Iraqi Journal of Science, 2023, Vol. 64, No. 3, pp: 1041-1048
DOI: 10.24996/ijs.2023.64.3.1

ISSN: 0067-2904

Synthesis, Characterization and Evaluation of Some Meldrum's Acid Derivatives as Lubricant Additives

Hadeel A.K Hussien, Zainab A. K. Al-Messri*
Department of Chemistry, College of Science, University of Baghdad, Baghdad, Iraq

Received: 20/11/2021 Accepted: 3/3/2022 Published: 30/3/2023

Abstract

Aminomethylene Meldrum's acid derivatives were synthesized by a three--5 component, one-pot reaction of Meldrum's acid with triethyl orthoformate and different aromatic amines. The prepared compounds were characterized using fourier transform infrared (FT-IR), nuclear magnetic resonance (1H NMR and 13C NMR) and evaluated as anti-corrosion and anti-rust additives by blending with base lubricating oil, according to the American Society of Testing and Materials (ASTM-D130 and ASTM-D665). The blends of the synthesized compounds with the base lubricating oil showed better anti-rust and anti-corrosion effects than the base oil Blank).

Keywords: Meldrum's acid, One-pot reaction, Lubricant additives, Anti-rust, Corrosion inhibitors.

تحضير و تشخيص و تقييم بعض مشتقات حامض الملدرمز كمحسنات لزيوت التزييت

هديل عبد الكريم حسين ، زينب عبد الزهره خضير*
قسم الكيمياء، كلية العلوم، جامعة بغداد ، بغداد، العراق

الخلاصة

حضرت مشتقات5-aminomethylene Meldrum's acid من تفاعل الوعاء الواحد لثلاث مكونات و هي Meldrum's acid وtriethyl orthoformate و امينات اروماتية مختلفة و شخصت المركبات المحضرة بوساطة مطيافية الاشعة تحت الحمراء (FT-IR)و الرنين المغناطيسي النووي 1 H NMR , 13C NMR)) و قيمت المركبات المحضرة كمضادات للتاكل و الصدأ من خلال مزجها مع زيت الأساس من نوع 60 stock المجهز من شركة مصافي الوسط العراقية / مصفى الدورة اعتمادا على اختبارات ASTM-D130 و ASTM-D665 على التوالي. حيث اظهرت نتائج افضل من الزيت الاساس.

1. Introduction

Lubricants are significant classes of refining industry chemicals that are commonly utilized to prevent wear and friction between moving components in modern equipment by interposing a film of another substance in order to extend its lifetime. Internal combustion engines, vehicles, industrial gearboxes, compressors, turbines, and hydraulic systems all need lubricants [1,2]. The manufacturers have increased the performance of the oils using some

*Email: zainababdulzahra@sc.uobaghdad.edu.iq

additives. The additives include pour point depressants, detergents, dispersants, viscosity index improvers, antioxidants, anti-corrosion, and rust inhibitors [3]. Many additives serve multifunction, zinc dialkyl dithiophosphates, for example, are anti-corrosion and antioxidants that are best known for their anti-wear properties [4]. Over-based magnesium stearate was used in Iraqi lubricating oils as detergents, antioxidants, and viscosity improvers [5,6]. Some pyranopyrazole and pyranopyrimidine derivatives have been prepared and used as antioxidants and anti-corrosion [7,8]. 5-Alkylidene Meldrum's acid derivatives [9] have recently been produced and employed as multifunctional lubricating oil additives. Meldrum's acid is a powerful organic Chiron that is commonly utilized to produce natural products [10-12]. and heterocyclic compounds [13,14]. Meldrum's acid derivatives also exhibit a wide spectrum of biological activities; they act as antimicrobials [15-17], antituberculosis agents [18], antimalarial, and antioxidants [19]. In our work, 5-aminomethylene derivatives of Meldrum's acid were synthesized by multicomponent reactions (MCRs) of Meldrum’s acid, triethyl orthoformate, and different substituted aromatic amines. The synthesized derivatives were evaluated as multifunction additives (anti-corrosion and anti-rust) for medium lubricating oils.

2. Experimental

2.1. Materials and Instruments

All chemicals used were supplied by Fluka AG, Sigma-Aldrich, Merck, and BDH Chemicals. Meldrum’s acid was synthesized by condensing malonic acid with acetone in the presence of sulfuric acid and acetic anhydride [9]. Melting points were recorded using an Electro-thermal Stuart Scientific apparatus. Infrared spectra were recorded on the Shimadzu (8400S) Spectrophotometer at the University of Baghdad, College of Science, Department of Chemistry. Nuclear Magnetic Resonance spectra (^{1}H and ^{13}C) were recorded on a VARIAN model at 500 MHz and 125 MHz, respectively, using dimethyl sulfoxide (DMSO-d_6) as a solvent at the University of Tehran, Iran.

2.2. Test methods

2.2.1. Thin layer chromatography

TLC was achieved on sheets of aluminum, which were coated with silica gel (60) supplied by the Merck Company. Petroleum ether and ethyl acetate solvents in the ratio 3:2 were used as the eluent, and iodine vapor was used to detect spots.

2.2.2. Rust preventing characteristics test (ASTM - D665)

A polished steel rod was immersed in a mixture of lubricant and water (300 mL: 30 mL), and warmed to 60 °C for 4 hours. The test rod was checked for signs of rust. This test was repeated twice, and both test rods must be rust-free to pass [20].

2.2.3. Copper corrosion test (ASTM - D130)

A polished copper strip was immersed in a sample of lubricant (30 mL) and heated at 100 °C for 3 hours. Upon completion of the test, the copper strip was cleaned and inspected. The stains on the copper strip were compared using the ASTM D130 color scale, which ranges from 1a to 4c. The ratings progress down the scale as the test coupon's corrosion staining worsens, with a 4c (the worst) rating, which appears as blackened, corroded, and pitted. While 1a denotes freshly polished copper coupons with minor staining that is barely noticeable, 1b denotes a slight tarnish [21].

*2.3. Synthesis of 5-aminomethylene Meldrum’s acid derivatives **1- 10***

A mixture of Meldrum’s acid (1.44 g, 10 mmol), appropriate amine (10 mmol) and triethyl orthoformate (5 mL) in anhydrous acetonitrile (50 mL) was heated to reflux for two hours.

Then, the solvent was evaporated to give the crude product, which was recrystallized from ethanol [22].

*2,2-Dimethyl-5-(((4-nitrophenyl)amino)methylen)-1,3-dioxan-4,6-dione (**1**)*

$C_{13}H_{12}N_2O_6$, 292.25 g/mole, yellow crystal, m.p. 220-222 ^oC, yield 55%. FT-IR (cm^{-1}); 3234 (N-H), 3076 (νC-H aromatic), 2920, 2849 (νC-H aliphatic), 1732, 1678 (νC=O), 1629 (νC=C), 1516 (NO_2 asymmetric), 1344 (ν NO_2, symmetric), 862 (δ o-o-p) (1,4-aromatic substitution). ^1H NMR (δ, ppm); 11.40 (1H, NH), 8.70 (1H, CH), 8.68-7.84 (4H, Ar-H), 1.70 (6H, $2CH_3$). ^{13}C NMR (δ, ppm); 166.1 (C=O), 155.6, 140.3, (C=CH), 146.9, 122.1-107.4 (Ar-C), 91.6 (O-C-O), 29.2 (6H, $2CH_3$).

*2,2-Dimethyl-5-((p-tolyl-amino)methylen)-1,3-dioxan-4,6-dione (**2**)*

$C_{14}H_{15}NO_4$, 261.28 g/mole, green-yellowish crystal, m.p. 152-154 ^oC, yield 74%. FT-IR (cm^{-1}); 3256 (νN-H), 3078 (νC-H aromatic), 2991, 2939 (νC-H aliphatic), 1736, 1684 (νC=O), 1628 (νC=C), 825 (δ o-o-p, 1,4-aromatic substitution). ^1H NMR (δ, ppm); 11.22 (1H, NH), 8.53 (1H, CH), 7.46-7.24 (4H, Ar-H), 2.33 (3H, CH_3), 1.68 (6H, $2CH_3$). ^{13}C NMR (δ ppm); 165.5 (C=O), 138.4, 155.7 (C=CH), 132.7-106.7 (Ar-C), 88.9 (O-C-O), 29.1 (6H, $2CH_3$), 23.1 (3H, CH_3).

*2,2-Dimethyl-5-(((furan-2-methyl)amino)methylen)-1,3-dioxan-4,6-dione (**3**)*

$C_{12}H_{13}NO_5$, 251.24 g/mole, orange crystal, m.p. 134-136 ^oC, yield 51%. FT-IR (cm^{-1}); 3261 (νN-H), 3124 (νC-H aromatic), 2945 (νC-H aliphatic), 1720, 1676 (νC=O), 1616 (νC=C), 1011 (C-O-C). ^1H NMR (δ ppm); 9.92 (1H, NH), 8.28 (1H, CH), 7.68-6.42 (3H, Furan-H), 4.72 (2H, CH_2), 1.56 (6H, $2CH_3$). ^{13}CNMR (δ ppm); 165.9 (C=O), 152.4, 162.2 (C=CH), 146.1-106.2 (Furan-C), 90.0 (O-C-O), 21.0 (6H, $2CH_3$), 47.9 (CH_2).

*2,2-Dimethyl-5-((thiazol-2-yl-amino)methylen)-1,3-dioxan-4, 6-dione (**4**)*

$C_{10}H_{10}N_2O_4S$, 254.26 g/mole, brown powder, m.p. 206-208 ^oC, yield 43%. FT-IR (cm^{-1}); 3230 (νN-H), 3084 (νC-H aromatic), 2989, 2833 (νC-H aliphatic), 1711, 1666 (νC=O), 1624 (νC=C), 1583 (C=N). ^1H NMR (δ ppm); 11.47 (1H, NH), 8.75 (1H, CH), 8.75-6.89 (2H, thiazole-H), 1.68 (6H, $2CH_3$). ^{13}C NMR (δ ppm); 165.6 (C=O), 149.7, 154.0 (C=CH), 129.4-106.9 (thiazole-C), 89.0 (O-C-O), 29.0 (6H, $2CH_3$).

*2,2-Dimethyl-5-(((4-chlorophenyl)amino)methylen)-1,3-dioxan-4,6-dione (**5**)*

$C_{13}H_{12}ClNO_4$, 281.69 g/mole, white powder, m.p. 216-218 ^oC, yield 74%. FT-IR (cm^{-1}); 3259 (νN-H), 3097 (νC-H aromatic), 2995, 2943 (νC-H aliphatic), 1720, 1676 (νC=O), 1628 (νC=C), 827 (δ o-o-p, 1,4-aromatic substitution), 638 (C-Cl).

*2,2-Dimethyl-5-(((2-thioxo-2,3-dihydrothiazol-5-yl)amino)methylen)-1,3-dioxan-4,6-dione (**6**)*

$C_{10}H_{10}N_2O_4S_2$, 286.32 g/mole, yellow powder, m.p. 232-234^oC, yield 34%. FT-IR (cm^{-1}); 3248 (νN-H), 3124 (νC-H aromatic), 2924 (νC-H aliphatic), 1740, 1716 (νC=O), 1608 (νC=C), 3340 (NH-C=S), 1329 (C=S). ^1H NMR (δ ppm); 13.13 (1H, NH), 7.13 (1H, CH), 1.03 (6H, $2CH_3$). ^{13}C NMR (δ ppm); 161.9 (C=O), 137.1, 145.4 (C=CH), 116.4-104.2 (Ar-C), 73.0 (O-C-O), 27.3, 28.3 (6H, $2CH_3$) 181.3 (C=S).

*2,2-Dimethyl-5-((pyridin-2-yl-amino)methylen)-1,3-dioxan-4,6-dione (**7**)*

$C_{12}H_{12}N_2O_4$, 248.24 g/mole, orange powder, m.p. 172-174 ^oC, yield 43%. FT-IR cm^{-1}; 3256 (νN-H), 3077 (νC-H aromatic), 2988, 2943 (νC-H aliphatic), 1736, 1684 (νC=O), 1620 (νC=C), 1560 (C=N). ^1H NMR (δ ppm); 11.34 (1H, NH), 9.22 (1H, CH), 8.44-7.27 (Pyridine-

H), 1.68 (6H, $2CH_3$). ^{13}C NMR (δ ppm); 163.1 (C=O), 139.2, 149.4 (C=CH), 137.0-104.1 (Pyridine-C), 87.8 (O-C-O), 26.4 (6H, $2CH_3$).

*2,2-Dimethyl-5-((pyrimidin-2-yl-amino)methylen)-1,3-dioxan-4,6-dione (**8**)*

$C_{11}H_{11}N_3O_4$, 249.23 g/mole, pink powder, m.p. 210-212 °C, yield 75%. FT-IR (cm^{-1}); 3313 (νN-H), 3068 (νC-H aromatic), 2995, 2943 (νC-H aliphatic), 1736, 1693 (νC=O), 1612 (νC=C), 1562 (C=N.). ^1H NMR (δ ppm); 11.04 (1H, NH), 9.13 (1H, CH), 8.84-7.42 (3H, pyrimidine-H), 1.75 (6H, $2CH_3$). ^{13}C NMR (δ ppm); 166.7 (C=O), 158.0, 162.0 (C=CH), 153.8-107.7 (pyrimidine-C), 92.5 (O-C-O), 29.3 (6H, $2CH_3$).

*2,2-Dimethyl-5-((benzo[d]thiazol-2-yl-amino)methylen)-1,3-dioxan-4,6-dione (**9**)*

$C_{14}H_{12}N_2O_4S$, 304.32 g/mole, dark red powder, m.p 183-185 °C, yield 31%. FT-IR (cm^{1}) ; 3242 (νN-H), 3065 (νC-H aromatic), 2989, 2920 (νC-H aliphatic), 1728, 1691 (νC=O), 1610 (νC=C), 1560 (C=N). ^1H NMR (δ ppm); 12.35 (1H, NH), 9.10 (1H, CH), 8.05-7.01 (4H, Ar-H), 1.61 (6H, $2CH_3$). ^{13}C NMR (δ ppm); 162.6 (C=O), 142.7, 152.9 (C=CH), 136.0-107.2 (Ar-C), 90.6 (O-C-O), 29.3 (6H, $2CH_3$).

*2, 2-Dimethyl-5-(((1-(1H-indol-3-yl)-ethyl)amino)methylen)-1,3-dioxan-4,6-dione (**10**)*

$C_{17}H_{18}N_2O_4$, 314.34 g/mole, brown crystal, m.p. 246-248 °C, yield 34%. FT-IR (cm^{-1}); 3292 (ν N-H), 3013 (νC-H aromatic), 2987,2910, (νC-H aliphatic), 1788,1701 (νC=O), 1606 (νC=C). ^1H NMR (δ ppm); 11.04 (1H, NH), 8.21 (1H, CH), 7.59-6.99 (5H, Ar-H), 1.56 (6H, $2CH_3$), 1.07 (2H, CH_2). ^{13}C NMR (δ ppm); 161.9 (C=O), 137.3, 158.6 (C=CH), 127.0-109.7 (Ar-C), 99.4 (O-C-O), 23.3 (6H .$2CH_3$), 55.2 (2H, CH_2).

2.4. Formulation of oil blends

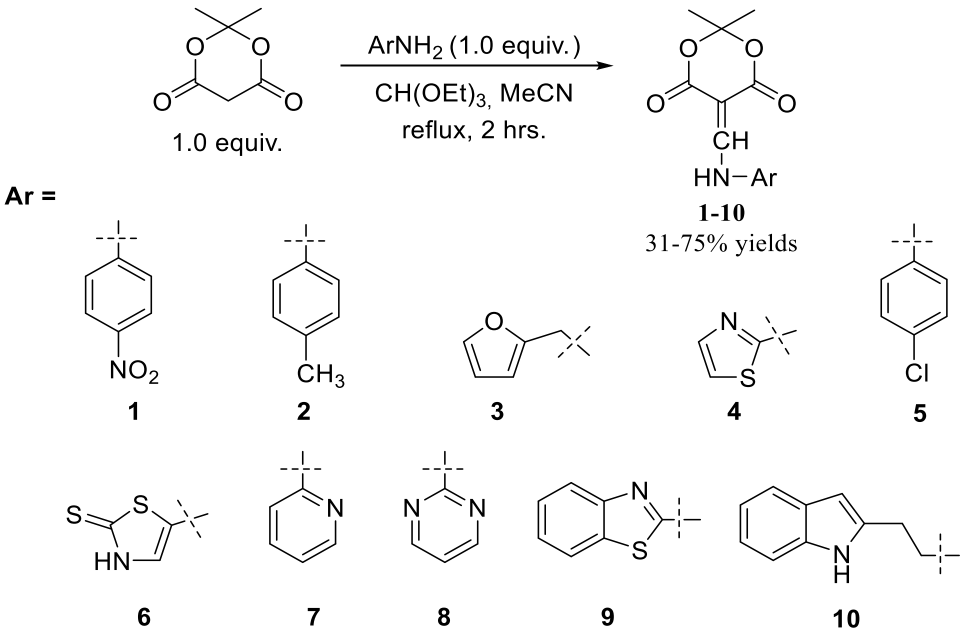
Blends of 5-aminomethylene Meldrum's acid compounds **1-10** were achieved by dissolving and mixing 0.2% weight/weight of each compound with base oil sixty stock at 70 °C with stirring for one hour and then leaving the mixture stirring for another hour at room temperature [5]. The properties of the base oil delivered by the Iraqi Midland Refineries Company are listed in Table 1.

Table 1: The properties of the (sixty stock) base oil

No.	Specifications	Properties	Test method
1	Kinematic viscosity (mm^2/s) at 40 °C	58.030	ASTM/ D445
2	Kinematic viscosity (mm^2/s) at 100 °C	8.170	ASTM/ D445
3	Viscosity index (V.I)	105	ASTM/ D2270
4	Pour point (P.P)	0.00	ASTM/ D97
5	Specific gravity	0.88430	ASTM/ D4052
6	Flash point	243	ASTM/ D92
7	Rust preventing	Fail	ASTM/ D665
8	Copper corrosion	2a	ASTM/ D130
9	Color	2	ASTM/ D1500

3. Results and discussion

Meldrum's acid was reacted with triethyl orthoformate and various aromatic amines in a one-pot, two-step multicomponent reaction (MCR) to produce the 5-aminomethylene Meldrum's acid derivatives **1-10**, as shown in Scheme 1.



Scheme 1 - Synthetic route of 5-aminomethylene Meldrum's acid derivatives

The structure of the synthesized compounds was confirmed by FT-IR, ^{1}H NMR, and ^{13}C NMR spectroscopies. The FT-IR spectra showed stretching bands between 3313 and 3230 cm^{-1} attributed to the νN-H bands, and bands between 3124 and 3013 cm^{-1} are due to the aromatic C-H stretching. Aliphatic C-H stretching appeared at 2995-2833 cm^{-1}. The C=C stretching is represented by a band in the region 1629-1606 cm^{-1}. Meldrum's acid's carbonyl groups showed a strong stretching band between 1788 and 1666 cm^{-1} [23]. The ^{1}H NMR spectra of the compounds showed signals at 13.13-9.92 ppm for the NH proton, 9.22-7.13 ppm for the olefinic C-H proton [24]. The signals from 8.84 to 6.42 ppm belong to the aromatic protons. Finally, the signals between 1.75 and 1.03 ppm are due to the two methyl groups [25]. The ^{13}C NMR spectra showed characteristic signals. The signals from 162.2 to 137.1 ppm are attributed to the olefinic carbons. The signals between 153.8 and 104.1 ppm are for aromatic carbons. The signals at 99.4-73.0 ppm are for the quaternary ketal carbons. The two methyl groups of Meldrum's acid appeared between 29.3 and 21.0 ppm [26]. The produced compounds' oil blends were tested for anti-rust and anti-corrosion properties using the American Society of Testing and Materials ASTM-D665 and ASTM-D130, respectively. Most oil blends of synthetic derivatives passed the rust prevention test compared to base oil (blank), which failed the test but showed good anticorrosion properties. The examined copper strip appeared orange with slight tarnish (1a or 1b), as compared with the base oil (blank), which appeared red with moderate tarnish, as shown in Tables 2 and 3 [27].

Table 2: ASTM copper strip corrosion classifications

Classification	Designation	Description
1a	Slight tarnish	Light orange (newly polished strip)
1b	Slight tarnish	Dark orange
2a	Moderate tarnish	Dark red
2b	Moderate tarnish	Violet
2c	Moderate tarnish	Multi colored and violet-blue
2d	Moderate tarnish	Silvery
2e	Moderate tarnish	Brassy or golden
3a	Dark tarnish	Magenta overcast
3b	Dark tarnish	Multi colored and red-green
4a	Corrosion	Black, brown, or dark gray
4b	Corrosion	Matt black
4c	Corrosion	Glossy black

Table 3: Results of rust preventing and copper corrosion tests

No.	Rust preventing test (ASTM/D665)	Copper corrosion test (ASTM/D130)
Blank	Fail	2a
1	Pass	1b
2	Fail	2c
4	Fail	1b
5	Fail	1b
6	Pass	1a
7	Pass	1a
8	Pass	1b
9	Pass	1a
10	Pass	1a

4. Conclusion

A one-pot multicomponent reaction was used to successfully synthesize various derivatives of 5-aminomethylene Meldrum's acid. The prepared compounds were assessed as antirust and corrosion inhibitors for engine lubricating oil by blending a 0.2 weight percentage with base oil. Most blends had good anti-rust and anti-corrosion properties.

References

[1] K. C. Ludema and O. O. Ajayi, "*Friction, wear, lubrication: a textbook in tribology" CRC press*, 2018.

[2] G. M. Parsons, "*Biodiesel and Engine Lubrication, Part 2," Chevron Products Company Publishers, San Ramon, Califonia*, 2007.

[3] L. R. Rudnick, "*Lubricant Additives: Chemistry and Applications" 2ed, Taylor & Francis Group, New York*, 2009.
[4] D. W. Johnson, "*The tribology and chemistry of phosphorus-containing lubricant additives", IntechOpen Book Series*, 2016.
[5] A. H. A. Mohammed, M. R. Ahmad, and Z. A. Al-Messri, "Synthesis, characterization and evaluation of overbased magnesium fatty acids detergent for medium lubricating oil," *Iraqi Journal of Chemical and Petroleum Engineering*, vol. 14, no. 3, pp. 1-9, 2013.
[6] A. H. A. Mohammed, M. R. Ahmad, and Z. A. K.Al-Messri, *Iraqi Patent,* 3967, 2014.
[7] M. R. Ahmad, A. H. A. Mohammed, Y. Ali, and Z. A. K. Al-Messri, "Synthesis, Characterization and Evaluation of Some Pyranopyrazoles and Pyranopyrimidines Derivatives as Antioxidants for Lubricating Oils," *Iraqi Journal Science*, vol. 55, no. 1, pp. 1-11, 2014.
[8] A. R. Salih and Z. A.K. Al-Messri, "Synthesis of pyranopyrazole and pyranopyrimidine derivatives using magnesium oxide nanoparticles and evaluation as corrosion inhibitors for lubricants," *Eurasian Chemical Communications*, vol. 3, no. 8, pp. 533-541, 2021.
[9] H. A. K. Hussien and Z. A. K. Al-Messri, "Synthesis, Characterization and Evaluation of 5-alkylidene Meldrum's acid derivatives as Multifunction Lubricating Oil Additives", *Eurasian Chemical Communications*, vol. 3, no. 9, pp. 598-605, 2021.
[10] P. Mahulikar and R. Mane, "Application of Meldrum's acid in natural product synthesis. Synthesis of ar-turmerone and α-curcumene," *Journal of Chemical Research*, vol. 2006, pp. 15-18, 2006.
[11] S. Ghosh and C. K. Jana, "Rapid access to cinnamamides and piper amides via three component coupling of arylaldehydes, amines, and Meldrum's acid," *Green Chemistry*, vol. 21, pp. 5803-5807, 2019.
[12] M. M. Heravi, F. Janati, and V. Zadsirjan, "Applications of Knoevenagel condensation reaction in the total synthesis of natural products," *Monatshefte für Chemie-Chemical Monthly*, vol. 151, pp. 439-482, 2020.
[13] S. Asadi, F. Alizadeh-Bami, and H. Mehrabi, "Efficient synthesis of N-amino pyridine-2, 6-diones via a novel one-pot, the three-component reaction between 2-cyanoacetohydrazide, Meldrum's acid, and aryl aldehyde," *Organic Chemistry Research*, vol. 6, no. 2, pp. 212-218, 2020.
[14] S. Asadi, M. Hajipour, and H. Mehrabi, "Meldrum's acid as an excellent catalyst for the facile, efficient and one-pot synthesis of N, 3-diarylimidazo [1,2-a]pyridin-2-amines", *Arkivoc*, part x, pp. 27-39, 2021.
[15] N. Janković, J. Muškinja, Z. Ratković, Z. Bugarčić, B. Ranković, M. Kosanić, and Srđan Stefanovićc, "Solvent-free synthesis of novel vanillidene derivatives of Meldrum's acid: biological evaluation, DNA and BSA binding study," *RSC advances,* vol. 6, no. 45, pp. 39452-39459, 2016.
[16] G. M. Sampaio, A. M. Teixeira, H. D. Coutinho, D. M. S. Junior, P. T. Freire, R. R. Bento, and L E. Silva, "Synthesis and antibacterial activity of a new derivative of the Meldrun acid: 2,2-dimethyl-5-(4H-1,2,4-triazol-4-ylaminomethylene)-1,3-dioxane-4,6-dione(C9H10N4O4),"
[17] *EXCLI journal*, vol. 13, pp. 1022-1028, 2014.M. M. C. da Silva, J. B. de Araújo-Neto, A. C. J. de Araújo, P. R. Freitas, C. D. de M Oliveira-Tintino, I. M. Begnini, R. A. Rebelo, L. E. da Silva, S. L. Mireski, M. C. Nasato, M. I. L. Krautler, J. Ribeiro-Filho, H. D. M. Coutinho, and S. R. Tintino, "Potentiation of Antibiotic Activity by a Meldrum's Acid Arylamino Methylene Derivative against Multidrug-Resistant Bacterial Strains," *Indian Journal of Microbiology*, vol. 61, no. 1, pp. 100-103, 2021.
[18] N. Rajeev, K. S. Sharath Kumar, Y. K. Bommegowda, K. S. Rangappa, and M. P. Sadashiva, "Catalyst free sequential one-pot reaction for the synthesis of 3-indole propanoates/propanoic acid/propanamides as antituberculosis agents," *Journal of the Chinese Chemical Society*, vol. 68, no. 1, pp. 39-44, 2021.
[19] H. S. Sandhu, S. Sapra, M. Gupta, K. Nepali, R. Gautam, S. Yadav, R. Kumar, S. M. Jachak, M. Chugh, M. K. Gupta, O. P. Suri, and K. L. Dhar, "Synthesis and biological evaluation of arylidene analogues of Meldrum's acid as a new class of antimalarial and antioxidant agents," *Bioorganic and Medicinal Chemistry*, vol. 18, no. 5, pp. 5626-5633, 2010.

[20] H. Ries, H. Cook, and C. Loane, "*Monomolecular Films of Rust-Preventive Additives" in Symposium on Steam Turbine Oils*, 1957.
[21] ASTM International, Designation: D130, Standard Test Method for Corrosiveness to Copper from Petroleum Products by Copper Strip Test, USA, 2017.
[22] H. Briehl, A. Lukosch, and C. Wentrup, "Reactive nitrogenous molecules from Meldrum's acid derivatives, pyrrole-2, 3-diones, and isoxazolones," *The Journal of Organic Chemistry*, vol. 49, no. 15, pp. 2772-2779, 1984.
[23] R. M. Silverstein, F. X. Websters, and D. J Kiemle, "*Spectrometic identification of organic compounds", 7th ed., New York, John Wiely and Sons*, 2005.
[24] S. M. H. Al-Majidi, H. A. R. Ibrahim, and Y. A. H. AL-Issa, Synthesis and Identification of Some New Derivatives of ([Benzyl Thio) Benzimidazole-N-(Methylene-5-Yl)]-4,5-Di Substituted 1,2,4-Triazole and Evaluation of Their Activity as Antimicrobial and Anti-Inflammatory Agents, *Iraqi Journal of Science*, vol. 62, no. 4, pp. 1054-1065, 2021.
[25] J. H. Li and J. H. He, "A one-pot reaction of Meldrum's acid with ethyl orthoformate and hydrazines," *Journal of Chemical Research*, vol. 2006, pp. 37-38, 2006.
[26] B. D. Mistry, "*A Handbook of Spectroscopic Data (UV, IR, PMR, ^{13}C NMR and Mass Spectroscopy)", Oxford Book Company, Jaipur, India*, 2009.
[27] M. A. Khiar, R. Brown, and P. Lewin, "Sacrificial copper strip sensors for sulfur corrosion detection in transformer oils," *Measurement*, vol. 148, p. 106887, 2019.