Synthesis, Coating and Biological evaluation of Ecofriendly fatty acid Hydrazide Derivatives

Shaker N.O., N. A. Alian, M. M. El-Sawyand S.A. Bakr

Chemistry Department, Faculty of Science (Girls), Al –Azhar University, Nasr City, Cairo, Egypt.
maha_el_sawy@yahoo.com

Abstract: Fatty acid hydrazides are used as starting materials to synthesize some important fatty acid hydrazides derivatives by reacting fatty hydrazides (Olive and sunflower oils as well as oleic and linoleic acids) with carbon disulphide, monochloroacetic acid and maleic anhydride to obtain fatty oxadiazole, oxapyridazinone and pyridazine derivatives, respectively. The synthesized compounds were characterized with the help of IR and H^1- NMR spectra. Surface coating properties of the prepared compounds were studied in terms of drying time, chemical resistance & mechanical tests. All the prepared compounds showed good coating properties. The prepared compounds were tested for antibacterial activity against the representative group of Gram-positive and Gram-negative bacteria such as *p.flourescense, Bacillus subtilis and Escherichia coli*. and also tested for antifungal activity against *Aspergillusniger and Candida albicam*.
[Shaker N.O., N. A.Alian,M. M.El-Sawyand S.A.Bakr. **Synthesis, Coating and Biological evaluation of Ecofriendly fatty acid Hydrazide Derivatives.** *Life Sci J* 2015;12(11):73-79]. (ISSN:1097-8135). http://www.lifesciencesite.com. 8

Keywords: Fatty acid hydrazide, oxadiazole, oxapyridazinone, maleazine, antibacterial, antifungal coating properties.

1.Introduction

Hydrazides, the acylated derivatives of hydrazine are usually encountered as the simple or monosubsituted ($RCONHNH_2$), or as sym-disubsituted (RCONHNHOCR) compounds. The latter have been referred to as sec-hydrazides.[1]

Besides being useful themselves for a number of biological properties, hydrazides are important starting materials for a wide range of derivatives utilizable in Pharmaceutical products and as surfactants. Hydrazides have been known to be associated with antibacterial[2], antifungal[3], anthelmintic[4] and anticonvulsant[5] activities.

Various thiosemicarbazide derivatives are reported to possess useful pharmacological properties like antitubercular[6], antidepressant[7], anti-inflammatory[8] and analgesic[9] activities. In addition to the antibacterial[10] activities exhibited by several triazole derivatives, they are also known for their fungicidal, herbicidal[11], analgesic and anti-inflammatory[12] activities. Oxadiazoles and their derivatives are well known chemotherapeutic agents and their utility has muscle relaxant[13], hypoglycemic[14, 15] and fatty hydrazides and their derivatives are reported earlier to possess antibacterial and antifungal activities[16-20].

The reactions between carbonyl-functional resin and various hydrazide cross-linkers were examined under various conditions revealed that this reaction system had many other interesting characteristics. If this reaction system is used with comprehensive and appropriate understanding, it would be possible to develop various novel coatings in response to demand from the environment-conscious society. [21]

A series of novel N,N'-bis(1-carboxy-15-hydroxy-*n*-pentadec-8-yl)alkyl or -aryl amides derivatives such as hydrazides, thiosemicarbazides, oxadiazoles, and triazoles have been synthesized. These newly synthesized oleochemicals have been studied and characterized by FTIR, ^{1}H NMR, and ^{13}C NMR spectroscopies and elemental analyses. They are used as antislip and antiblock additives for polyethene films in which they are incorporated. They are also used as wall repellents for textiles and as mold release agents, and they are employed in rubber goods and printing inks. Because of this interest in the biological and industrial potential of oleochemicals,[22]a resinous composition for water paint crosslinkable at the normal temperature is provided, which is suitable for painting interiors and exteriors of buildings, bridges, vehicles which is non-odored, as well as which has advantages in safe and hygienic aspects.[23]

Six azoles with n-pentyl side chain were synthesized from n-hexanoic acid. Three N-glycosides namely: 5-pentyl-2-(D-amino arabinoside)-1,3,4-oxadiazole, 5-pentyl-2-(D- aminoglycoside)-1,3,4-thiadiazole, and 3-pentyl-4-(D-amino xyloside)-4 H-1,2,4-triazole-5-thiol were prepared from already synthesized n-pentyl azoles, respectively. Surface active properties of water soluble synthesized compounds were studied in terms of surface tension, cloud point and critical micelle concentration. The antibacterial activities were assessed using the paper disk diffusion and broth dilution methods against gram-positive and gram-negative bacteria. Some of

the synthetic compounds showed promising activity against micro-organisms under test in comparison to commercially available antibiotics polymixine and oxytetra- cycline.[24]

The long-chain fatty acid hydrazide (1) was prepared from the corresponding long-chain fatty ester with hydrazine hydrate (2). Reacting 1 with phenyl isothiocyanate afforded the corresponding thiosemicarbazide (3). The later 3 underwent intramolecular cyclization in basic medium, and gave the s-triazole derivative, which was methylated and afforded 3-heptadecanyl-5-(methylthio)-4-phenyl-4H-1,3,4-triazole, which was then treated with hydrazine hydrate and afforded the corresponding 1-(5-heptadecanyl-4-phenyl-4H-1,2,4-triazol-3-yl) hydrazine. On the other hand, thiosemicarbazide 3 underwent intramolecular cyclization in acid medium and afforded the corresponding thiadiazole derivative. Treatment of thiosemicarbazide 3 with ethyl chloro(arylhydrazono) acetate derivatives. Similarly, when the thiosemicarbazide 3 was treated with the phenylcarbamoylarylhydrazonyl chloride, it afforded (3-Aryl-N-5-(phenylcarbamoyl)-1,3,4-thiadiazol-2(3H)-ylidene)octadecanehydrazide. Also the reaction of thiosemicarbazide3 with 2-oxo-N-arylpropanehydrazonoyl chlorides and N-phenylbenzohydrazonoyl chloride gave the corresponding thiadiazole derivatives. A solution of thiosemicarbazide 3 was treated with the haloketones, afforded the thiadiazine derivatives. Analogously, the thiosemicarbazide 3 was reacted with α-haloketones and afforded the corresponding products. The structure elucidation of all synthesized compounds is based on the elemental analysis and spectral data.[25]

HimamiVarshney et_al[26] reported the synthesis of many heterocyclic moiety substituted with fatty acid residue. 2,5-Dimethyl pyrrole & 1,3-benzoxazin-4-one derivatives were systhesized from cyclization of fatty acid hydrazide with acetonyl acetone and fatty esters with anthranilic acid in $POCl_3$.

On the other hand **Himani Varshaney et_al**[27] also prepared a series of novel unsaturated hydroxyl and non-hydroxy fatty acid residue substituted 1,3,4-selenadiazoles from the reaction of hydrazide with acetyl chloride in anhydrous sodium carbonate in THF and H_2O at o°c to form N\-acetyl undec-10-enoic hydrazide, N-acetyl-(9Z)-octadec-9-enoic hydrazide, N- acetyl -(9Z, 12R)-12-hydroxy-9-enoic hydrazide, and N-acetyl-(9R, 12Z)-9-hydroxy-12-enoic hydrazide.

An attempt therefore, has been made to prepare ecofriendly fatty hydrazide derivatives for their evaluation as coating, anti bacterial and antifungal products with a view to explore new value added areas of application for these compounds with enough avaibility.

2. Experimental

2.1. Materials;

Hydrazine hydrate, maleic anhydride, oleic acid, linoleic acid, olive oil, sunflower, Carbon disulphide and chloroacetic acid (Fine Chemicals, Mumbai, India). All other reagents and solvents used throughout this study were obtained from the chemicals pure grade.

FT-IR spectra were recorded in KBr on a Shimadzu FT-IR spectrometer.

^{1}HNMR (400 MHz) spectra were recorded on a Jeol EX400 NMR Spectrometer in DMSO-d_6 using the DMSO signal as a reference.

2.2. Synthesis of acid chlorides:

The preparation of fatty acid chlorides were carried out by treatment of fatty acids with thionyl chloride. According to the procedure described by **Weil et al.**[28]

The preparation of Oleyl chloride is desdribed as example:

Oleic acid (1mol, 282 gm) was placed in a 250 ml flask fitted with a reflux condenser and seperatory funnel containing redistilled thionyl chloride. The flask was heated gently in a water bath, thenthionyl chloride (2 moles, 238 gm) was added dropwise from the seperatory funnel during the course of 30-40 min. The hydrogen chloride evolved was received in a water trap attached to the top of the condenser. When the thionyl chloride has been added, the contents of the flask were heated for 30 min. The reaction flask was left 2 hours, after which the excess thionyl chloride was distilled off under reduced pressure and kept over night under vacuum. The products were used in the following steps of synthesis without further purification.

2.3. Synthesis of fatty hydrazides (Ia-Id)

To a solution of fatty acid chloride (0.01 mol) in dry acetone (30 ml), hydrazine hydrate (0.02 mol, 0.64g) was added and the reaction mixture was heated under reflux for 4hrs and then left to cool. The products were collected and recrystallized using acetone.

2.4. Synthesis of fatty oxadiazole derivatives (IIa-IId)

Fatty hydrazides (Ia-d) (0.01 mol), were reacted with carbon disulphide (0.01 mol, 0.76g) in presence of potassium hydroxide as catalyst. The mixture was refluxed for 6 hrs until the release of H_2S was ceased. The mixture was then cooled and acidified with dilute HCl. The crude compounds were recrystallized from chloroform to give oxadiazole derivatives.

2.5. Synthesis of fatty oxapyridazinone derivatives (III a – IIId)

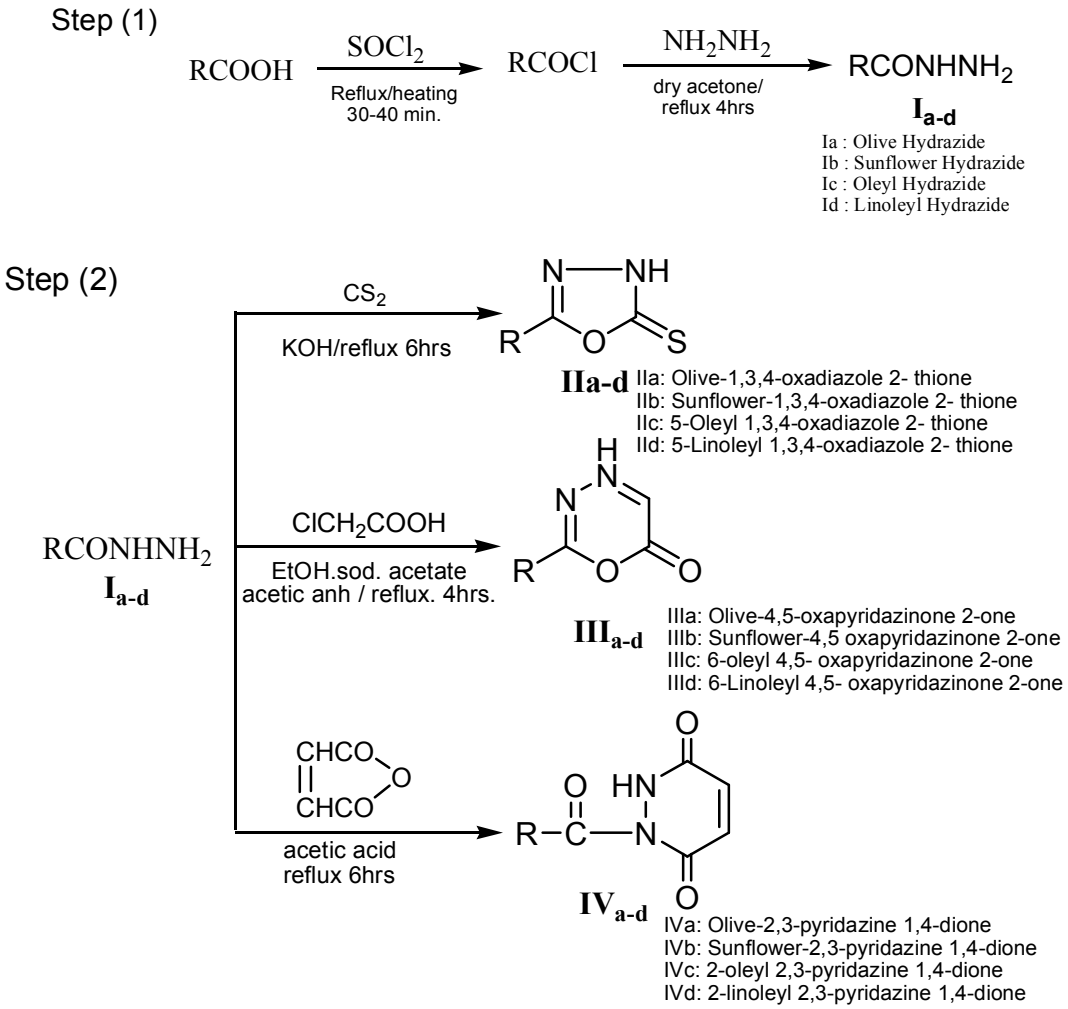
A mixture of (0.01 mol) fatty hydrazides (Ia–d) and (0.01mol, 9.4gm,) choloroacetic acid in ethyl

alcohol in presence of sodium acetate and acetic anhydride was heated under reflux for 4hrs then the reaction was cooled and poured on water, the products were obtained and crystallized using ethanol.

2.6. Synthesis of fatty Maleazine derivatives (IVa – IVd)

Maleic anhydride (0.01 mol, 0.98g) dissolved in acetic anhydride and fatty hydrazide (0.01 mole) in acetic acid were refluxed for about 6hrs, cooled then poured on water, the products were collected and crystallized using ethanol.

The synthetic route and abbreviations of the prepared coating compounds are shown in the following scheme:



3. Evaluation of the prepared compounds:

The drying performance tests and thickness for the products were carried out using standard methods[29]. Moreover, the prepared fatty hydrazide compounds were separately formulated and their characteristics were compared.

3.1. Preparation of test panels:

Glass and mild steel plates (50 x 150 mm) were degreased by dipping into petroleum ether, then the surfaces were cleaned by fine cloth, washed and wiped. The plates were washed with ethyl alcohol and allowed to dry in air. Films were applied onto clean plates and left for half an hour to remove slowly the greatest part of the solvent and then stove at the required temperature for the specified time in a thermostatically controlled well – ventilated oven.

The prepared compounds were thinned to brushable consistency and appropriate quantities of cobalt and lead octoate driers were added. Coatings were applied on previously prepared mild steel and glass panels with a brush to obtain a uniform coat. Drying on film formation of coatings were carried out at room temperature, 80˚C, 100˚C and 150˚C for one hour **(table 3).**

3.2. Evaluation of film characteristics:

3.2.1.Film resistance:

Water, alkali (10% NaOH) acid (10% HCl, 20% H_2SO_4) resistance and solvents (acetone, ethanol, methanol, ethyl methyl ketone, and toluene) resistance were carried out according to standard methods.[30,31] **(table 4).**

3.2.2. Mechanical tests:

After tack-free drying, the film properties such as: bending test, adhesion (tape test), impact, gloss and scratch hardness by pencil test, were determined by standard methods.[32-36] **(table 5).**

3.3.Biological activity:[37]

All the prepared compounds were tested for their bactericidal activity against (*P.flourescence, Bacillus sublitis*, *Escherichia coli*,) and their antifungal activity against (*Aspergillusniger* and *Candida albicam*). **(table 6)**

The antibacterial activities of the tested compounds were evaluated using the paper disk diffusion method. DMSO which is known as bacterial static in the above mentioned concentration was used as negative control and standard disks (Mast Diagnostics, UK), saturated with known antibiotic polymixine and oxytetracycline as positive control were applied. After incubation at 37˚C for 24 h, the zone of inhibition of growth around each disk was measured in millimeters and zone diameters were interpreted in accordance with CLSI and BCCLS.

4. Results and Discussion

In this study different fatty hydrazide derivatives were prepred (Ia-d). Firstly,oxadiazole derivatives were prepared by reaction of (Ia-d) with carbon disulphide to obtain (IIa-d). The prepared compounds were confirmed by spectroscopic methods. IR spectra shows nN-H at 3228, nC=N at 1620, nC=S at 1490, nC-H aliphatic at 2920–2850cm^{-1}. And H^1NMR exhibited signals at σ (6.5,S,NH), σ(0.84, t, 3H,CH_3), σ (2.4,m,$(CH_2)_{17}$,34H).

Secondly, oxapyridazinone derivatives were prepare by reaction of (Ia-d) with monochloroacetic acid in ethyl alcohol. IR spectra of (IIIa) shows nN-H at 3838, nC=O at 1703 and 1651, nC-H aliphatic at (2927– 2858.3) cm^{-1}. And H^1NMR exhibited signals at σ (0.9, t,3H,CH_3), σ (1.3, m, $(CH_2)_{17}$, 34H), σ (6,S, H, CH) and σ (5, S,H,NH proton).

Finally maliezine derivatives (IVa.d) were produced by the reaction of compound (Ia-d) with maleic anhydride in acetic anhydride. IR spectra of IVa shows nN-H at 3394.5, nC=O at 1643.2, nC-H aliphatic at (2927.7–2858.3) cm^{-1}. And H^1NMR exhibited signals at σ (0.9, t,3H,CH_3), σ (1.4, m, $(CH_2)_{16}$, 32H),
σ (6,S, 2H,CH of pyridazine ring) and σ (4.1, S,H,NH) **(table 2).**

Table 1 illustrates the physicochemical properties of the prepared compounds: color, molecular formula, molecular weight and yield %.

In order to show the performance of prepared hydrazide derivatives, the following tests were performed:

Drying time:

The drying time test was conducted by applying the coating on a glass and mild steel plates. They were air dried, then stoved at 80˚C for 1 hour, at 100˚C for 1 hour and at 150˚C for 1 hour. **Table 3** illustrates the drying time of air dried and stovedhydrazide derivatives films. The data indicate that the drying time varying between (4,5, 6 & 7) hours in open air while increasing temperature to 80˚C for 1 hour, at 100˚C for 1 hour and at 150˚C for 1 hour. It can be concluded that the optimum stoving schedule was found to be 1hr at 150˚C as all the films were hard dry except IIIb&IIIc films were hard dry at 100˚C.

Chemical resistance:

Water, alkali (10% NaOH), acid (10% HCl & 20% H_2SO_4) and solvents (acetone, methanol, ethanol, ethyl methyl ketone & toluene) resistances were evaluated for the prepared hydrazide derivatives films. **Table 4** illustrates the films resistance of hydrazide derivatives, it is clear that the films resist water, acetone, methanol, ethanol, toluene and ethyl methyl ketone, 10% HCl & 20% H_2SO_4 for more than 60 days except resistance of NaOH.

Mechanical test:

Table 5 illustrate that hydrazide derivatives films exhibited good overall mechanical properties, e.g. gloss, adhesion, impact, scratch hardness, bending and flexibility.

Biological activities:

All the prepared compounds were tested for their antibactericidal activity against (*P. flourescence, Bacillus sublitis*, *Escherichia coli*) and antifungal against (*Aspergillusniger* and *Candida albicam*). The results obtained indicated that compounds II a-d are more active than IIIa-d & compounds IIIa-d are more active than IVa-d as bactericide or fungicide agents. **Table 6**

The present results highlight the importance of the prepared ecofriendly fatty hydrazide derivatives as coatings and therefore can be recommended for use in finishes where good mechanical properties with mild chemical resistance are required.

It can be concluded also that, some of the synthetic compounds showed promising activity against microorganisms under tests *(p.flourescense, Bacillus subtilis and Escherichia coli*, *Aspergillusniger and Candida albicam*) in comparison to commercially available antibiotics polymixine and oxytetracycline.

Table 1: Physicochemical properties of the prepared compounds:

Comp.	Colour	Molecular formula	Molecular weight	Yield %
IIa	light brown	$C_{20}H_{35}N_2O_2S$	367.57	67
IIb	light brown	$C_{20}H_{31}N_2O_2S$	363.54	82
IIc	yellow	$C_{20}H_{35}N_2O_2S$	367.57	66
IId	yellow	$C_{20}H_{31}N_2O_2S$	363.54	76
IIIa	yellow	$C_{21}H_{35}N_2O_3$	363.52	75
IIIb	brown	$C_{21}H_{31}N_2O_3$	359.49	65
IIIc	brown	$C_{21}H_{35}N_2O_3$	363.52	76
IIId	light brown	$C_{21}H_{31}N_2O_3$	359.49	69
IVa	light brown	$C_{23}H_{37}N_2O_4$	405.56	84
IVb	yellow	$C_{23}H_{33}N_2O_4$	401.53	82
IVc	yellow	$C_{23}H_{37}N_2O_4$	405.56	79
IVd	light brown	$C_{23}H_{33}N_2O_4$	401.53	77

Table 2: Spectra Analysis of the Prepared Compounds

Compounds	IR Spectra	H^1NMR Spectra
IIa	nNH at 3228, nC=N at 1620, nC=S at 1490, nC-H aliphatic at 2920–2850 cm^{-1}.	exhibited signals at σ (6.5, S,NH), σ (0.84, t, 3H,CH_3), σ (2.4,m,$(CH_2)_{17}$,34H)
IIb	nNH at 3437, nC=O at 1654, nC=N at 1618, nCH aliphatic at (2919– 2852) cm^{-1}.	exhibited signals at σ (0.9, t,3H,CH_3), σ (1.3, m, $(CH_2)_{16}$ 30H) and σ (3.6, S,H,NH proton)
IIc	nNH at 3413, nC=N at 1639.4 nC=S at 1431.1, nC-H aliphatic at 2923–2854 cm^{-1}.	exhibited signals at σ (6.6, S,NH), σ (0.85, t, 3H, CH_3,), σ (2.4, m, $(CH_2)_{17}$,34H) .
IId	nNH at 3224.8, nC=O at 1708, nC=N at 1608, n CH aliphatic at (2927.8–2854.5)cm^{-1}.	exhibited signals at σ (0.9, t,3H,CH_3), σ (1.3, m, $(CH_2)_{16}$, 32H) and σ (4.1, S,H,NH proton)
IIIa	nNH at 3838., nC=O at1703and1651, n CH aliphatic at (2927–2858.3)cm^{-1}.	exhibited signals at σ (0.9, t,3H,CH_3), σ (1.4, m, $(CH_2)_{17}$, 34H), σ (5,S, H,NH) and σ (6, S,H,CH)
IIIb	nNH at 3413, nC=O at 1706, n C-H aliphatic at 2923–2854cm^{-1}.	exhibited signals at σ (0.9, t,3H,CH_3), σ (1.3, m, $(CH_2)_{16}$ 32H), σ (6,S, 1H,CH of pyridazine ring) and σ (4.1, S,H,NH proton)
IIIc	nNH at 3745, nC=O at1712and 1658, n CH aliphatic at (2927–2858)cm^{-1}.	exhibited signals at σ (6.6, S,NH), σ (0.85, t, 3H, CH_3,), σ (2.4, m, $(CH_2)_{17}$,34H) .
IIId	nNH at 3413.8 nC=O at 1709, n C-H aliphatic at 2923–2854 cm^{-1}.	exhibited signals at σ (6.5, S,NH), σ (0.84, t, 3H,CH_3), σ (2.4,m,$(CH_2)_{16}$, 32H)
IVa	nNH at 3394.5,n C=O at 1643.2, n CH aliphatic at (2927.7– 2858.3) cm^{-1}.	exhibited signals at σ (0.9, t,3H,CH_3), σ (1.3, m, $(CH_2)_{17}$ 34H), σ (6,S, 2H,CH of pyridazine ring) and σ (4.1, S, H, NH proton)
IVb	nNH at 3741, nC=O at 1708 and 1647.1, nCH aliphatic at (2927–2858.3)cm^{-1}.	exhibited signals at σ (0.9, t,3H,CH_3), σ (1.4, m, $(CH_2)_{16}$, 32H), σ (5,S, H,NH) and σ (6, S,H,CH)
IVc	nNH at 3690, nC=O at 1738, n CH aliphatic at 2938 cm^{-1}.	exhibited signals at σ (0.9, t,3H, CH_3), σ (1.3, m, $(CH_2)_{17}$, 34H), σ (6, S, 2H, CHof pyrdazine ring) and σ (4.1, S, H, NHproton)
IVd	nNH at 3648, nC=O at 1703, 1656 and 1640, nCH aliphatic at (2940–2851)cm–1.	exhibited signals at σ (0.9, t,3H, CH_3), σ (1.4, m, $(CH_2)_{16}$, 32H), σ (5, S, H, NH) and σ (6, S, H, CH)

Table 3:Drying Characteristics of the prepared compounds

Compounds	Air drying (hrs)	Stoving at 80˚C 1 hr	Stoving at 100˚C 1 hr	Stoving at 150˚C 1 hr
IIa	7	T	D	HD
IIb	7	T	D	HD
IIc	6	T	D	HD
IId	6	T	D	HD
IIIa	5	T	D	HD
IIIb	5	D	HD	------
IIIc	4	D	HD	-------
IIId	7	T	D	HD
IVa	7	T	D	HD
IVb	6	T	D	HD
IVc	6	T	D	HD
IVd	5	T	D	HD

D Dry; **T** Tacky; **HD** Hard Dry

Table 4: Film Resistance of the prepared compounds:

Comp.	Water resistance (months)	Alkali resistance (10% NaOH) (hours)	Acid resistance (months)		Solvents resistance (months)				
			HCl 10%	H_2SO_4 20%	Acetone	Methanol	Ethanol	Ethyl methyl ketone	Toluene
IIa	> 2	3	> 2	> 2	> 2	> 2	> 2	> 2	> 2
IIb	> 2	3	> 2	> 2	> 2	> 2	> 2	> 2	> 2
IIc	> 2	4	> 2	> 2	> 2	> 2	> 2	> 2	> 2
IId	> 2	4	> 2	> 2	> 2	> 2	> 2	> 2	> 2
IIIa	> 2	5	> 2	> 2	> 2	> 2	> 2	> 2	> 2
IIIb	> 2	6	> 2	> 2	> 2	> 2	> 2	> 2	> 2
IIIc	> 2	7	> 2	> 2	> 2	> 2	> 2	> 2	> 2
IIId	> 2	2	> 2	> 2	> 2	> 2	> 2	> 2	> 2
IVa	> 2	2	> 2	> 2	> 2	> 2	> 2	> 2	> 2
IVb	> 2	3	> 2	> 2	> 2	> 2	> 2	> 2	> 2
IVc	> 2	3	> 2	> 2	> 2	> 2	> 2	> 2	> 2
IVd	> 2	4	> 2	> 2	> 2	> 2	> 2	> 2	> 2

Table 5: Mechanical properties of the prepared compounds:

Compounds	Thickness of film (mm)	Gloss % at 60˚c	Adhesion test	Impact	Scratch hardness	Bending test
IIa	0.019	75	good	pass	2H	pass
IIb	0.021	78	good	pass	2H	pass
IIc	0.015	80	good	pass	2H	pass
IId	0.032	85	good	pass	2H	pass
IIIa	0.034	88	good	pass	2H	pass
IIIb	0.025	90	good	pass	2H	pass
IIIc	0.014	99	good	pass	2H	pass
IIId	0.034	60	good	pass	2H	pass
IVa	0.033	64	good	pass	2H	pass
IVb	0.026	68	good	pass	2H	pass
IVc	0.017	76	good	pass	2H	pass
IVd	0.024	80	good	pass	2H	pass

Table 6: Biological activities of the prepared compounds:

Compounds	*P.flourescense*	*Bacillus subtilis*	*Escherichia coli*	*Aspergillus niger*	*Candida albicam*
IIa	++	++	+	+	+
IIb	++	++	+	+	+
IIc	++	++	+	+	+
IId	+++	+++	+	++	+
IIIa	+	+	+	-	+
IIIb	+	+	+	+	+
IIIc	++	+	+	+	+
IIId	++	++	-	++	+
IVa	++	++	+	-	+
IVb	+	+	+	-	+
IVc	+	+	-	-	+
IVd	++	++	-	-	+

References

1. Markley, K.S., Fatty Acids, Their Chemistry, Properties and Uses, Interscience Publishers, New York, 1964.
2. Mir, I.; Siddiqui, M.T. and Comrie, A., Tetrahedron, 26, 5235, 1970.
3. Degener, E.; Scheinplug, H. and Schemelzer, H.G., Brit. Pat. 1035,474, Oct. 15, 1967, ChemAbstr, 68, 95567, 1968.
4. Cavier, R. and Rips, R., J.Med. Chem., 11, 8706, 1965.
5. Renz.J.; Bourguin, J.P. Winkler, H. BruieshWeiler, C. Reush, L. and Schewarb, G., Swiss 419136(1670), ChemAbstr, 68, 29709, 1968.
6. Brown, R.K.; Snider, R.F. and Stevenson, M.D., J.Org. Chem., 21, 261, 1956.
7. Allias,A.; and Meier, J., Er. Pat, 1363,855, ChemAbstr, 61, 146141, 1967.
8. Alverex, E.F.; Pajares, M.B. and Lopez, O.N., ChemAbstr, 67, 32590, 1967.
9. Lieberman, D.; Rist, N. Grumbach, F. Moyewx, M. Gautheir, V. Rouqix, A. Millard, J. Himbert, J. G. and Cals, S., Bull Soc. Chim. France, 1440 (1954); ChemAbstr, 50, 14109, 1956.
10. Sengupta, A. K.; Garg, M. and Chandra, U., J. Ind.Chem. Soc., 56, 1230, 1974.
11. Greenfield, S. A.; Seidal, M. C. and Von Mayer, W. C.,Ger. Offen 1,966,806, Oct. 24, 1974, ChemAbstr, 82, 150485, 1975.
12. George, T.; Mehta, D.V. Tahiramani, R. David, J. and Talwalker, P.K., J.Med.Chem., 14, 335, 1971.
13. Piala, J.J.; and Yale, H.L., U.S.3, 166, 566(1965); Chem Abstr, 62, 10444, 1965.
14. Hanford, W. E.; (to M. W. Kellog Co.), U. S. Pat. 2,717,200, Sept. 6, 1955. J. Ind. Council Chem., Vol. 25, No. 2, 130, 2008.
15. O'Neal, J.B.; Rosen, H. Russel, P.B. Adams, A.C. and Blumenthal, A., Chem Abstr, 57, 9168, 1962.
16. Sherman, W.R.; J. Org. Chem., 26, 88, 1961.
17. Raval, D.A.; and Toliwal, S.D., J. Oil Tech. Assn. of India., 26(1), 27, 1994.
18. Eissa, A.M.F.; Olaj, Szappan, Kozmetika., 51(4), 155, 2002.
19. Yousef, E.A.A.; Zaki, M.E.A. and Megahed, M.G., Heterocyclic Communication, 9(3), 293, 2003.
20. Rauf, Abdul.; Sharma, Shweta. andGangal, Saloni., Arkivoc(xvi).,137, 2007.
21. Yasuharu Nakayama, Progress in Organic Coatings, 51, 280–299, 2004.
22. K and Raviraj S. Pattanashettar, *Ind. Eng. Chem. Res.*, *43* (17), pp 4979–4999, 2004.
23. Makoto Hori et al, Resinous compositions for a water paint, patent 2011.
24. TaiebBrahimi, F., Mohamed B. and Adil A. Othman, Arabian Journal of Chemistry, http:\ dx.doi.ovg\ 10.1016\ J. arabjc. 06.016, 2013.
25. S, Basuny AM, Arafat SM, J Oleo Sci. 64(9), 1019-32, 2015.
26. HimaniVarshaney, Aiman Ahmed, Abdul Rauf, Fohad M. Husain and Iqbal Ahmad, Journal of Saudi Chemical Society, http://dx.doi.org/10.1016/j.jscs.2014.04.00s.
27. HimaniVarshaney, Aiman Ahmed and Abdul Rauf, Arabian Journal of Chemistry http:// dx. doi. org/ 10.1016/j.arabjc.2014.08.002.
28. Weil I.K.; Scircon A.J. and Biscleine R.J.:JAOCS 37 p.295 (1960).
29. Indian Standard Specification, I.S, Standard Test Method for Water and Alkali Resistances, Philadelphia, ASTM: D 1647-89.
30. Standard Test Method for Solvent Resistance, Philadelphia, ASTM: D.
31. Standard Test Method for Bend Test, Philadelphia, ASTM : Da.
32. Standard Test Method for Adhesion, Philadelphia, ASTM: D.
33. Standard Test Method for Impact, Philadelphia, ASTM: D–Standard Test Method for Gloss, Philadelphia, ASTM: D–, 2004.
34. Standard Test Method for Film Hardness by Pencil Test, Philadelphia, ASTM: D–a, 2004.
35. A.M.F. EISSA, Anionic Surface Active Agents from fatty acid hydrazides containing heterocyclic moity, OLAJ, SZAPPAN, KOZMETIKA, 155-161, 2002.

11/1/2015