

Kinetic Studies on the Cellulose-Propylene System in Presence of Ziegler-Natta Catalysts

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Synopsis

Kinetics of the uptake of polypropylene by the cellulose were studied. According to the experimental results, the reaction begins with an induction period which cannot be explained on the basis of the reaction mechanism of Ziegler-Natta catalysts. Most probable reason of this period may be the initial unsaturation of the system.

Studies on the effect of concentration of the catalyst (TiCl_4) showed that, above the order of magnitude of 10^{-2} mole/l, uptake of polypropylene by the cellulose is directly proportional to the concentration of TiCl_4 . The maximum catalytic activity was observed at the molar ratio of AlEt_3 to TiCl_4 equal to 1.5.

Concentration of propylene supported by the cellulose can be increased by elevation of the temperature.

By increasing the concentration or the specific surface area of the cellulose at a constant concentration of the cellulose at a constant concentration of the catalyst, a kind of saturation appears on the cellulose fibers with respect to the adsorption of the metal chloride. Thus, increasing the specific surface area does not involve an increase in the uptake of polypropylene, in fact, increasing the concentration of the cellulose results in a drop in the amount of propylene to be taken up.

On the basis of the infrared spectroscopical and the X-ray diffraction results, no differences were found between the mechanical mixture of the components and the cellulose fibers coated with polypropylene.

Mechanical strength of paper sheets was poor compared with that of sheets from pure cellulose. Cellulose encapsulated with polypropylene can be used in the paper industry as an interlayer in laminated papers.

INTRODUCTION

As the plastics industry has rapidly developed and the choice of its products has grown, pure cellulosic products have more and more been displaced from several fields (such as from packaging).

According to the interpretation of this backwardness in the cellulose industry by some specialists, industrial processing of cellulose follows a well known technology developed long ago and characteristics of the cellulose and its derivatives are fixed with scarcely any possibilities of changing them. During the last 10-15 years, however, it turned out that it is not so. Coupling the cellulose with plastic materials proved to be possible. "Plastics based on cellulose" retain the favorable properties of both systems.

Cellulose—plastics systems can be formed essentially in two ways:

- (a) Coupling the cellulose with semiprocessed or finished plastics.
- (b) Chemical modification of the cellulose.

Many papers in the literature deals with modifications of the cellulose. It is very difficult to review them and they include several contradictory data.

Reactions between cellulose and vinyl monomers, with a few exceptions, are of radical mechanism. Among these reactions a great many processes have been published for grafting initiated by high-energy irradiation,¹⁻³ photochemical methods,⁴ peroxides, hydroperoxides,^{5,6} azo and diazo compounds,⁷ redox systems^{8-12,13} and mechanochemical processes.^{14,15} Modification of cellulose by ionic mechanism is also known.¹⁶

Conclusions from papers on graft polymerizations of concern to the paper industry are not always unequivocal but a notion can be formed from the literary reviews viz., characteristics of papers can favorably be varied by grafting.¹⁹ Thus publications considering the regards of the paper industry are remarkable.

Papers published in 1964,^{17,18} showed that a third way is also possible for modification of cellulose: initiated polymerization of a vinyl monomer in presence of cellulose but without any chemical connection with it. Presumably surface of the elemental cellulose fibers is only encapsulated by the polymer formed.^{17,19} Normally, this process cannot be designated as grafting but it promises to be very interesting.^{20,21} Cellulose fibers containing 0.1 per cent of water are dispersed in hydrocarbon medium. This dispersion is treated with a Ziegler-Natta catalyst then ethylene is added to it. The catalyst is adsorbed on the surface of cellulose. After a rapid initiation, polymer spots are formed at a high rate and encapsulate the surface of the cellulose. On the surface of the individual fibers separate polymerization processes take place. No bonds or bridges exist between the individual fibers. Density of a wide range of fibers is near to that of water which facilitates the sheet formation. It is used in form of dispersion for production of paper. Nonencapsulated cellulose is also used as an additive since the capsulated fibers are nonpolar in nature. By using these additives, strong, wet fabric is formed. The pulp can be handled similarly to that from wood cellulose. Its humidity is adjusted to the usual value. The paper is produced by the conventional machines of the paper industry.

Only few papers have been published in this field and they cover principally the technology rather than the revelation of conditions of the reaction. Only two works are known^{19,22} to give a kinetic analysis of the polymerization reaction. The greatest merit of these papers is that it proves the polymerization to take place on the surface of the cellulose by adsorption of the catalyst (a metal halide) on the cellulose fibers. Adsorption can be extended by addition of a metal alkyl.

The present paper aims to study preparation of the coupled system of cellulose-propylene and to determine the principle factors and the optimum conditions of the polymerization reaction.

Technique of Polymerization

A bleached, sulphite type pine-cellulose made in USSR was used which contained 85 to 90 per cent of alpha-cellulose. The cellulose samples were ground in Jokro mill for various periods. Degree of grinding was determined by a Schopper-Riegler equipment. The wet, ground cellulose was filtered under vacuum, washed many times with methanol then dried up to constant weight in a vacuum oven at 60°C. Dried cellulose was stored in desiccators over phosphorus pentoxide.

Polymerization

Polymerization was carried out in a 1000-ml three-necked flask equipped with a reflux condenser, a high speed stirrer, and a thermometer. Four hundred milliliters of toluene was placed into the flask then 4 g of cellulose was added after weighing with an accuracy of 1 mg. To avoid losses from crumbling, cellulose was dampened by 50 ml of toluene and the residual cellulose was flushed into the reaction mixture by another 50 ml of toluene. Cooling, heating and stirring was started simultaneously at that time. After a ventilation with nitrogen for half an hour, TiCl_4 then in 10 min Et_3Al was added to the mixture by means of hypodermic syringes.

Propylene was introduced into the reactor at constant flow rate. Flow rate was measured by a differential manometer. Polymerization conditions were as follows:

Toluene	500 ml
Cellulose	2-5 g
TiCl_4	$1.26 \cdot 10^{-2}$ to $5.06 \cdot 10^{-2}$ mole/l
Al/Ti ratio	0.5 to 4 mole/mole
Flow rate of propylene	24 l/h
Temperature	0-50°C

The polymerization process was stopped by shutting off the propylene stream followed by addition of acidic methanol. The product was filtered and washed with distilled water then with methanol again. After the final filtration, the product was dried up to constant weight in a vacuum oven at 80°C.

Experimental Results

Polymerization of propylene on the cellulose fibers was studied in form of suspension in toluene medium at a concentration of 4 g cellulose per liter. The reaction time was varied within a wide range (15 min to 2.5 hr) in order to obtain maximum conversion. Uptake of polypropylene (in per cent of weight of cellulose) is shown in Figure 1 against the polymerization time.

The reaction shows a relatively long induction period. In this stage of the process, conversion of the monomer does not exceed 4 per cent. In the following rapid period, polymerization is essentially completed while the last period is slow again including a conversion of 1-2 per cent only. The

middle rapid period takes about 50 min. During this time the conversion rises to 45 per cent under conditions outlined above. The third (slow) stage of the kinetic curve may certainly be attributed to the aging of the complex catalyst (Fig. 1).

Using TiCl_4 in concentration of 10^{-3} mole/l in order of magnitude, polymerization does not take place. Above the concentration of 10^{-2} mole/l growth of weight of cellulose is directly proportional to the concentration of TiCl_4 (Fig. 2).

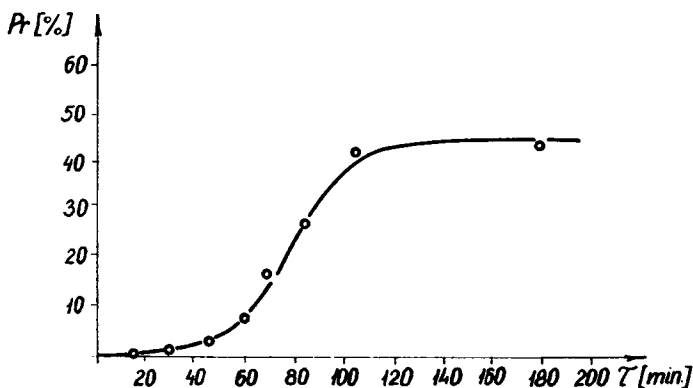


Fig. 1. Kinetics of the uptake of polypropylene by the cellulose. Degree of grinding: 43°SR , Al/Ti molar ratio: 2, concentration of TiCl_4 : $2.52 \cdot 10^{-2}$ mole/l, temperature: 30°C .

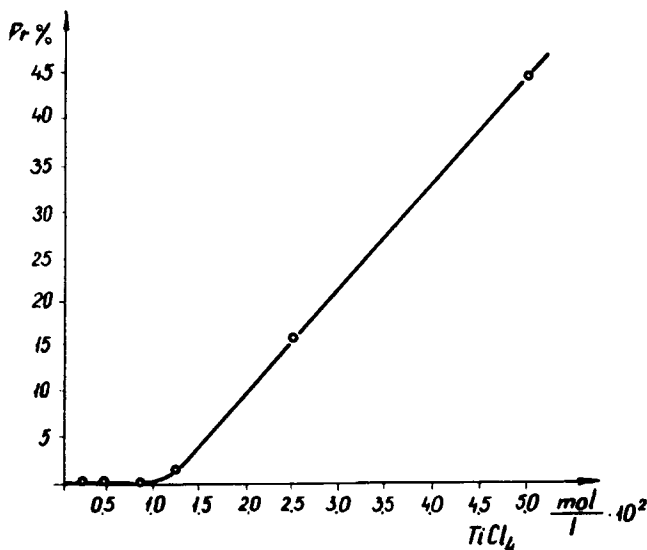


Fig. 2. Growth of weight of cellulose plotted against the concentration of TiCl_4 . Concentration of TiCl_4 : $0.3 \cdot 10^{-2}$ to $5 \cdot 10^{-2}$ mole/l Al/Ti molar ratio: 2, temperature: 30°C , time: 70 min, concentration of cellulose: 4 g/l, degree of grinding: 43°SR .

The polymerization rate of propylene is known to be directly proportional to the concentration of the metal halid. The order of magnitude of the concentration used is identical to that of catalysts for polymerization of propylene in absence of cellulose.²³

In the investigations of the effect of AlEt_3 to TiCl_4 molar ratio on the conversion, cellulose with 43 SR° of grinding was used. Molar ratio of the

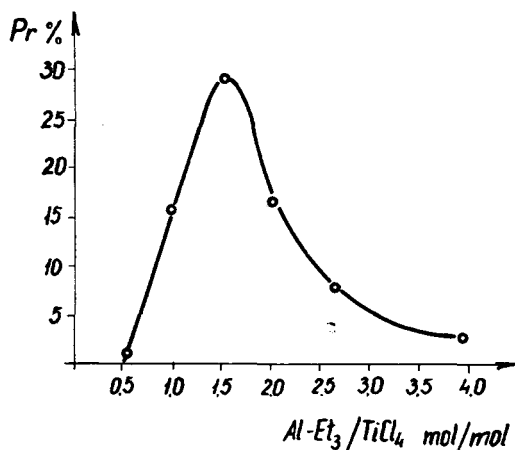


Fig. 3. Uptake of polypropylene plotted against the Al/Ti molar ratio. Concentration of cellulose: 4 g/l, degree of grinding: 43° SR, concentration of TiCl_4 : $2.52 \cdot 10^{-2}$ mole/l, time: 70 min, temperature: 30°C.

components of catalyst (Al to Ti) was varied in the range of 0.5 to 4. Concentration of cellulose was 4 g/l. Catalytic activity of the system is essentially influenced by the nature of the organometallic compound and its molar ratio to TiCl_4 . The molar ratio of catalyst components affects also the crystallinity and intrinsic viscosity of the polymer. For molar ratios of AlR_3 to TiCl_4 less than the unit or with TiCl_4 alone, polymerization of propylene leads to low molecular weight oils.²³ Increment per cent of weight of cellulose is plotted against temperature in Figure 4.

Considerable conclusions cannot be drawn from these few experimental data. It is apparent, however, that the amount of polypropylene on the cellulose increases with the temperature in the range studied. It should be considered in the analysis of the curve that concentration of propylene increases as temperature decreases (adsorption) and the polymerization rate is directly proportional to the partial tension of propylene. On the other hand, the reaction between the components of the catalyst becomes quicker with increasing temperature and structural changes of the complex take place as the degree of reduction of TiCl_4 varies. It can actually be stated that with increasing temperatures the number of active sites degree of reduction responsible for the polymerization is considerably augmented resulting in the increasing amount of polypropylene taken up by the cellulose (Fig. 4).

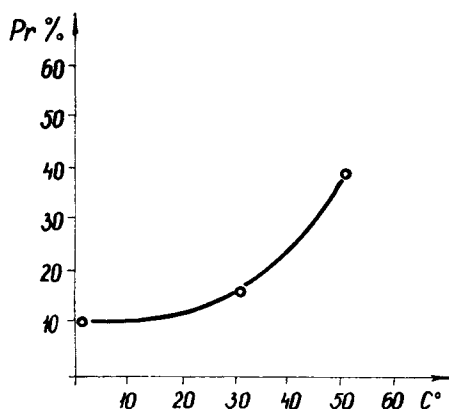


Fig. 4. Effect of the temperature. Concentration of cellulose 4 g/l, degree of grinding: 43° SR, time: 70 min, concentration of TiCl_4 : $2.52 \cdot 10^{-2}$ mole/l, Al/Ti molar ratio: 2.

Grinding of cellulose performs two parts: defibration into single fibers and increasing the surface area of cellulose fibers. Slight increase in the surface area can be reached by transversal cutting of cellulose fibers while longitudinal cutting results in high surface area. During the grinding several cutting effects may appear. The most important ones: chopping, shearing and crushing effects.

Chopping effect alone leads neither to detachment of the fibers, nor to considerable increase in the surface area, in fact, the external layer covering the fibers does not get loose as well. Crushing effect can cause the fibers to be decomposed or fibrillized thus the external layer of the cellulose fibers gets damaged. This effect does not result in considerable shortening of the fibers, only a longitudinal splitting takes place on the surface. Shearing effect does not cause cutting of the fibers but fibrillation and longitudinal splitting of the fiber wall can be observed.

In the Jokro mill used for the experiments, chopping and crushing effects predominate. The course of grinding and characteristics of the ground matter were determined by the method of Schopper and Riegler for measuring the degree of grinding. Degrees of grinding were selected according to the paper industry. In the paper industry, a sample of cellulose is characterized by comparison with samples of different degrees of grinding. The first reference point represents an unground cellulose giving a notion of frictional forces in the paper. The second point corresponds to 25–30° SR just below the formation of lateral bonds among the fibers. The third point at 45 to 50 SR° characterizes an advanced stage of formation of these bonds. The fourth point refers to the optimum strength (65–70° SR) where the elemental fibers have reached their strength.

Grinding times and the corresponding degrees of grinding are shown in Table I for the cellulose under investigation. Effectivity of grinding was studied in order to establish the influence of increasing surface area on uptake of polypropylene by the cellulose.

TABLE I
Degree of Grinding as a Function of the Grinding Time

Grinding time min	Degree of grinding SR
0	16
5	22
15	43
25	65

Evaluation of the data is difficult because of scattering of the experimental points. In the interpretation, however, it should be considered that the experimental error is about 1-2% (losses due to pulverization, weighing etc.). It can be established that the amount of polypropylene taken up by the cellulose is practically independent on the degree of grinding (Fig. 5).

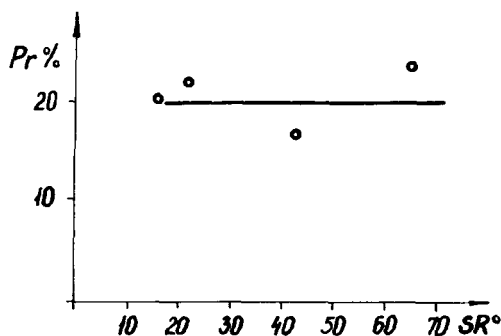


Fig. 5. Uptake of polypropylene plotted against the degree of grinding. Polymerization conditions: of Figure 1.

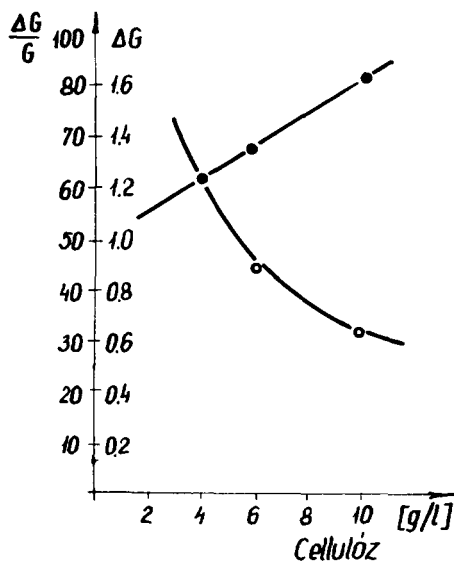


Fig. 6. Effect of concentration of the cellulose. Polymerization conditions: of Figure 1.

● ΔG , ○ $\Delta G/G$

Concentration of cellulose in the solvent was varied in the range of 4 to 10 g/l. Uptake of polypropylene in grams (ΔG) and in per cent of weight of cellulose ($\Delta G/G \cdot 100$) is shown in Figure 6. Increasing the ratio of cellulose to solvent, i.e., the concentration of cellulose, polymerization does not stop proved by the slow but consistent increase in the amount of polypropylene. But it does not carry satisfactory conviction since at a constant concentration of catalyst the amount of polymer taken up can be increased by increasing the concentration of cellulose. The amount of polypropylene per 1 g of cellulose is, however, a much more significant factor which tends to decrease (Fig. 6). Paper sheets were prepared from cellulose samples of various degrees of grinding and various polypropylene content on the basis of the previous experimental results. Characteristics of the samples used for preparation of paper sheets are collected in Table II.

TABLE II
Polypropylene Content of Cellulose Samples Having Various Degrees of Grinding

Degree of grinding °SR	Polypropylene content %
22	3,4
22	27,0
43	4,7
43	11,4
43	54,0
65	10,3
65	30,7

Formation of the sheet was preceded by agitating the material with water in a disintegrator. After adjusting the proper density, the sheets were formed in a Rapid-Köthen equipment. The wet sheets were dried by means of hot water heating and vacuum at 95–97°C for 8 min. Mechanical strength of the sheets produced very low in this stage and it cannot be measured by the testing equipments of the paper industry. The sheets of low mechanical strength therefore were pressed in order to melt the polypropylene. According to some preliminary experiments, the most suitable press conditions were as follows:

Pressure	250 kp/cm ²
Temperature	170°C
Preheating period	0.5 min
Press time	4 min

Unfortunately, the equipment did not permit a heating above 170°C. Surface of sheets produced are glossy and smooth to the touch. Slight discoloration and inhomogeneity appears especially for sheets of higher polypropylene content. Tensile and tear strengths of the paper sheets produced were determined at 20°C and at 65% relative humidity. The former was measured by a System Schopper tensile apparatus and the latter

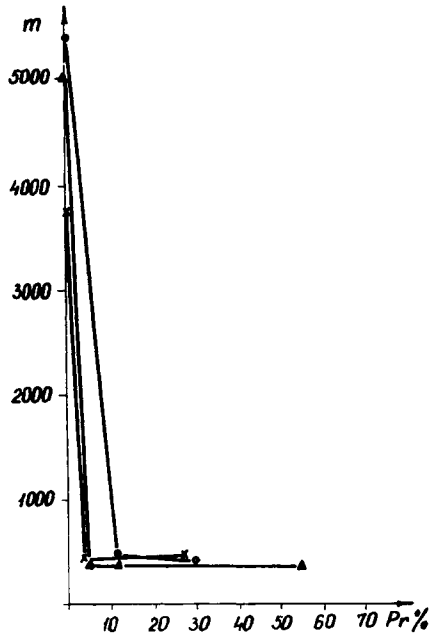


Fig. 7. Tensile strength:
 ○ Degree of grinding of cellulose: 22° SR
 ▲ " " " 43° SR
 × " " " 65° SR

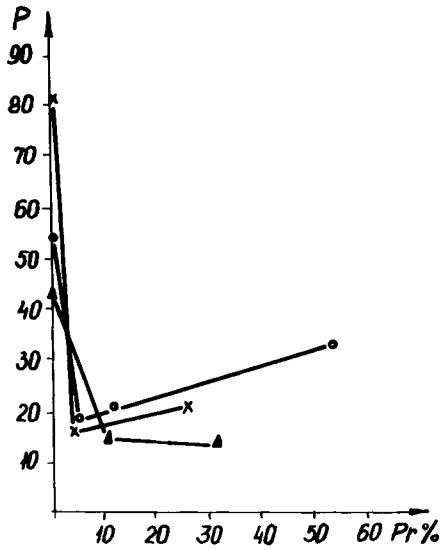


Fig. 8. Tear strength:
 × Degree of grinding of cellulose: 22° SR
 ○ " " " 43° SR
 ▲ " " " 65° SR

by an Elmendorf equipment. The results are shown in Figures 7 and 8. Tensile and tear strength of the paper sheets is markedly decreased by uptake of polypropylene independently on the degree of grinding of the cellulose, 3 to 10 per cent of propylene is satisfactory for a considerable deterioration of the tensile strength. Above this propylene content, tensile strength is independent on the amount of propylene taken up by the cellulose fibers (Fig. 7). Similar characteristics were found for tear strength with the exception that increasing the uptake of polypropylene to a high level, tear strength of a paper sheet of pure cellulose fibers may be restored (Fig. 8).

DISCUSSION

The relatively long induction period cannot be explained by the reaction mechanism of Ziegler-Natta catalysts. Assuming an ionic coordination mechanism, the reaction rate increases gradually as the reduction of Ti^{4+} has commenced. Degree of reduction depends on time and temperature but under these conditions the slow polymerization in the first stage is not reasonable. This contradiction can be attributed to two factors: (Fig. 1).

(1) Interactions between the catalyst system and the cellulose i.e. modification of the former in presence of cellulose.

(2) Initial insufficiency of the concentration of propylene in the system.

The first assumption is supported by the fact that cellulose can act as an electron donor in some systems.²² Several informations are available about polymerization of propylene in presence of electron donors. They can briefly be summarized as follows:

Electron donors (such as amines, ethers etc.) inhibit or promote the polymerization reaction mostly by forming complexes with the catalyst modifying the electron structure of the two-component catalyst system. The extent of the effect of the complex is not completely unequivocal. Reactivity of an electron donor depends on its molecular structure. Steric hindrances, basicity, concentration etc. should be considered in any case. Naturally, electron donor characteristics of cellulose may appear, in fact, this may be proved on the basis of the experimental results (induction period).

To verify the second assumption, the time dependence of the adsorption of propylene was investigated in the polymerization system in absence and in presence of catalyst (Fig. 9). According to the diagram, the curves divide only after a certain period, at a given concentration of propylene. Deviation of the curves from one another is caused by the polymerization of propylene.

Comparing Figures 9 and 1 it is evident, that polymerization reaches a considerable rate only when the system is near the saturation of propylene. Saturation is achieved in 25 to 30 min which is identical to the induction period. The only problem which of the both possible processes is the primary one.

Change in the polymerization rate during the final stage can be attributed to the changes in activity, i.e., in composition of the complex catalyst.

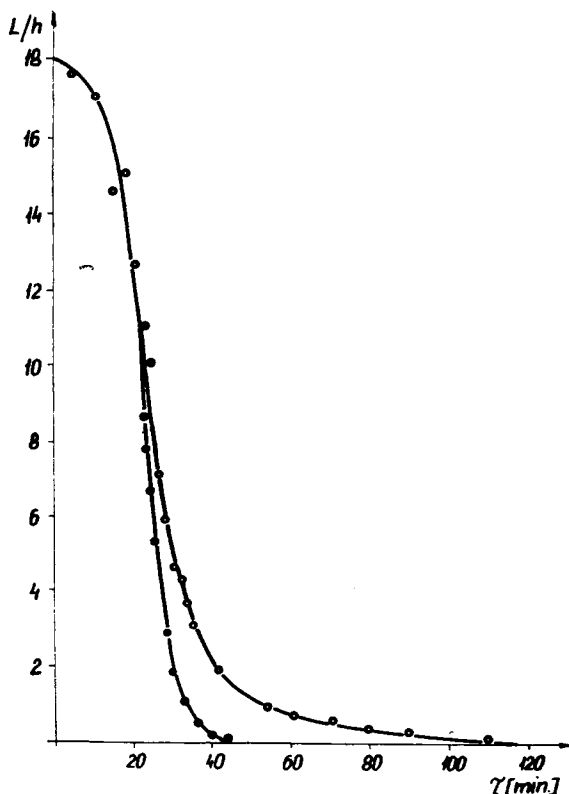


Fig. 9. Changes in the consumption rate of propylene. ● Adsorption; ○ Adsorption + polymerization.

Characteristic feature in syntheses of high molecular weight compounds from alpha-olefins is the intricate change in catalytic activity of the system as a function of time. Polymerization of propylene is a pure heterogeneous process taking place on the surface of the titanium halide containing trialkyl aluminium adsorbed. The principle action of the polymerization is, presumably the introduction of the monomer between the metal atom and the alkyl group. Chemical activity of the catalyst may be attributed to both Al-R and Ti-R bonds.²³

Accepting the initiation mechanism by Ziegler-Natta catalysts, it is evident that increasing the concentration of $TiCl_4$, probability of formation of Ti-R complex bond increases as well and so does the polymerization rate (Fig. 2). Decreasing the concentration or the active surface area of the initiator, the sites available for the monomer also grow less. Concentration of the monomer is relatively high in this case thus the polymerization rate is low. Concentration of catalyst is also important with respect to the molecular weight of the product. Number of the active sites affects reversely the molecular weight of the polymer, proper selection of the concentration of catalyst may therefore be very important.

It was shown for the polymerization of propylene²³ that the maximum activity of the catalyst is reached at the molar ratio of AlEt_3 to TiCl_4 equal to 2. The lower molar ratio observed in this study may probably be attributed to the interaction between the components of the catalyst and the cellulose (Fig. 3). Considerable amount of TiCl_4 adsorbed on the cellulose may, presumably, interact with the cellulose. Cellulose as an electron donor can modify the electron orbits of components of the catalyst altering their activity. Assuming that reduction rate of titanium tetrachloride adsorbed by the cellulose is higher than that of the titanium tetrachloride remained in the solution, polymerization reaction takes place principally on the surface of cellulose. AlEt_3 certainly provides the reduction of TiCl_4 also in this special case but reduction of Ti^{4+} adsorbed on the cellulose may be more rapid due to the electron donor effect mentioned. Thus the reaction occurs mainly on the surface. Naturally a part of TiCl_4 remains in the solution but its catalytic activity is negligible in comparison with the former because of its low concentration. In all probability, the molar ratio of Al to Ti equal to 1.5 obtained in these investigations includes also the TiCl_4 remaining in the solution. This relation discussed is important also in solution of the consistent problem: what is the reason of the induction period (Fig. 1). Provided that cellulose as an electron donor accelerates the reduction of Ti^{4+} , the overall polymerization rate should be increased as well. In this case induction period cannot be considered thus its reason can only be the initial unsaturation of the polymerization system for propylene. As it was demonstrated, uptake of propylene by the cellulose is practically independent on the degree of grinding of the latter (Fig. 5). It is evident from the experiments that increasing the surface area of cellulose should involve the increasing concentration of the catalyst (TiCl_4). Expansion of the surface area is in vain when proper amount of catalyst to be adsorbed is not available. Uptake of polypropylene by cellulose is constant presumably because the extent of adsorption cannot increase at a given concentration of catalyst. Analogous phenomena were found by varying the concentration of the cellulose (Fig. 6). In the discussion of the results, the following factors should be considered:

(1) Besides the cellulose to solvent ratio, concentration of catalyst referring to the cellulose varied as well.

(2) If the polymerization of propylene were not disturbed and influenced by the cellulose, the absolute growing of weight of cellulose (ΔG) had to be constant, i.e., independent on the concentration of cellulose.

(3) It was established that under these conditions, uptake of polypropylene by the cellulose is independent on the specific surface area of the latter.

The first and third factors are correct being proved by unequivocal conclusions or experimental results. The second assumption could, however, not be evidenced by the experimental results. Concentration of cellulose involved a change in the equilibrium of the polymerization system. It may well be supposed therefore, that the present catalyst system can interact

with the molecules of the cellulose. At the same time it was proved again that the polymerization takes place on the surface of the cellulose. The relation between the amount of cellulose coated with polypropylene and the concentration of catalyst cannot be interpreted otherwise. At a constant concentration of catalyst increasing the amount of cellulose extends the surface available for adsorption. Cellulose fibers are possibly not saturated with respect to the adsorption of Ti^{4+} , specific concentration of titanium and simultaneously the amount of polypropylene decreases on them. However, provement of these conceptions and the experimental results leave the question open about the electron donor effect of the cellulose. Even if Ti^{4+} interacted with the cellulose and the latter deformed the electron structure of the complex catalyst, it would not be sure that the catalytic activity would change.

Complex formation usually involves similar problems since formation of any new bonds (complex, valence etc. ones) is reflected in the nature of the initiation. Presence of cellulose does not result in a retardant effect on the polymerization of propylene, in fact, polymerization is detected to take place on the surface of cellulose. This phenomenon can be explained as follows:

- (1) Electron donor character of the cellulose is weak.
- (2) The electron donor effect depends on the concentration and the structure.

Micromorphological characteristics of the products were studied by infrared spectroscopy and X-ray diffraction. Infrared spectra were recorded by a Zeiss UR-LO spectrophotometer in the range of 2.5 to 25 μm

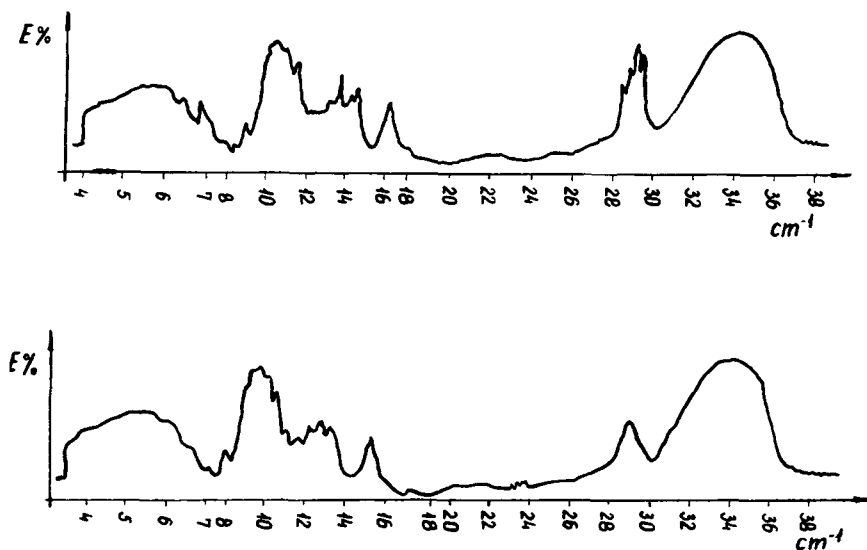


Fig. 10. Infrared spectra of the cellulose and the cellulose—propylene system. Above: cellulose—propylene system (45:55 w/w ratio); Below: cellulose.

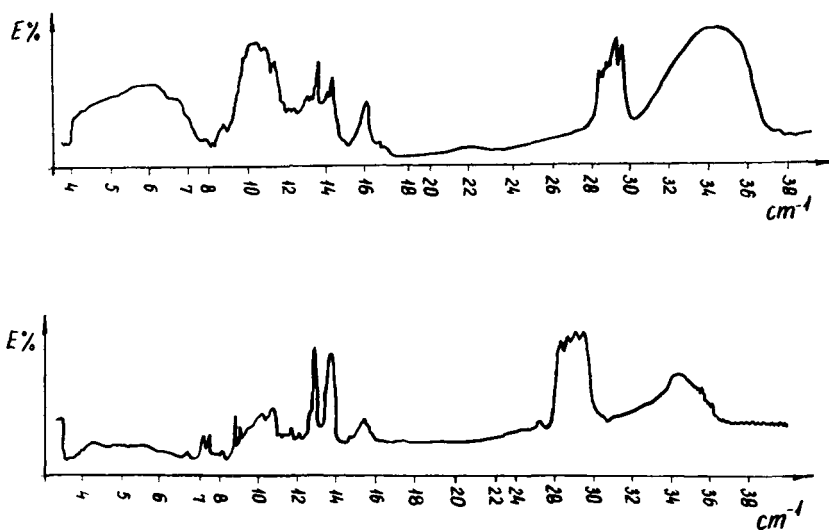


Fig. 11. Infrared spectra of polypropylene and of a mechanical mixture of cellulose—polypropylene. Above: Mechanical mixture of cellulose—polypropylene (45:55 w/w ratio); Below: Polypropylene.

wave length, using the tablet technique. Infrared spectra of cellulose, polypropylene, their mechanical mixture and their encapsulated composition are shown in Figures 10 and 11. Infrared spectrum of the cellulose encapsulated with polypropylene shows the bands of both polypropylene and cellulose to be present. Bands of polypropylene are not so sharp as those of the homopolymer since the broad, indistinct bands of the cellulose surpass them. New bands and shifts of wave length are not found in the spectrogram. In comparison with the spectrum of the mechanical mixture, no differences can be observed.

X-ray diffraction records were taken by CuK irradiation in a Guinier chamber at 40 kV — 20 mA dose rate with 8-hr exposures. The record for cellulose consists of a single crystalline band corresponding to $D = 2.85 \text{ \AA}$ (Fig. 12, band 4 from above). In the record of polypropylene 4 bands are observed. Their angles refer to $D = 6.25, 5.25, 4.80$ and 4.10 \AA . Location and intensity of bands agree with literary data²⁴ Figure 12, band 1 from above.

In case of the mixture, bands of both components are present (Fig. 12, band 2 from above). The record of cellulose encapsulated with polypropylene is identical to that of the mixture (Fig. 12, band 3 from above). Intermediate structures or changes characteristic to stresses in the crystals do not appear (Fig. 12).

Based on the data of the infrared spectroscopical and X-ray diffraction measurements, no chemical connection can be assumed between the polypropylene and the cellulose fibers (Figs. 11 and 12). In formation of a paper sheet, the connection among the cells is effected by both mechanical

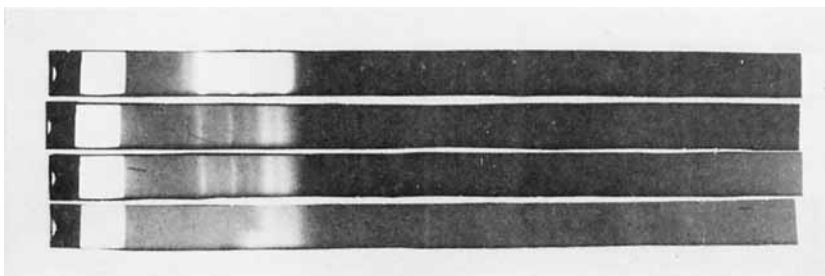


Fig. 12. X-ray diffraction studies

1st band from above:	Polypropylene
2nd " " "	Mechanical mixture of cellulose—polypropylene (55:45 w/w).
3rd " " "	Cellulose encapsulated with polypropylene (55:45 w/w)
4th " " "	Cellulose

and chemical forces. The mechanical connection is produced by hydrogen bonds. Longspun cells are capable of felting whereas hydrogen bonds are permitted by OH groups of hemicelluloses accompanying the cellulose. Mechanical properties of a paper produced from cellulose are predetermined by the size of the fibers, strength of the individual cells and number of the hydrogen bonds.

Strength of a paper containing polypropylene is determined by its porosity and by the bonds of cellulose-to-cellulose, polypropylene-to-polypropylene, and polypropylene-to-cellulose fibers. It was, therefore, to be expected that mechanical strength of the paper would be reduced (Figs. 7 and 8). It should however, be considered that presence of the thermoplastic polypropylene markedly improve several other detrimental properties of the cellulose. Sheets formed from this coupled composition exhibit considerable higher chemical resistance, water repulsion, resistance to oxidation as well as heat resistance. The latter permits a long-term sheet formation at 170°C. Poor mechanical strength of the cellulose encapsulated with polypropylene does not involve its unsuitability for application in the paper industry. Laminated sheets are known to peel, i.e., the plastic layer is disjoined from the paper. Using small amount of coated cellulose in the production of the paper, it rises to the surface of the sheet because of its lower specific gravity and this interlayer can provide good contact with both the cellulose and plastic film.¹⁹

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