

## FOREWORD

The work embodied in the present thesis was undertaken by the candidate early in 1968 under the guidance of Dr. S. N. Poddar. When the work in the present programme was started, there were only a few reports in literature regarding the ligational behaviour of pyrazole or its substituted products, although voluminous work on imidazole, the m-isomer of pyrazole, was reported. During last two years, a number of publications have appeared dealing mainly on the complex-forming capabilities of unsubstituted pyrazole, its mono- and dimethyl derivatives as well as on some other pyrazole derivatives.

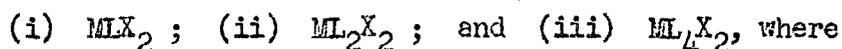
In the "Introduction" of the present thesis, the work on the metallic complexes of imidazole, its substituted products and derivatives has been presented, considering their analogy with the pyrazoles and their derivatives. This chapter also gives a review of the work on metallic complexes of pyrazole, substituted pyrazoles and derivatives of pyrazole. The review is not claimed to be exhaustive but every effort has been made to include therein all the important work in this field reported so far.

The work reported in the present thesis consists in the study of some transition-metal complexes of 3,4,5-trimethyl pyrazole. The ligand was synthesized by condensing 3-methylacetylacetone with hydrazine hydrate and it forms stable, well-defined colourless crystalline

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solid with definite melting point. For preparing the metallic complexes, three common transitional metal ions, namely copper (II), nickel (II) and cobalt (II) were chosen. The ligand, 3,4,5-trimethyl pyrazole generally behaves as a monodentate coordinating ligand, although removal of imino-hydrogen atom takes place if the reactions with the metal ions are carried out in alkaline medium. The resulting metal-chelate is obviously polymeric in nature and forms amorphous powder, insoluble in all common solvents and having comparatively higher thermal stability than the other metal-complexes of the ligand studied during the present investigation.

The coordination complexes 3,4,5-trimethyl pyrazole with copper (II), nickel (II) and cobalt (II) ions, which have been isolated during the present study are of three types depending upon the number of the ligand molecules associated per metal ion. These can be formulated as



M is an ion of the metals stated above, L is a molecule of 3,4,5-trimethyl pyrazole and X is a monovalent anion or  $\frac{1}{2}$  of a divalent anion. The mono-complex could be isolated only in the case of copper (II) ion while the bis- and the tetrakis- complexes were isolated with all the three metallic ions. The compound containing 3,4,5-trimethyl pyrazolate ion could be

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prepared only with cobalt (II) ion. The compositions of the complexes were determined by elemental analyses. Various physico-chemical methods, viz., the study of magnetic susceptibility, conductivity, visible and infrared spectra were used to elucidate the probable structure of the complexes.