In Fig. 1, the change in frequencies for various mass loads is shown.

\[ \frac{\beta_1}{\beta_2} \cot \beta_1 x_1 + \cot \beta_2 (x_3 - x_1) = 0, \]

i.e.,

\[ \frac{\beta_1}{\beta_2} \sin \beta_1 (x_3 - x_1) \cos \beta_1 x_1 + \cos \beta_2 (x_3 - x_1) \sin \beta_2 x_2 = 0, \]

where \( x_1 = l_1 \) and \( x_3 = l_1 + l_4 \).

This tallies with De's previous investigation.

The author is grateful to Prof. P. K. Ghosh, Head of the Department of Applied Mathematics, Professor S. N. Ghosh of Applied Physics, Calcutta University and Dr. M. Ghosh, Director of the Institute of Theoretical Physics, for their keen interest and constant encouragement.

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A CONVENIENT METHOD FOR THE SYNTHESIS OF 2-[(2-BENZIMIDAZOLYL) AMINO]-6-METHYL-4-PYRIMIDINOLS

2-[(2-Benzimidazolyl) amino]-6-methyl-4-pyrimidinols (I) are valuable precursors for the preparation of various potent antimalarial drugs such as 2-[(4) (diaryl)alkyl] amino]-6-methyl-2-pyridinylamino]-benzimidazoles \( \text{III} \) (II)

Recently Werbel and coworkers \( ^1 \) prepared 1 in 11-51\% yields by the condensation of suitably substituted \( \alpha \)-phenylenediamines with 2 (cyanoamino-4-hydroxy-6-methylpyrimidine (III) which in turn is obtained (in less than 50\% yield) by the condensation of di cyan maleic acid with ethyl acetoacetate in the presence of sodium ethoxide.
Although reported method does not involve a number of steps, it is not satisfactory as: (i) yields are very poor, (ii) time required particularly for the condensation of (III) with ethyl acetocetate is too long (>24 hrs.). To overcome the above mentioned difficulties, we wish to report in this paper a new and simple method for the synthesis of title compounds (I).

In the present method, different 2-guanidinobenzimidazoles9-8 (IV), obtained in better yields (50-70%) than that of III, were condensed with ethyl acetocetate and afforded the required pyrimidinols in excellent yields (71-85%). Time required for the above condensation is only 50-60 minutes and the use of sodium ethoxide has also been avoided.

To study the general applicability of the method developed six new compounds (1-5, Table I and one in experimental part) have also been synthesised. All the known compounds thus obtained were found to be identical (m.p.s and elemental analysis) with their authentic samples. Other compounds (new) were also characterized by their high m.p.s, elemental analysis and converting one of them (I R = X = Z = H) into its 4-chloro and then 4-piperidino derivatives which were confirmed by their analytical data.

Experimental

All the melting points are uncorrected. 2-Guanidinobenzimidazoles were prepared in the laboratory by the action of different o-phenylenediamines on dicyandiamide according to the conditions given in literatures9-8 and characterized by their m.p.s.

2-[(2-Benzimidazolyl) amino]-6-methyl-4-pyrimidinol (I, R = X = Z = H)

A mixture of 2-guanidinobenzimidazole (17.50 g, 0.1 mole) and ethyl acetocetate (15.60 g, 0.12 mole) in distilled xylene (100 ml) was heated under reflux for 50-60 min. A white solid began to separate during the refluxing time. When the refluxing was over, 70-80 ml of xylene was removed by distillation and after cooling the solid was collected by filtration, washed with a little ether and dried. Recrystallization from dimethyl sulphoxide followed by a wash with little methanol gave 20.48 g (85%) 1(R = X = Z = H) m.p. >360°C (Found: C, 59.89; H, 4.41 N, 28.87%. Calculated for C12H11N2O: C, 59.75; H, 4.57; N, 29.04%).

Following the above procedure other pyrimidinols were also prepared. Their data are given in Table I.

2-[(2-Benzimidazolyl) amino]-4-chloro-6-methylpyrimidine

Following the procedure of Werbel et al. the above 4-chloro derivative was obtained from I (R = X = Z = H) and phosphorous oxychloride in 75%: m.p. 350-42°C (decomp.) (Found: C, 55.23; H, 3.82; N, 27.18%. Calcd. for C12H10N2Cl: C, 55.49; H, 3.85; N, 26.97%).

2-(4-Piperidino-6-methyl-2-pyrimidinylamino) benzimidazole

A mixture of 2-[(2-benzimidazolyl) amino]-4-chloro-6-methyl-pyrimidine (5.19 g, 0.02 mole) and purified piperidine (6.80 g, 0.08 mole) was heated under reflux for 2 hrs. The cooled mixture

Table I

<table>
<thead>
<tr>
<th>Sl. No.</th>
<th>X, Z</th>
<th>M.P. °C</th>
<th>Yield %</th>
<th>Solvent for crystallization</th>
<th>Formula</th>
<th>Analysis</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>C</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>C found</td>
</tr>
<tr>
<td>1. 5- (or 6) Br</td>
<td>&gt;360</td>
<td>75</td>
<td>DMSO</td>
<td>C12H16BrN3O</td>
<td>45.00</td>
<td>3.13</td>
</tr>
<tr>
<td>2. 5- (or 6) Me</td>
<td>&gt;332-36 †</td>
<td>71</td>
<td>DMSO-EtOH</td>
<td>C13H15N3O</td>
<td>61.18</td>
<td>5.09</td>
</tr>
<tr>
<td>3. 5, 6- (Me)2</td>
<td>&gt;33 (d)</td>
<td>78</td>
<td>DMSO-EtOH</td>
<td>C14H16N4O</td>
<td>61.32</td>
<td>5.23</td>
</tr>
<tr>
<td>4. 5- (or 6) OMe</td>
<td>347-49 †</td>
<td>82</td>
<td>DMSO</td>
<td>C15H18N4O2</td>
<td>62.47</td>
<td>5.57</td>
</tr>
<tr>
<td>5. 5- (or 6) NO2</td>
<td>&gt;360</td>
<td>81</td>
<td>DMSO</td>
<td>C12H19N4O3</td>
<td>57.56</td>
<td>4.79</td>
</tr>
</tbody>
</table>

* The values of R, X, Z and yields are given for reported compounds prepared [H, 5-(or 6) Cl, 75%; H, 5-6-Cl, 74%; H, 4-NO2, 83%].

† — decomposition.
was dissolved in dilute hydrochloric acid; treated with charcoal and filtered. The filtrate was made basic with 30% aqueous sodium hydroxide solution in cold (10–15°C), and solid collected by filtration was crystallized from ethanol: yield 3.1 g (≈50%), mp 210–12°C (Found: C, 66.58; H, 6.41; N, 27.61%; Calcd. for C₁₇H₂₇N₆: C, 66.23; H, 6.50; N, 27.27%).

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**POLYMERISATION OF SESAME OIL**

**UNSaturated** oils can undergo polymerisation by auto-oxidation, heating in air or oxygen, in presence of catalyst like clay, activated earths and metal ions. The polymerisation of a number of oils, synthetic fatty acid esters, and glycerides have been studied. In the present investigation, the polymerisation of sesame oil in presence of red lead (Pb₃O₄) has been studied.

Sesame oil was polymerised by suspending red lead (Pb₃O₄) and spreading the mixture in a glass dish. It was allowed to stand at room temperature for 5 weeks. The clear pale yellow oil becomes viscous and orange red in colour. This was decanted and centrifuged to remove suspended red lead.

The iodine number, saponification value, peroxide value and refractive index were determined for the oil before and after reaction according to the AOAC procedures. The intrinsic viscosity was determined in acetone using an Ubbelohde viscometer. Gel permeation chromatography was carried out in a 60 × 1 cm column packed with sephadex LH 20 in ethanol. The eluted products were estimated by dichromate oxidation. The triglycerides of sesame oil before and after reaction were separated by TLC, transmethylated with methanolic HCl and the methyl esters of the fatty acids characterised by GLC in a varian aerograph series 1400 using a 8' × ½” stainless steel column packed with 15% DEGS on chromosorb Q. The individual triglyceride species before and after reaction were separated by argentation TLC.

Sesame oil before reaction has an iodine number of 116, saponification value 210, refractive index at 25°C 1.467 and intrinsic viscosity 0.053 dl/gm. After the reaction the iodine value decreased to 47.5, saponification value increased to 233, refractive index to 1.473 and intrinsic viscosity to 0.058 dl/gm. The peroxide value of the oil after the reaction was 18 mE/Kg whereas that of the oil stored under the same conditions was 67 mE/Kg.

From the gel permeation chromatography, the percentage composition of the oil before reaction was glycerides 80%, free fatty acids 15% and after the reaction the glycerides were 50%, free fatty acids 15% and a new compound whose molecular weight was estimated as 1600 was found to an extent of 30%.

The fatty acid composition of the triglycerides before the reaction on percentage basis is: C₁₆:₄ 14%, C₁₈:₀ 5%, C₁₈:₁ 39%, C₁₈:₂ 41% and the fatty acid composition of triglycerides remaining after reaction was: C₁₆:₀ 15%, C₁₈:₀ 7.5%, C₁₈:₁ 52% and C₁₈:₂ 20%.

Argentation TLC separates triglycerides depending on the number of double bonds present in the molecule. For sesame oil triglycerides 9 spots fluorescent in UV when sprayed with 2, 6 dichlororfluorescin were obtained in a solvent system chloroform : methanol (99 : 1 V/V) whereas for the triglycerides remaining after the reaction only 6 spots were obtained.

Hence it is concluded that the highly unsaturated triglycerides of the sesame oil have undergone polymerisation.

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