)$$

$$\delta C\text{-}CH_3(ppm) = 0.720(\pm0.333)\sigma_I + 127.37(\pm0.013) \quad \ldots(33)$$
$$(r = 0.960, s = 0.26, n = 10)$$

$$\delta C\text{-}CH_3(ppm) = 0.919(\pm 0.266)\sigma_R + 127.800(\pm 0.094) \quad \ldots(34)$$
$$(r = 0.976, s = 0.20, n = 10)$$

$$\delta C\text{-}CH_3(ppm) = 0.520(\pm 0.311)F + 127.42(\pm 0.165) \quad \ldots(35)$$
$$(r = 0.944, s = 0.29, n = 10)$$

$$\delta C\text{-}CH_3(ppm) = 0.628(\pm 0.187)R + 127.813(\pm 0.089) \quad \ldots(36)$$
$$(r = 0.976, s = 0.21, n = 10)$$

The correlation of ^{13}C chemical shifts (δ, ppm) of C-Ph_2 carbons of hydrazine derivatives gave good and satisfactory correlations with Hammett σ, σ^+, σ_I, σ_R and R parameters (eq. 37-42). The filed components of the substituents were failing in correlations. This is due to the inability of the effect of substituents on the stretches and along with resonance-conjugative structure as shown in Figure 2. All correlations gave positive ρ values.

This may mean that the normal substituent effects operated in all system.

$$\delta C\text{-}Ph_2(ppm) = 0.323(\pm 0.088)\sigma + 159.49(\pm 0.039) \quad \ldots(37)$$
$$(r = 0.979, s = 0.12, n = 10)$$

$$\delta C\text{-}Ph_2(ppm) = 0.176(\pm 0.059)\sigma^+ + 159.54(\pm 0.045) \quad \ldots(38)$$
$$(r = 0.972, s = 0.13, n = 10)$$

$$\delta C\text{-}Ph_2(ppm) = 0.496(\pm 0.189)\sigma_I + 159.35(\pm 0.078) \quad \ldots(39)$$
$$(r = 0.968, s = 0.14, n = 10)$$

$$\delta C\text{-}Ph_2(ppm) = 0.470(\pm 0.194)\sigma_R + 159.61(\pm 0.063) \quad \ldots(40)$$
$$(r = 0.964, s = 0.15, n = 10)$$

$$\delta C\text{-}Ph_2(ppm) = 0.404(\pm 0.210)F + 159.36(\pm 0.094) \quad \ldots(41)$$
$$(r = 0.852, s = 1.16, n = 10)$$

$$\delta C\text{-}Ph_2(ppm) = 0.312(\pm 0.040)R + 159.61(\pm 0.067) \quad \ldots(42)$$
$$(r = 0.961, s = 0.15, n = 10)$$

Some of the single parameter correlation plots are shown in Figures 7-

In the study of multi-correlation analysis [25] with these NMR data of hydrazines gave good and satisfactory correlations with σ_I and σ_R or F and R parameters. The generated regression equations are presented in (43-50).

$$\delta CH_3(ppm) = 2.281(\pm 0.026) + 0.221(\pm 0.052)\sigma_I + 0.279(\pm 0.053)\sigma_R \quad \ldots(43)$$
$$(R = 0.995, P > 95\%, n = 10)$$

$$\delta CH_3(ppm) = 2.295(\pm 0.036) + 0.188(\pm 0.069)F + 0.209(\pm 0.048)R \quad \ldots(44)$$
$$(R = 0.994, P > 95\%, n = 10)$$

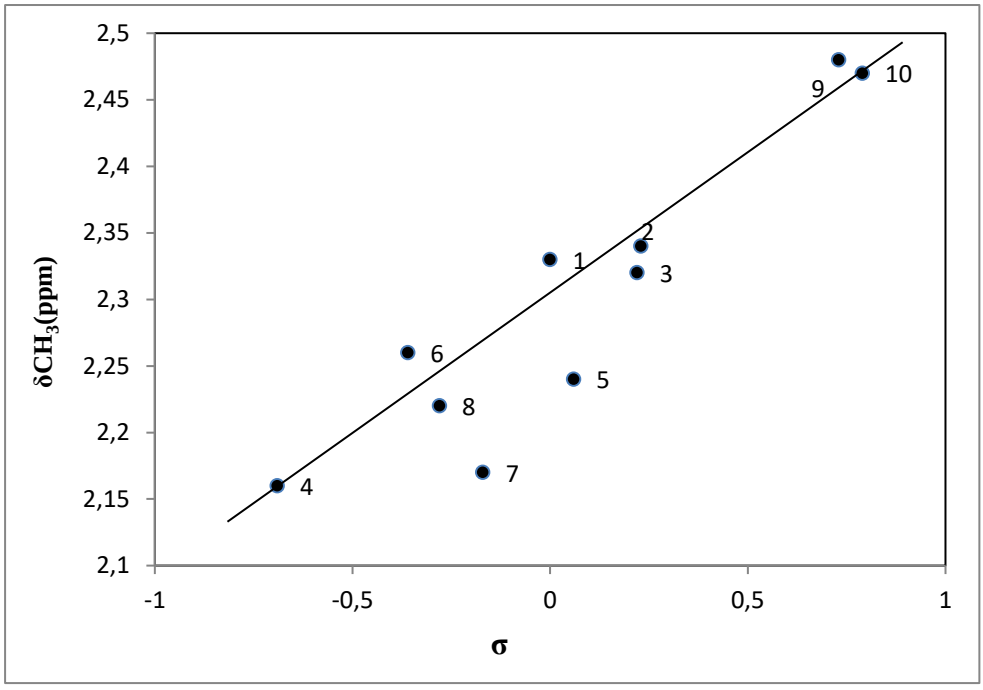


Fig. 7. Plot of δCH_3(ppm) of (*Z*)-1-(1-substitutedphenylethylidene)-2-(diphenyl methylene) hydrazines versus Hammett σ constant.

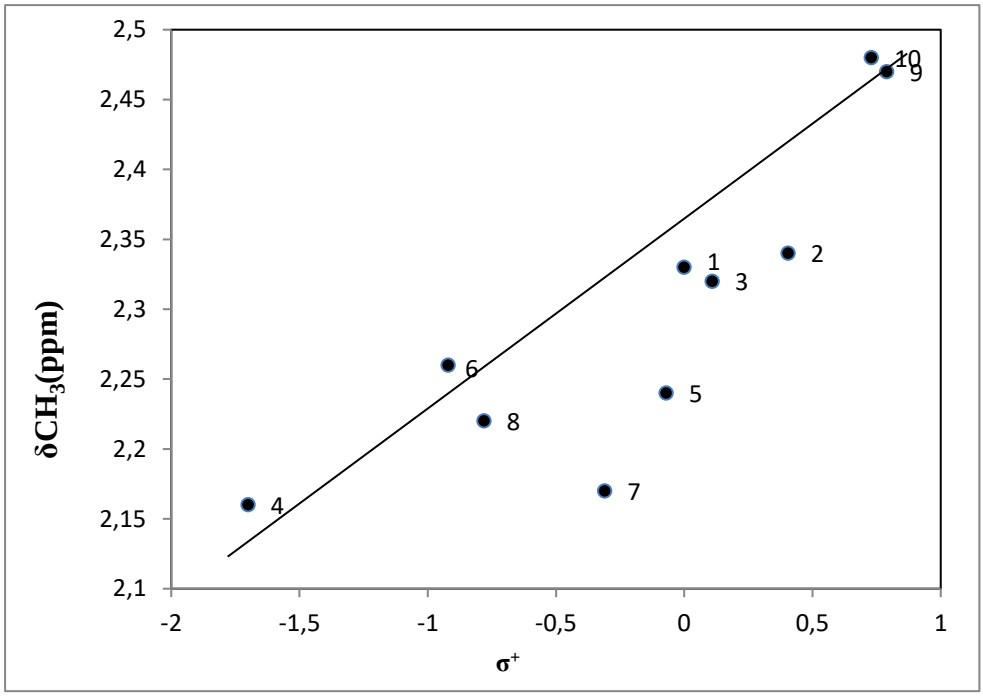


Fig. 8. Plot of δCH_3(ppm) of (*Z*)-1-(1-substitutedphenylethylidene)-2-(diphenyl methylene) hydrazines versus Hammett σ^+ constant.

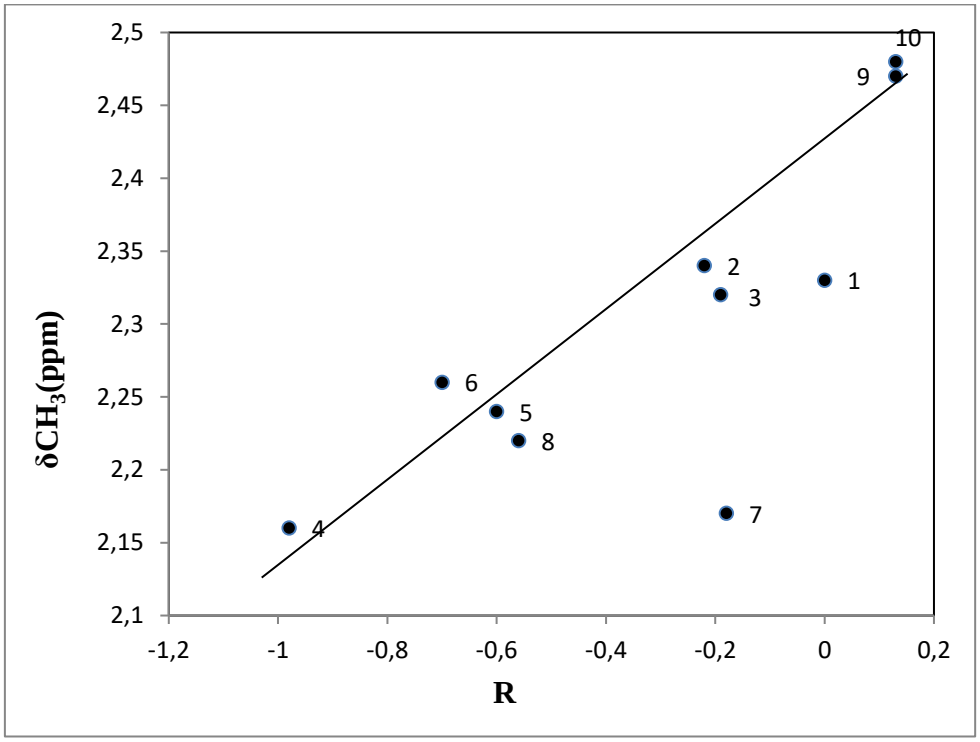


Fig. 9. Plot of δCH_3(ppm) of (*Z*)-1-(1-substitutedphenylethylidene)-2-(diphenyl methylene) hydrazines versus R parameter.

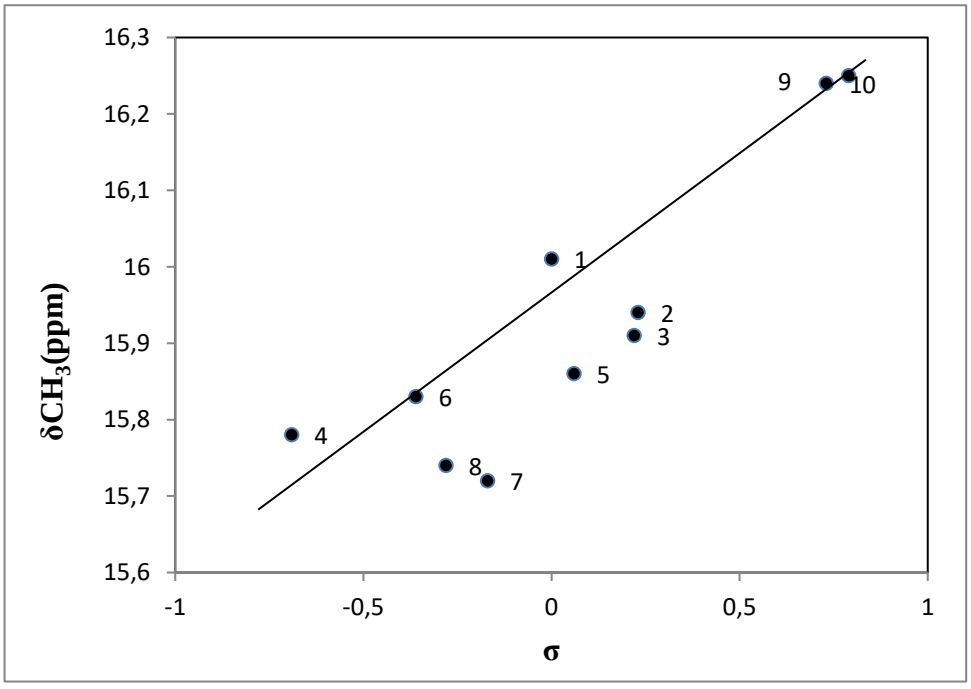


Fig. 10. Plot of δC-CH_3(ppm) of (*Z*)-1-(1-substitutedphenylethylidene)-2-(diphenyl methylene) hydrazines versus Hammett σ constant

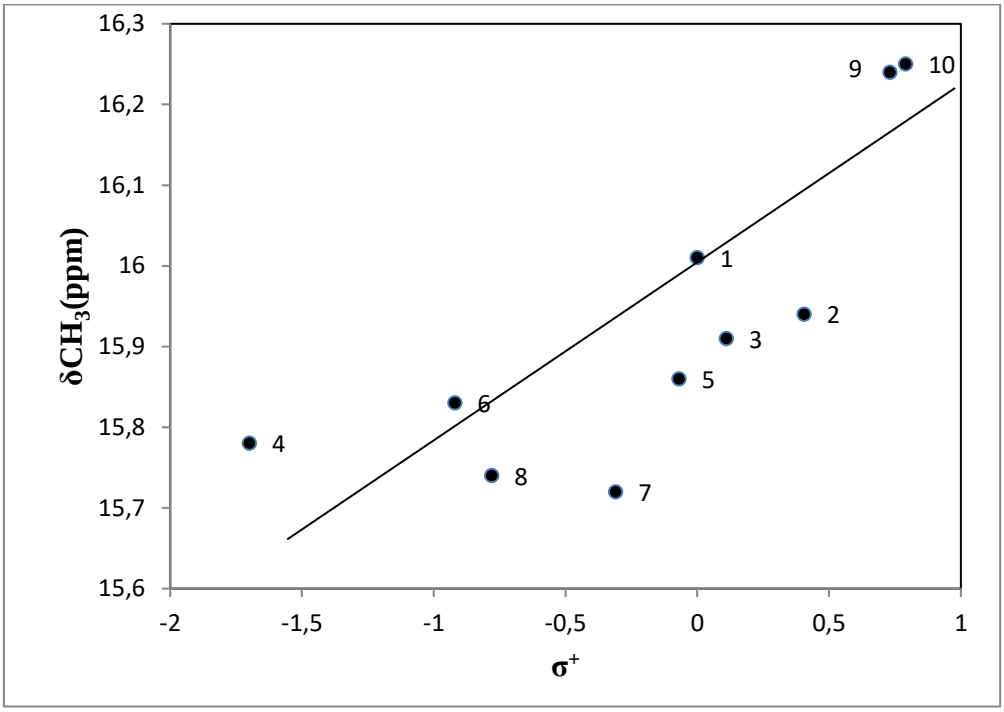


Fig. 11. Plot of δC-CH_3(ppm) of (*Z*)-1-(1-substitutedphenylethylidene)-2-(diphenyl methylene) hydrazines versus Hammett σ^+ constant

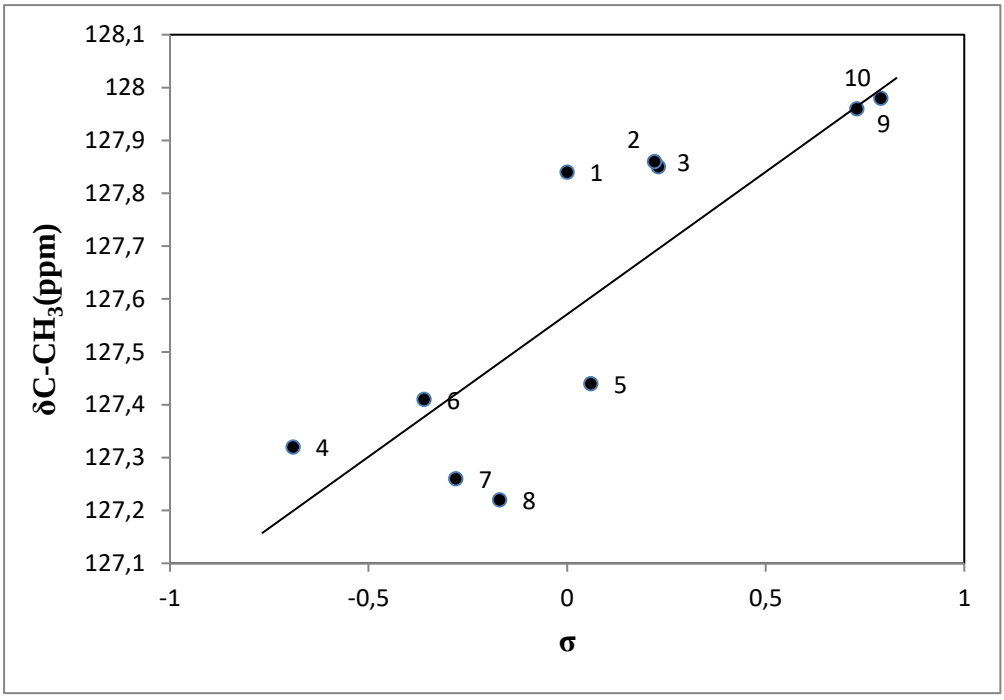


Fig. 12. Plot of δC-CH_3(ppm) of (*Z*)-1-(1-substitutedphenylethylidene)-2-(diphenyl methylene) hydrazines versus Hammett σ constant

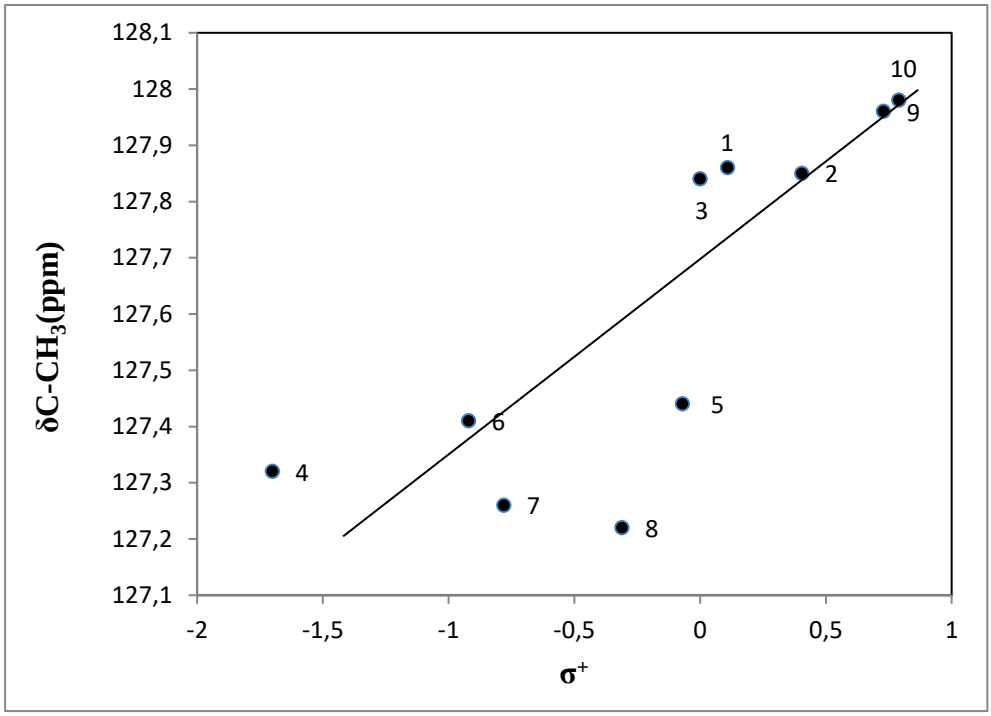


Fig. 13. Plot of δC-CH_3(ppm) of (*Z*)-1-(1-substitutedphenylethylidene)-2-(diphenyl methylene) hydrazines versus Hammett σ^+ constant

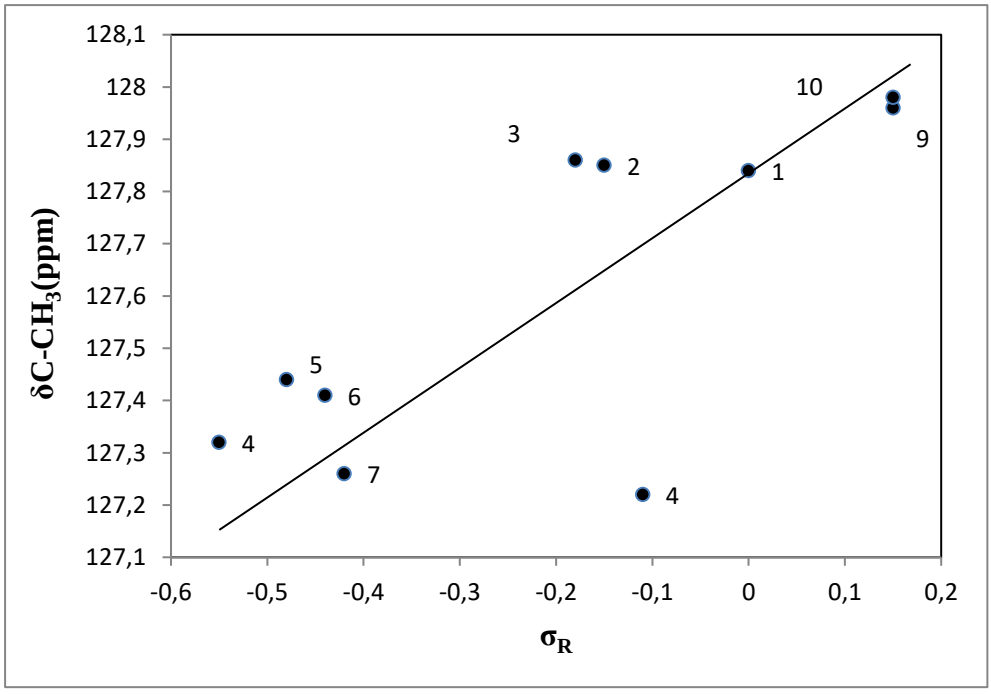


Fig. 14. Plot of δC-CH_3(ppm) of (*Z*)-1-(1-substitutedphenylethylidene)-2-(diphenyl methylene) hydrazines versus Hammett σ_R constant

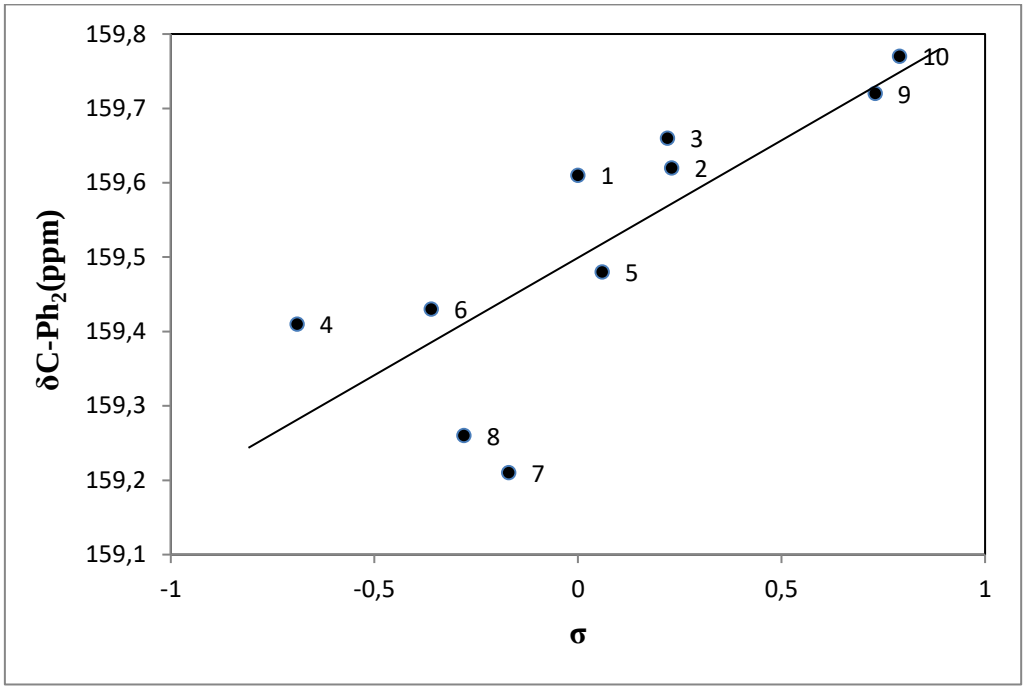


Fig. 15. Plot of δC-Ph_2(ppm) of (*Z*)-1-(1-substitutedphenylethylidene)-2-(diphenyl methylene) hydrazines versus Hammett σ constant

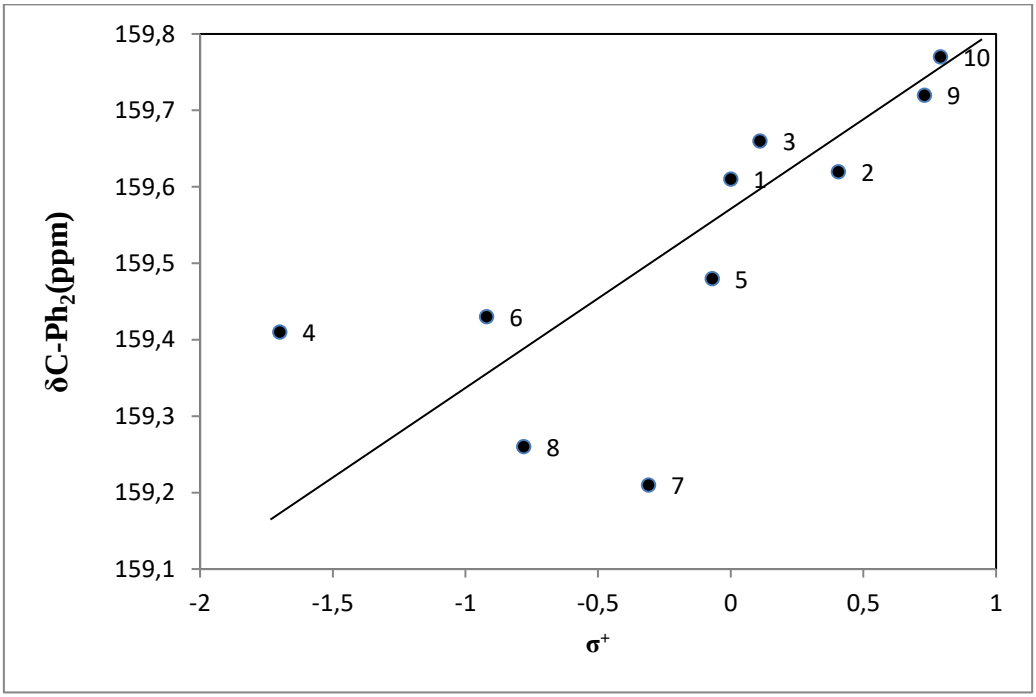


Fig. 16. Plot of δC-Ph_2(ppm) of (*Z*)-1-(1-substitutedphenylethylidene)-2-(diphenyl methylene) hydrazines versus Hammett σ^+ constant

$$\delta CH_3(ppm) = 15.917(\pm 0.060) + 0.326(\pm 0.119)\sigma_I + 0.482(\pm 0.120)\sigma_R \quad \ldots(45)$$
(R = 0.991, P > 95%, n = 10)

$$\delta CH_3(ppm) = 15.922(\pm 0.075) + 0.303(\pm 0.142)F + 0.336(\pm 0.100)R \quad \ldots(46)$$
(R = 0.985, P > 95%, n = 10)

$$\delta C\text{-}CH_3(ppm) = 127.611(\pm 0.122) + 0.473(\pm 0.244)\sigma_I + 0.763(\pm 0.245)\sigma_R \quad \ldots(47)$$
(R = 0.985, P > 95%, n = 10)

$$\delta C\text{-}CH_3(ppm) = 127.665(\pm 0.135) + 0.359(\pm 0.257)F + 0.579(\pm 0.018)R \quad \ldots(48)$$
(R = 0.994, P > 95%, n = 10)

$$\delta C\text{-}Ph_2(ppm) = 159.45(\pm 0.085) + 0.385(\pm 0.170)\sigma_I + 0.344(\pm 0.171)\sigma_R \quad \ldots(49)$$
(R = 0.981, P > 95%, n = 10)

$$\delta C\text{-}Ph_2(ppm) = 159.47(\pm 0.094) + 0.330(\pm 0.179)F + 0.267(\pm 0.126)R \quad \ldots(50)$$
(R = 0.976, P > 95%, n = 10)

4. CONCLUSIONS

A series containing ten (*Z*)-1-(1-(substituted phenyl)ethylidene)-2-(diphenylmethylene) hydrazines were prepared and their purities were examined by literature method. The infrared and NMR spectra of these hydrazines were recorded. The characteristics C=N and N-N stretches (ν, cm^{-1}) and the chemical shifts (δ, ppm) of methyl protons and carbons were assigned. These spectral have been correlated with Hammett substituent constants, F, R and Swain-Lupton's parameters using single and multi-regression analysis. From the results of statistical analyses, the single and multi-regression equations are drafted. From these equations, almost all regressions gave good and satisfactory correlation co-efficients except F parameter in single parameter analysis. Similarly all multi-regression analysis gave good and satisfactory correlation co-efficients.

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