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Zeolite Catalysed evaluation of substituted 2-imidazolones derivatives

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Abstract:

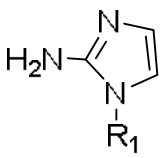
2-imidazolones have gained immense significance in human life due to variety of their applications. Though there are several methods for the synthesis of 2-imidazolones, most of them required longer reflux time of 8 to 10 hours. Hence the proposed work was undertaken to workout simple methodology for the synthesis of 2-imidazolones and to improve the yield of the products, by employing Zeolite as a catalyst. The work presented here describes the synthesis of some substituted 2-imidazolones obtained from substituted benzoins and urea in CH3COOH as a solvent in presence of Zeolite as a catalyst. Substituted benzoins in turn were obtained from aromatic aldehydes by their condensation in presence of aqueous NaCN. The characterisation of synthesized compounds was made on the basis of chemical properties, elemental and spectral analysis.

Keywords:

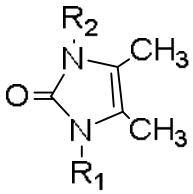
Substituted benzoins, Urea, Methyl urea, Phenyl urea, Zeolite catalyst, 2-imidazolones

INTRODUCTION

2-Imidazolones are the heterocyclic compounds containing nitrogen atoms at 1 and 3 position and C=O group at 2 position. Imidazolones are believed to be associated with several pharmacological activities. Many natural products are believed to contain imidazolones. For example Leucetta and the Oroidin families of alkaloids have been reported to contain either 2-aminoimidazole (1) or 2-imidazolone (2) moiety.

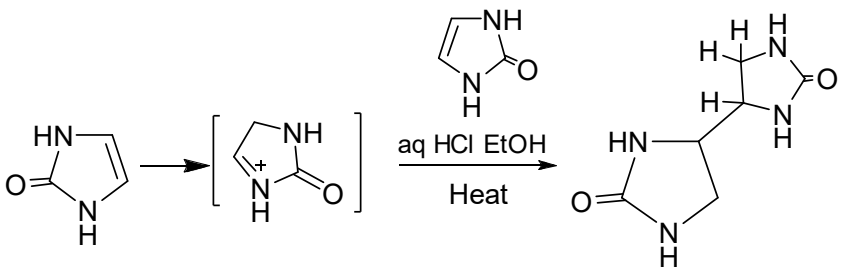


(1)



(2)

1,3 Azoles (e.g. 2-imidazolones) are found to exist in their carbonyl tautomeric forms. There is less aromatic character in such systems which can be illustrated by the acid catalyzed dimerization of 2–imidazolone (3) which acts as an enamide in the process.

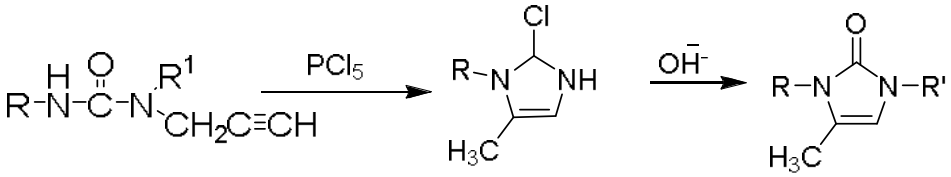


(3)

The Leucetta and Oroidin families of alkaloids [15] have been identified which contain either 2-aminoimidazole or 2-imidazolone moiety[16-17].

Glass D et al.[18] reported 4-(4-Guanidinobenzoyl)-2-imidazolones and related compounds having phosphodiesterase inhibitors and novel cardio tonics with combined histamine H_2 receptor agonist and PDE 111 inhibitor activity.

Stoffel and speziale[19] described the preparation of 2-imidazolones by a novel ring closure of propynylureas with phosphorous pentachloride. 2-imidazolone (5) was obtained via a stable isolable imidazolium chloride (4).



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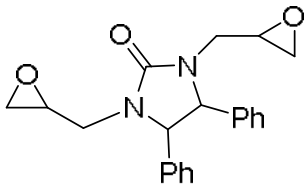
Marie Pascale[20] synthesized several 2-imidazolone derivatives (6) and screened their fungicidal and herbicidal activities.



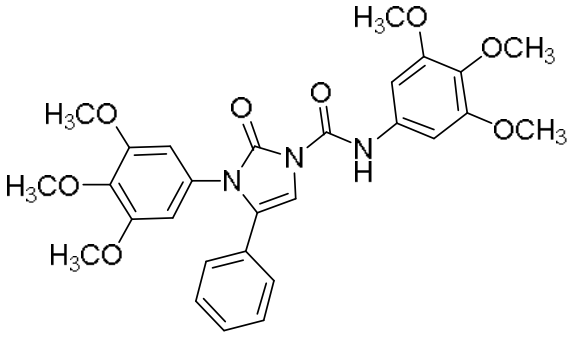
(6)

Jie-Fei Cheng et al.[21] carried out A traceless solid phase synthesis of 2-imidazolones. Polymer-bound glycerol resin was reacted with bromo acetaldehyde diethyl acetal to give the cyclic acetal bromide on the solid support.

Inas M AlNashef[22] described a novel method for the synthesis of 2-imidazolones. The superoxide ion electrochemically generated reduction of oxygen or chemically generated by dissolving potassium superoxide in ionic liquids, react with alkyl imidazolium cations of imidazolium based ionic liquids at room temperature and atmospheric pressure to give the corresponding 2-imidazolones in excellent yield.

Tomokazu Katahira et al.[23] studied Stereo selective intermolecular radical addition of polyhaloacyl pendant groups to the 1,3-dihydro-2-imidazolone moiety the chiral synthesis of threo- diamino carboxylic acids.

Xue et al.[24] synthesized 2,3-Dihydro-*N*,3-bis(3,4,5-trimethoxyphenyl)-4-(substitutedphenyl)-2-oxo-imidazole-1-carboxamides(7) and 1-acetyl-1,3-dihydro-3-(3,4,5-trimethoxyphenyl)-4-(substitutedphenyl)-2*H*-imidazol-2-ones (8) and studied their antitumor activities.



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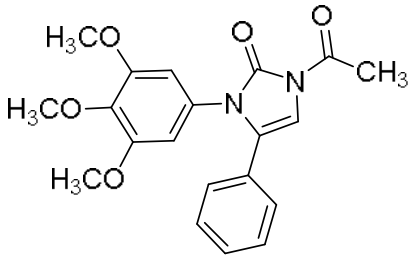
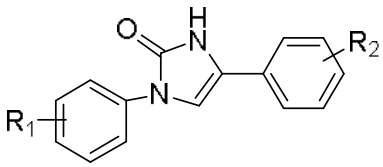


(8)

Congiu et al.[25] reported in vitro antitumor activity of new 1,4-diarylimidazole-2-ones (9) and their 2-thione analogues has been conceded by Compounds bearing a 3,4,5-trimethoxyphenyl ring linked to either N-1 or C-4 position of the imidazole (10) core demonstrated an interesting profile of cyto toxicity with preferential activity against leukemic cell lines. Compound exhibited a potent antitumor activity against MOLT-4 (GI50 = 20 nM) and SR (GI50 = 32 nM) cell lines.



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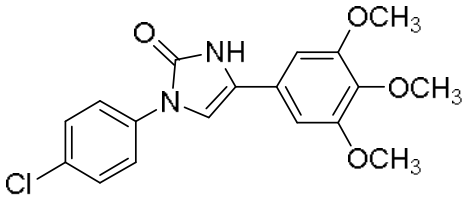
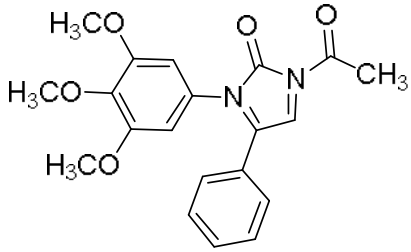


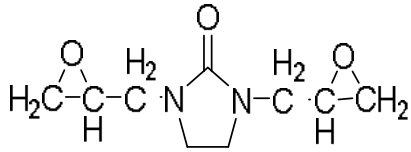
(10)

Xue et al.[26] synthesized and evaluated imidazol-2-one derivatives (11) as potential antitumor agents.



(11)

Kortkikh[27] prepared below mentioned compound (12) and tested for its antitumor activity.



(12)

Grigat[28] reported imidazoline derivatives (13) having below mentioned general structure which were found to be useful as a plant protection agents.

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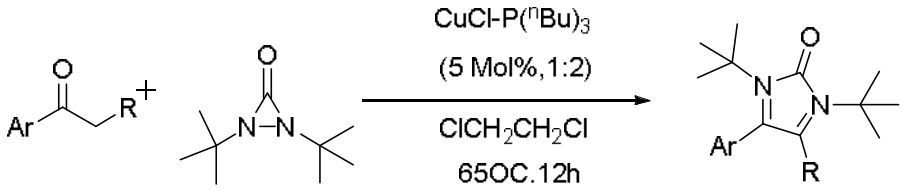
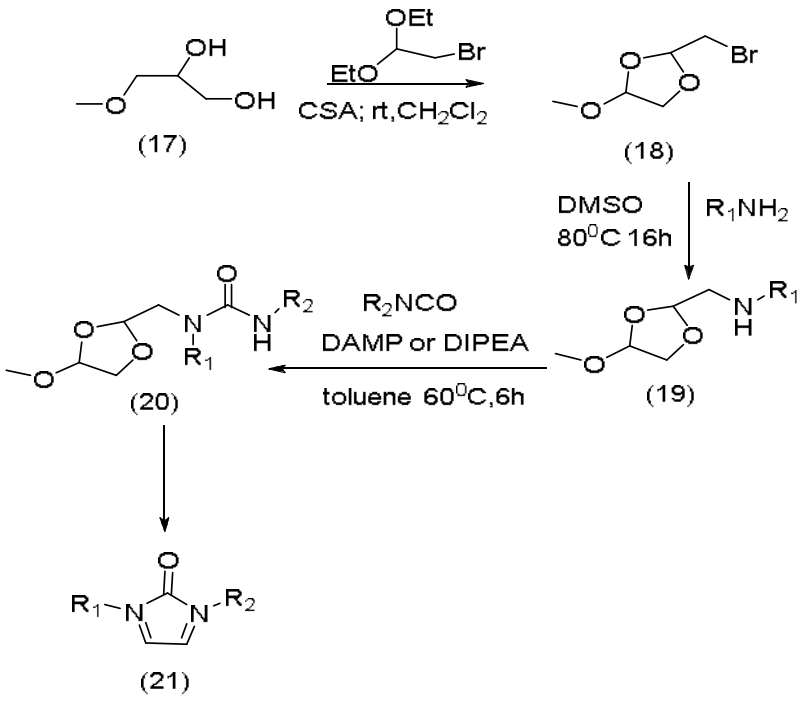
(13)

Zhao et al.[29] synthesised a variety of imidazolinone derivatives (16) via B-amination process of aryl ketones (14) under mild conditions using CuCl as a catalyst and di-tert-butyldiaziridinone (15) as the source of nitrogen.



(14) (15) (16)

Cheng et al.[30] reported the solid-phase synthesis of analogues of 2-imidazolones type (21). In their synthetic method, the polymer-bound glycerol resin (17) was first reacted with bromoacetaldehyde diethyl acetal to give the cyclic acetal bromide (18) on the solid support. Further, amination of the resin-bound acetal bromide was achieved to afford compound (19), which was further treated with isocyanates to afford the resin-bound urea acetals (20). Upon treatment of (20) with TFA, aldehyde urea was released from the resin, which immediately cyclized to the desired 2-imidazolones (21).



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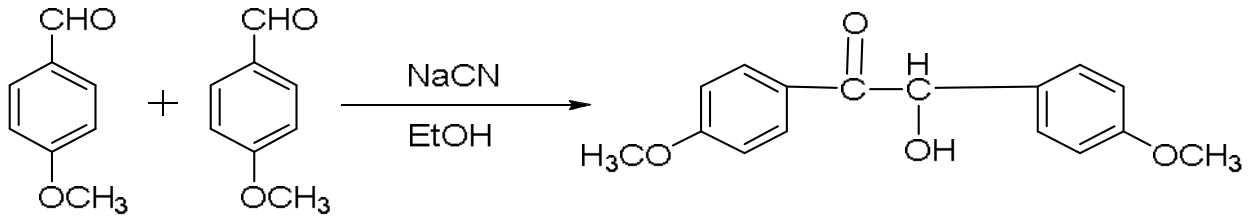
Experimental Method

Synthesis of 4, 4'-dimethoxybenzoin.

Method: In a round bottom flask, took 13.6 gms (0.1 mol) of anisaldehyde, added to it about 50 ml of ethyl alcohol. The mixture was shaken well. To this mixture added 4.9 gms aq. solution of sodium cyanide (0.1 mol). The reaction mixture was refluxed for 30-40 minutes. Cooled reaction mixture and poured it to ice cold water, obtained solid yellow product. Recrystallised it from water-ethanol mixture.

Yield: 65% Melting point: 113°C

Reaction:



Properties and Constitution of the Compound

1. It is an yellowish crystalline solid having melting point 113°C.
2. With NH_2OH in alkaline medium, it formed oxime.
3. With 1% solution of m–dinitrobenzene, it gave red colour on addition of dilute NaOH solution indicating presence of carbonyl group (C=O).
4. From analytical data, the molecular formula of the compound was found to be $C_{16}H_{16}O_4$, molecular weight 272.30.

Properties and Constitution of the Compound (1b)

The IR spectrum of the compound showed the following main absorption bands.

Absorption Observed (cm^{-1})	Assignment	Literature Value (cm^{-1})
3427	O–H Str	3570-3400
3066	Ar,C–H str	3100-3000
2920	Aliph, C–H	2980-2840
1653	C=O str	1850-1630
1507	Ar, C=C str	1500-1450
1310	C–O str	1350-1210

Properties and Constitution of the Compound (1b)

The ^{1}H-NMR spectrum of the compound showed the chemical shifts which can be correlated as given below.

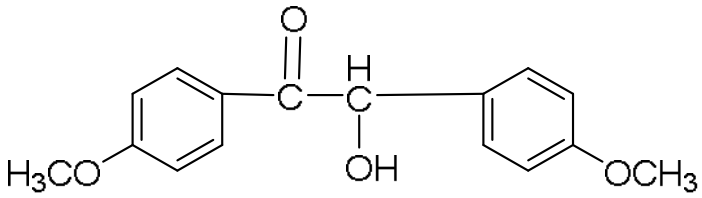
Chemical Shifts (δ)	Multiplicity	Assignment
7.86-7.85	d	4H,Ar–H
7.13-7.12	d	4H,Ar–H
3.87	s	1H, CH–OH
3.37	s	6H, $–OCH_3$
2.50	s	1H, Aliph, C–H

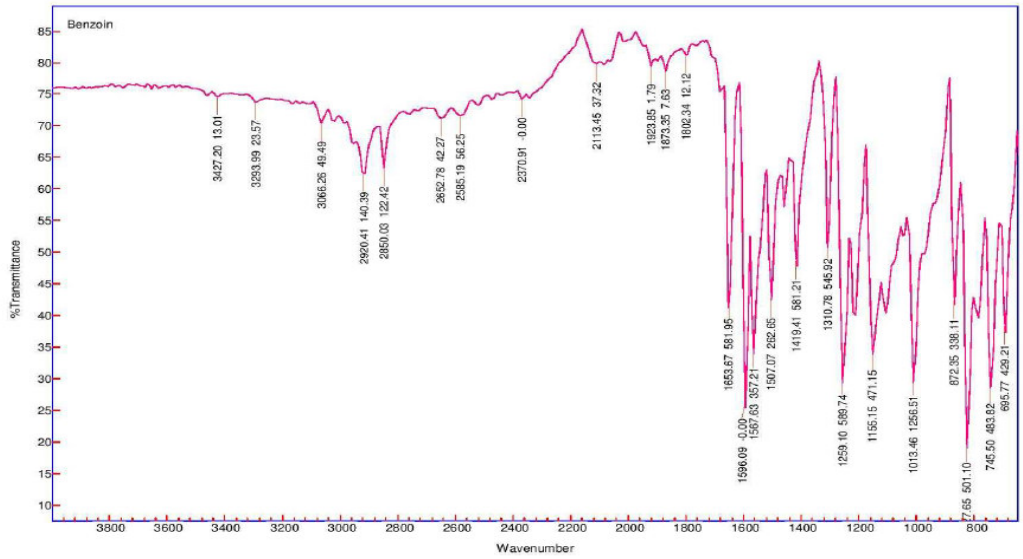
Properties and Constitution of the Compound (1b)

Elemental Analysis for $C_{16}H_{16}O_4$ (272.30)

Element (%)	C	H
Calculated	70.58	5.92
Found	70.55	5.90

On the basis of chemical properties, elemental and spectral analysis of the compound 1b was assigned the structure as 4,4'-dimethoxybenzoin.





SPECTRUM NO. 27

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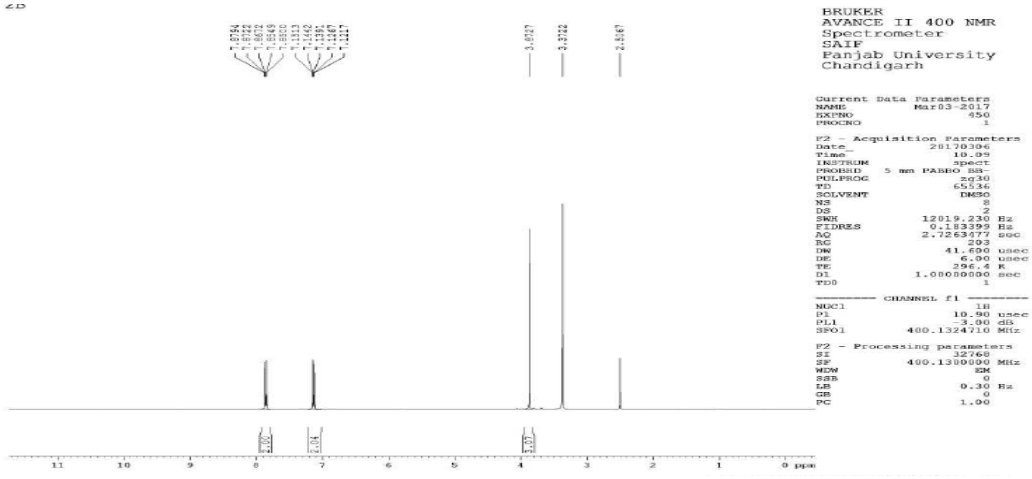
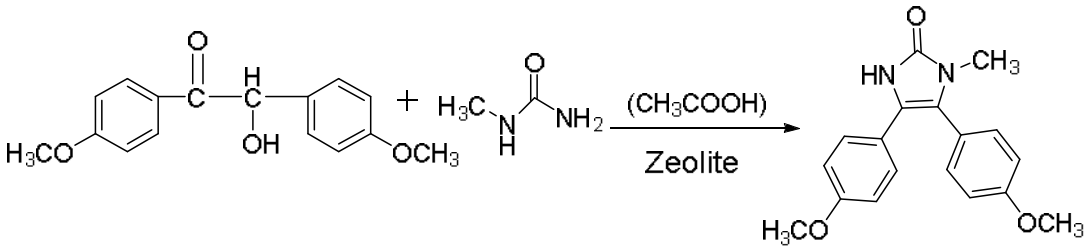


Step-II Synthesis of 1-H-3-methyl-4(4-methoxyphenyl)-5-(4-methoxyphenyl)-2-imidazolone

Method: Took 4,4'-dimethoxybenzoin 2.72 gms (0.01 mol) in a round bottom flask, added to it glacial acetic acid (20ml). The mixture was warmed slightly to dissolve the solute. To this solution, added methyl urea 0.74 gm (0.01mol), followed by zeolite (1gm) as a catalyst. The reaction mixture was refluxed for three hours. Allowed it to cool and poured it to ice cold water. The solid yellow product formed was filtered, washed 2,3 times with cold water and recrystallized from water-ethanol mixture.

Yield: 62% Melting point: 180°C

Reaction:



(2d)

Properties and Constitution of the Compound (2d)

1. It is an yellow crystalline solid having melting point 180°C.
2. The compound gave positive test for nitrogen.
3. It decolorized alkaline $KMnO_4$ indicating presence of C=C linkage.
4. With 1% solution of m–dinitrobenzene, it gave red colour on addition of dilute NaOH solution indicating presence of carbonyl group (C=O).
5. From analytical data, the molecular formula of the compound was found to be $C_{18}H_{18}N_2O_3$, molecular weight 310.35.

Experiment No.	Compound	Melting point	Yield
137	2d	180°C	62%

6. The IR spectrum of the compound showed the following main absorption bands.

Absorption Observed (cm^{-1})	Assignment	Literature Value (cm^{-1})
3650	N–H str	3600-3200
3073	Ar, C–H str	3100-3000
2922	Aliph, C–H str	2980-2840
1678	C=O str	1850-1630
1600	C=N str	1690-1620
1507	Ar, C=C str	1500-1450
1293	C–N str	1360-1310
1245	C–O str	1350-1210

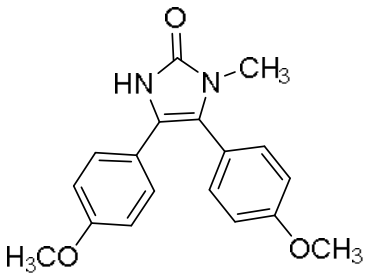
7. The ^{1}HNMR spectrum of the compound showed the chemical shifts which can be correlated as given below.

Chemical Shifts (δ)	Multiplicity	Assignment
7.57	s	1H, N–H
7.26-7.22	m	4H, Ar–H
7.17-7.05	m	4H, Ar–H
3.37	s	6H, $–OCH_3$
2.52	s	3H, $–CH_3$

8. Elemental Analysis for $C_{18}H_{18}N_2O_3$ (310.35)

Element (%)	C	H	N
Calculated	69.66	5.85	9.03
Found	69.62	5.44	9.00

On the basis of chemical properties, elemental and spectral analysis of the compound 2d was assigned the structure as 1-H-3-methyl-4(4-methoxyphenyl)-5-(4-methoxyphenyl)-2-imidazolone.



(2d)

Similarly other variedly substituted 2-imidazolones (Expt. No. 138 to 139) were prepared by following the same procedure and are given in table 2.2.

Results and Discussion

We newly synthesized variedly substituted -2-imidzolones by the condensation of each of three substituted benzoins with urea, methyl urea and phenyl urea respectively.

Table 2.2: List of synthesized compounds, their % yield and melting points.

Sr. No.	Compound	Percent Yield (%)	Melting Point (°C)
1	1-H-3-methyl-4-(4-methoxyphenyl)-5-(4-methoxyphenyl)-2-imidazolone. (2d)	62	180
2	1, 3,dihydro-4-(4-methoxyphenyl)-5-(4-methoxyphenyl)-2-imidazolone. (2e)	66	145
3	1-H-3-phenyl-4-(4-methoxyphenyl)-5-(4-methoxyphenyl)-2-imidazolone. (2f)	58	138

Antimicrobial Activity Method for the determination of antimicrobial activity

The newly synthesized compounds were screened for their antimicrobial activity against the test organisms *E.coli, S.typhi, P.vulgaris, B.subtilis, S.aurus* and *S.penumoniae* by using agar disc diffusion method at concentration of 100 µgm/ml in DMF as a solvent. Each standardized test organism (0.1ml) was spread on the solidified sterile agar plates.

Conclusion

Thus we could succeed in synthesizing variedly substituted-2-imidazolone with simple and easy to workout methodology. Use of Zeolite as a catalyst enabled us rapid route for the synthesis of 2-imidazolones which could reduce reflux time to as low as two and half hours. The catalyst is insoluble in solvent due to which isolation of the product became much easy. The synthesized compounds were screened for antimicrobial activity against the test organisms E.coli, S.typhi, P.vulgaris, B.subtilis, S.aurus and S.penumoniae

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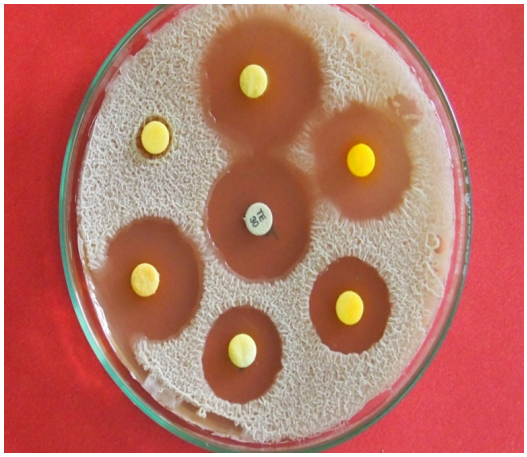

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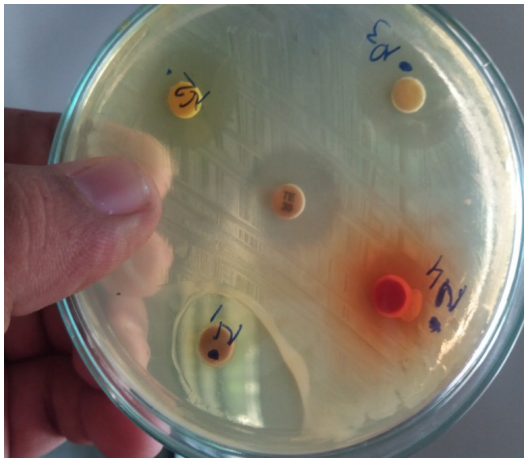

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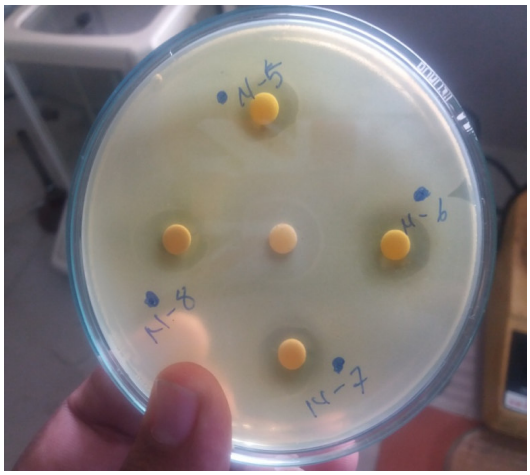

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