

Quarterly Report: August 1, 1992 - October 31, 1992

"Catalysis and Co-Catalysis of Bond Cleavages in Coal and Coal Analogs"

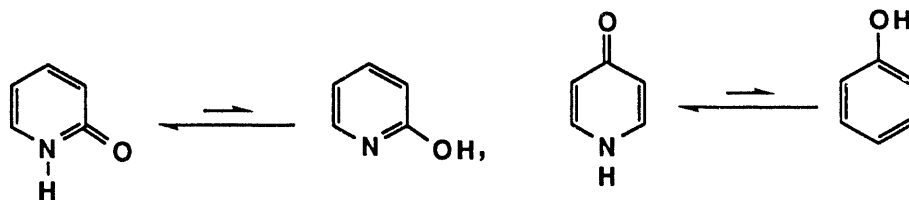
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PRINCIPAL INVESTIGATOR:

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RESEARCH PROGRESS:

Our last report demonstrated that 2-aminophenol and 4-aminophenol were exceptionally effective catalysts for transfer hydrogenolyses of dinaphthyl ether and dinaphthyl sulfide. We have therefore investigated the catalytic efficiency of 2-pyridone and 4-pyridone, the tautomers of 2-hydroxy and 4-hydroxypyridine.



As can be seen in the Table (entries 1 and 2) neither compound appeared to be a particularly effective catalyst for hydrogenolysis of di-2-naphthyl ether (one) under the conditions usually employed (5 hrs at 450°C). However, a comparison of the effectiveness of 2-pyridone and 2-aminophenol at shorter reaction times showed that 2-pyridone was as effective as 2-aminophenol if the reaction time were reduced to one hour. At longer reaction times, the effectiveness of 2-pyridone dropped sharply, suggesting that the catalyst is destroyed under the reaction conditions.

We previously reported that reaction of compound I with bromine in carbon tetrachloride solution yielded primarily a rearrangement product, with the cyclization product, II, a minor component. We have now discovered that if the reaction is carried out in acetonitrile solution only the

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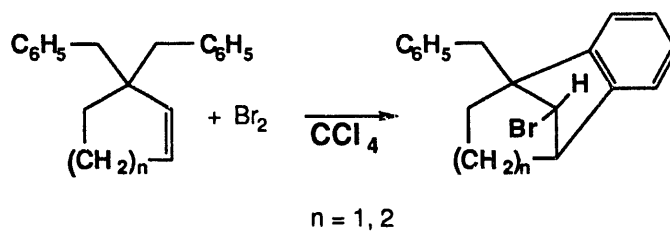
pure rearrangement product is obtained in essentially quantitative yield. The reason for this interesting solvent effect is not yet understood.

TABLE
TRANSFER HYDROGENOLYSIS OF DI-2-NAPHTHYL ETHER IN TETRALIN AT 450°C IN
STAINLESS STEEL REACTORS

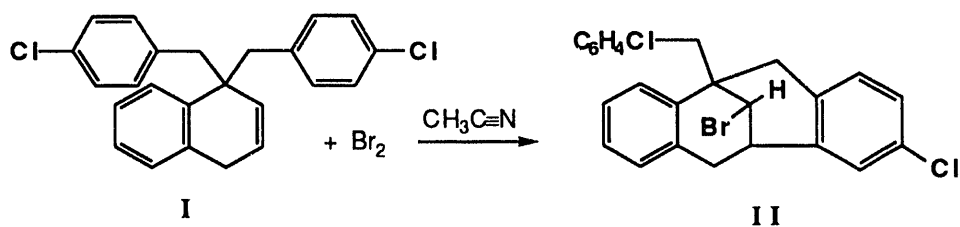
No. of Runs	Catalyst (mole/mole) ^a	Reaction Time (hrs)	% Disappearance of DNE	
1	2	2-Pyridone (14.6)	5.0	12.2 ± 1.1
2	2	4-Pyridone (13.9)	5.0	10.9 ± 1.6
3	1	2-Pyridone (14.6)	3.0	10.4
4	1	2-Aminophenol (13.6)	3.0	14.2
5	1	2-Pyridone (14.6)	1.1	7.2
6	1	2-Aminophenol (13.6)	1.1	6.1
7	1	2-Pyridone (14.2)	8.4	14.5
8	1	2-Aminophenol (13.6)	8.4	32.0
9	1	2-Pyridone (14.2)	22.5	19.1
10	1	2-Aminophenol (13.6)	22.5	40.5

a. Moles catalyst/moles DNE.

We have observed that the novel cyclization reaction proceeds with cyclohexene and cyclopentenes as well as with dihydronaphthalene derivatives:

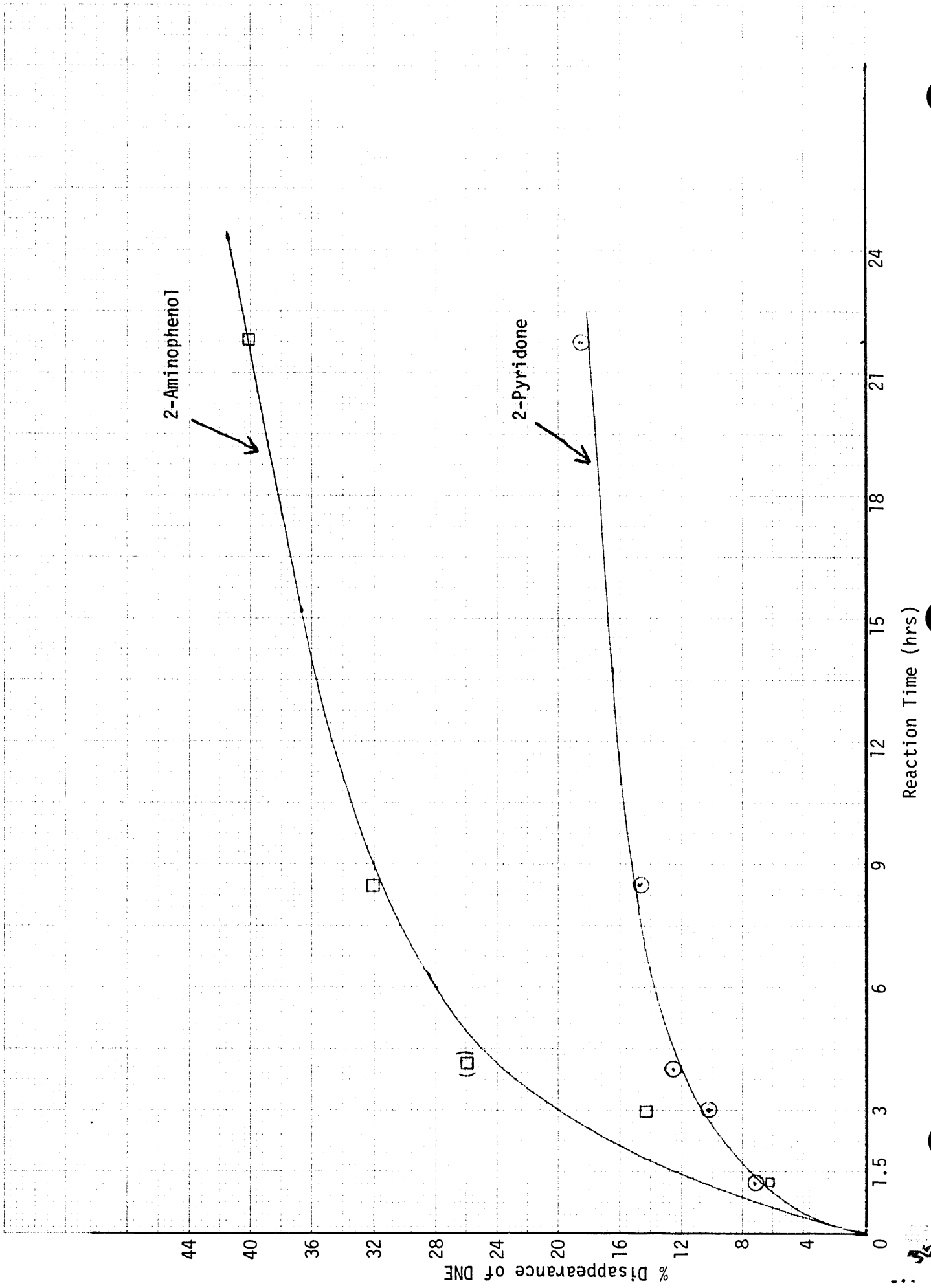


A manuscript describing this aspect of our work is in preparation.



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