**Experimental Section**

All melting points reported were obtained from a Mel-Temp

capillary melting point apparatus and were corrected. The

microanalysis were performed by Midwest Microlab, Inc.,

Indianapolis, Ind. Infrared spectra were recorded with a Perkin-

Elmer Model **137** or **137A** Infracord. The ultraviolet spectra

were recorded using a Bausch and Lomb Model **505** spectrophotometer.

The nmr spectra were recorded on a Varian Model

A-60 spectrometer, employing tetramethylsilane as an internal

reference. All nmr spectra are in agreement with the =signed

structures. The molecular weights were determined in specified

solvents on a Mechrolab vapor pressure osmometer, Model 301A.

**Preparation of Ylidenemalononitri1es.-The** method described

by Mowrylz was used for the preparation of all the ylidenemalononitriles

except **9c.** Using the suggestion offered by Cope and

co-workers,18 an additional amount of catalyst was used for the

condensation of malononitrile with hindered ketones. The general

procedure for the condensation of nonhindered carbonyl

compounds with malononitrile used in these experiments is

described as follows. For every **0.5** mole of carbonyl compound,

**0.6** mole of malononitrile, **4** g of anhydrous ammonium acetate,

and **12** ml of glacial acetic acid were used. Anhydrous benzene

was added and the benzene solution was refluxed until the amount

of water collected in the Dean-Stark trap remained constant

(usually **4-12** hr) . For sterically hindered carbonyl compounds,

the reflux time was longer and two to eight times the recommended

amount of catalyst was added. After refluxing, the

benzene solution was washed with water, bicarbonate solution,

and water again, and dried (NazSO4). Evaporation of the solvent

yielded either a crystalline dinitrile or an oil. The latter compounds

were distilled at reduced pressure (in some cases unreacted

carbonyl compounds may be recovered). The properties of the

ylidenemalononitriles prepared are summarized in Table I.

**General Preparation of Lactones** Using **PPA** .-The liquid

ylidenemalononitriles were mixed thoroughly, using a mechanical

stirrer, with ten times their weight of polyphosphoric acid at

room temperature. When mixing was complete, the mixture

was heated to **100'** for 12 hr with continuous stirring. The

mixtures were hydrolyzed in a tenfold weight of water. The

solid ylidenemalononitriles were treated in a similar manner

except that no mixing was required before heating. The mixtures

were hydrolyzed as mentioned above. The hydrolyzed

mixtures were saturated with NaCl and extracted with CHCla

if water-soluble products were obtained. The CHCls solutions

were washed with small volumes of water and dried (NazSO4).