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THE INFLUENCE OF COATING ADHESIVES
ON THE STIFFNESS OF PAPER

by
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A Thesis submitted to the
Faculty of the Department of Paper Technology
in partial fulfillment
of the
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TABLE OF CONTENTS

	<u>Page</u>
INTRODUCTION	1
OBJECTIVE	8
EXPERIMENTAL PROCEDURE	9
DATA	11
DISCUSSION	14
CONCLUSION	18
GRAPHS	19
SCHEMATIC DRAWINGS OF INSTRUMENTS	36
BIBLIOGRAPHY	39

ABSTRACT

The main purpose of this project was to study the effect coating adhesive has on the stiffness of paper. This work enables one to compare data on what influence adhesive with and without supercalendering has on the stiffness and caliper of the paper. The project was performed with six formulations with a different adhesive or adhesive combination in each. The adhesives were six types, casein, protein, starch, casein + latex, protein + latex and starch + latex.

It has been found in general that stiffness of all the paper samples increased when coating adhesive was applied on the surface. The starch gave the higher stiffness followed by protein and casein in that order. By adding the latex to the casein, starch and protein formulations the stiffness decreased in all cases. It was also noted that the stiffness was higher in the machine direction than in the cross direction of the paper. Stiffness and caliper decreased in all cases with supercalendering.

INTRODUCTION

Webster defines an adhesive as "a substance which causes bodies to adhere to each other" (6). This process by which bodies adhere to each other is still not clearly understood, although much progress in the field is being made.

For many years adhesives have been in use for bonding a variety of materials, particularly paper. Interest in adhesive and adhesive bonding has increased very significantly, however, in the past fifteen to twenty years, largely because of new developments in synthetic resins and because of the unusual demand for better methods of fabrication brought about by World War II, and the current military situation (6).

There are several current theories or ideas concerning the nature of adhesion which at first glance does not appear to be compatible. However, closer examination and study reveal that they are all probably true. Theories advanced by each author contain the most important factors for the particular system which was studied. However, the group of factors which are predominant vary from system to system (6).

The significant factor in adhesion is the molecular attraction operative between the adhered and the adhesive. These molecular attractive forces are the cause of adhesion, the intrinsic strength of an adhesive bond can be no stronger than the total of the molecular forces operative (5).

The forces responsible for molecular adhesion are essentially the same as those responsible for the cohesion of solids, namely primary and secondary valence forces (8).

The primary bonds in organic and polymeric systems are of the covalent type whereby an electron pair between atoms is shared resulting in relatively high bond energies of about 100 to 200 K-cal per mole. These forces hold the carbon-carbon and other atoms together in the backbone of polymer.

Secondary forces are of four types viz. London - dispersion, Keeson - orientation, Debye - induction and hydrogen bonds. Their interaction energies range up to about 10 k-cal per mole with hydrogen bonds being the strongest, the orientation forces ranking second, then the dispersion forces followed by the relatively weak induction forces. The basic nature of these forces is electrical in nature and is due to the fact that most molecules have polarity, i.e. have separated centers of positive and negative charge. The more asymmetric the molecule the greater the dipole moment and the greater the attraction. Orientation forces are interactions between permanent dipoles whereby the positive and negative centers attract each other and also exert an orienting effect on other molecules. Induction forces have their origin in the capacity of nonpolar molecules to become polarized under the influence of other molecules which have strong dipole moment. Dispersion forces are universal forces of great importance to all intermolecular systems, but especially if there are no polar molecules involved.

THEORY OF ADHESION

a. Adsorption Theory

A large proportion of investigators now believe that the most fundamental prerequisites for good adhesion is uniform and unlimited molecular contact between the adherends. This view is basis for the adsorption theory of adhesion. Molecular forces between two materials are theoretically sufficient for strong adhesion.

The wetting of the solid by the adhesive prior to its solidification to achieve good interfacial molecular contact is acknowledged to be a most critical factor in adhesion. The Young and Dupre relation showing the balance between contact angle originally is:

$$\rho_{sv}^0 = \rho_{sl} + \rho_{lv}^0 \cos \theta$$

Where θ is the surface tension, sv^0 and lv^0 refer to the solid and liquid in equilibrium with the saturated vapor and sl to the interfacial tension.

b. Mechanical Theory of Adhesion

The early research of McBain (9) resulted in his conclusion of the existance of two types of bonds.

1. Specific or chemical adhesion between smooth, dense surface and involving secondary valence force.

2. Mechanical adhesion between porous surface involving penetration and hooking of the dried adhesive with no need for intermolecular forces.

Bikerman (10) feels that proof for mechanical adhesion lies in the insensitivity of such bonds to impurities which cause weak boundary layer. It is difficult experimently to separate the specific and mechanical effects.

c. Electrical Theory of Adhesion

Skinner and co-workers (11) and Dergaguin and co-worker explained the increase found in the work of stripping a polymer film from an adherend with increase in rate of stripping as being due to electrostatic attraction between charged layers at the interface. Also, they felt that the action of intermolecular forces should be independent of velocity of testing. The elctrostatic theory was supported by studying the occurrence of electrical discharge during the breaking of certain joints.

d. Diffusion Theory of Adhesion

The diffusion theory of adhesion by Voyutskii and co-workers (12) states that adhesion is a result of interdiffusion of polymer molecules and their individual segments across the adhesive-adherend interface. Upon completion of the bonding process, the interface

no longer exists. Thus, adhesion is visualized as an interfacial mutual solubility phenomenon as contrasted to a surface phenomenon, as in the case of the adsorption and electrical theories. Most of the studies have been carried out using elastomeric polymers and have indicated that diffusion is inversely proportional to molecular weight, increases with temperature and time of contact and is dependent on the molecular compatibility of the adhesive and adherend as judged by solubility parameter and similarly in polarity.

e. Rheological Theory of Adhesion

Bikerman (10) feels that the strength of an experimental adhesive bond has only a small relation to molecular adhesion, because when such a bond is ruptured, failure never proceeds along the adhesive-adherend interface, therefore for a proper bond the forces between the adhesive and adherend do not influence the measured force to break the joint. Rupture occurs cohesively within one of the materials and therefore, is a rheological problem.

SUMMARY OF ADHESION

1. Actual strength of secondary bonds due to interaction of surface energies.
2. The extent and perfection of contact between adhesive and adherend.

3. Presence or absence of other stress concentration effects caused by variation in cross section, dimensional change during drying or conditioning.

4. Presence or absence of flaws and impurities at the interface or within the adhesive.

Stiffness

Stiffness is that property of a material that resists bending and flexing. This property is dependent upon the modulus of elasticity and moment of inertia.

The moment of inertia is an expression of the internal resisting moment that is set up in the body when subjected to a bending moment. The moment of inertia I of a body with rectangular cross section area is

$$bh^3/12 \tag{1}$$

where b = length of base

h = height or thickness of a body

From equation (1) it is apparent that stiffness will vary with the cube of the thickness. This is true for homogenous materials but for paper there are some limitations. Smith found that rigidity increases with the cube of caliper at constant density and with the square of caliper at constant weight.

Clark defines (1) stiffness as the ability of paper to support its own weight. He represents this definition as

$$L^{3/100}$$

where L is the overhang of a standard size sample clamped horizontally.

Casey defines (2) stiffness with the formula.

$$\frac{ET^2}{12} \times \frac{W}{L^2}$$

Where E = Young's modulus

W = Sample width

L = Sample length

T = Sample thickness

Another definition of stiffness that is more popular than is given by Carson. Carson defines (4) stiffness as the bending moment per unit specimen width and per unit specimen curvature at the torque axis. This may be expressed as

$$\frac{ML}{3b} f(\theta)$$

Where M = The bending moment at the torque axis

L = Bending length

b = Specimen width

θ = Bending angle

This definition by Carson agrees with the definition of flexural rigidity given by Pierce.

OBJECTIVE

The purpose of this experimental program was to study the effect coating adhesives has on the stiffness of paper.

EXPERIMENTAL PROCEDURE

Sixty grams of Penford gum 280 was stirred in 690 c.c. of water and heated to 180^oF. while stirring in an Eppenbach stirrer. After the starch had been held at 180^oF. for five minutes, one gram of TSPP and 500 grams of ultra white clay was added. This mixture was stirred for at least ten minutes in the mixer before using. The Keegan Coater was used for coating. The same procedure was followed for protein and casein. Latex was added to the above formulae for the latex series.

The caliper of the paper, percentage solid of the coating, temperature, tension, drying rate were held constant through the whole project. The viscosity varied with adhesive to adhesive.

After coating, the paper was conditioned for 24 hours in an atmosphere maintained at 50 \pm 2% relative humidity and 73 \pm 2^oF. temperature. It was then tested for stiffness. The samples were cut from the heart of the samples in the machine direction and in the cross machine direction with dimensions of 1-1/2 x 2-3/4 inches. Stiffness was run in the machine and cross machine direction of all samples.

Stiffness test fall into three categories according to Casey (2). Type one measures the force required to bend the paper through a given angle. Type two, measures the angle through which the paper is bent with a given load. Type three, measures the angle through which the paper bends under its own weight.

The Taber stiffness tester has been chosen for this work due to the two reasons. It is fairly easy to operate correctly, and the values for the stiffness are not arbitrary. The Taber instrument uses a specimen 1-1/2 x 2-3.4 inches which is fastened to a clamp on a pendulum. A force is applied to the lower end of the specimen by rollers attached to a power driven disk; and the specimen is deflected. The end point of the test is when the specimen has been deflected a predetermined amount. To compensate for curl, the test was run in both the directions. Refer to Tabor Bulletin 4506-10M for further details.

DATA

STARCH

	<u>MD</u>	<u>CD</u>
Stiffness Reading	58.75 - 76.00	35.80 - 73.20
Stiffness after 1 nip	49.40 - 62.80	32.20 - 61.30
Stiffness after 2 nips	46.40 - 60.20	30.40 - 58.40
Stiffness after 3 nips	45.80 - 56.80	30.00 - 56.40

STARCH + LATEX

Stiffness Reading	55.40 - 69.40	45.00 - 63.00
Stiffness after 1 nip	51.60 - 61.20	37.00 - 60.60
Stiffness after 2 nips	50.40 - 59.20	37.00 - 58.20
Stiffness after 3 nips	48.40 - 56.00	36.80 - 54.60

PROTEIN

Stiffness Reading	56.30 - 74.80	26.00 - 66.80
Stiffness after 1 nip	49.00 - 66.20	31.60 - 59.60
Stiffness after 2 nips	46.60 - 62.20	32.60 - 59.40
Stiffness after 3 nips	45.20 - 60.20	33.60 - 58.20

PROTEIN + LATEX

	<u>MD</u>	<u>CD</u>
Stiffness Reading	53.40 - 74.40	28.80 - 67.30
Stiffness after 1 nip	48.80 - 62.00	35.00 - 56.20
Stiffness after 2 nips	47.60 - 56.00	36.00 - 55.80
Stiffness after 3 nips	46.40 - 55.40	37.00 - 52.40

CASEIN

Stiffness Reading	52.40 - 76.80	31.00 - 68.00
Stiffness after 1 nip	46.80 - 57.60	33.00 - 63.00
Stiffness after 2 nips	45.80 - 55.40	34.70 - 58.80
Stiffness after 3 nips	45.00 - 55.00	35.60 - 54.30

CASEIN + LATEX

Stiffness Reading	48.00 - 76.30	22.00 - 72.70
Stiffness after 1 nip	47.00 - 57.00	28.00 - 53.00
Stiffness after 2 nips	46.70 - 54.20	30.00 - 52.40
Stiffness after 3 nips	45.30 - 53.20	31.00 - 52.00

CALIPER

	<u>0 Nip</u>	<u>1 Nip</u>	<u>2 Nips</u>	<u>3 Nips</u>
Paper Stock	4.08	3.52	3.32	3.28
Casein	4.50	3.76	3.62	3.52
Casein + Latex	4.48	3.72	3.56	3.40
Starch	4.46	3.82	3.70	3.50
Starch + Latex	4.58	3.76	3.62	3.54
Protein	4.62	3.90	3.70	3.66
Protein + Latex	4.62	3.70	3.66	3.62

STIFFNESS OF PAPER STOCK

	<u>MD</u>	<u>CD</u>
Stiffness Reading	47.9	44.1
Stiffness after 1 nip	45.8	41.5
Stiffness after 2 nips	44.6	40.8
Stiffness after 3 nips	43.7	40.3

COAT WEIGHT

Coat Weight of Starch	=	11.2177
Coat Weight of Starch + Latex	=	11.7655
Coat Weight of Protein	=	11.4139
Coat Weight of Protein + Latex	=	11.9400
Coat Weight of Casein	=	11.3657
Coat Weight of Casein + Latex	=	10.7580

DISCUSSION

Figure one shows the stiffness of the paper stock in both the machine direction and in the cross direction. The stiffness of the cross direction is lower because the majority of the fibers as they flow onto the wire orient themselves in the direction of their flow just as logs would do in flowing down a stream.

Figure two shows the stiffness of the coated side of the paper in the machine direction after applying the coating adhesives - starch and starch + latex. Figure three shows stiffness of the same paper, but of the opposite side. The readings of both sides of the coated paper sample have not been combined to take the average because there was a great difference in the stiffness reading of the same sample in both directions. By comparing figures two and three with figure one, it is obvious that the stiffness has been increased by applying the coating adhesive on the surface of the paper. By applying the coating adhesive, adhesion took place which effected the stiffness of the paper. Figure two and three shows the two curves. Figure two shows the starch and the starch plus latex. Starch alone gives a higher stiffness than the combination of the starch and latex. By adding the 10% latex on the basis of the percentage solid to the coating formulation, it decreases the stiffness by five percent on the coated side and nine percent on the opposite side.

In figures two and three, both of the curves are decreasing. This indicates the effect of supercalendering on the stiffness of the paper. Both of the curves show that, in both cases, the stiffness decreases. After the first nip, there was a great reduction in stiffness in all cases and in the additional nips, there was only a slight change in the stiffness of the paper. Thus, by increasing the number of the nips, the stiffness and caliper of the paper was decreased. Casey (2) found that stiffness varied with the cube of the thickness of the paper.

Figures four and five show the stiffness of the starch and starch + latex formulation in cross direction of the paper. The stiffness of the starch in cross direction on coated side was lower but in the uncoated side it was higher. Both figures show, how the supercalendering effects the stiffness reading.

Figures six and seven indicate the stiffness of the coated and uncoated sides, respectively, in the machine direction of the paper after applying the adhesive-protein and protein plus latex. Both graphs show that the protein formulation gives a higher stiffness than the protein plus latex formulation. There was not much difference in the stiffness readings for both formulations. Like the previous graphs, these graphs illustrate the effect of supercalendering on the stiffness.

Figures eight and nine show the stiffness of the coated side of the paper in the cross direction by applying the protein and

protein plus latex, formulations. Figure eight shows an unusual curve, that is to say, instead of being a decreasing function, it is an increasing function after the supercalendering. Maybe the reason is that curling took place on the coated side of the paper. In figure eight, both curves illustrate the stiffness on the coated side of the paper. Thus, due to the curling, it shows the lower stiffness and increases after supercalendering. So the stiffness in these cases should be higher than what was actually found from three nips.

Figures ten and eleven show the stiffness of the coated and uncoated side in machine direction after applying casein and casein plus latex, and it shows decreasing curve which is usual for supercalendering. Figures twelve and thirteen illustrate the stiffness and influence of supercalendering on stiffness in the cross direction of the coated and uncoated side respectively. Here again, figure twelve shows the increasing function after supercalendering. It has been caused by the curling, due to the one side coating.

Figure fourteen indicates that the caliper has been decreased by supercalendering in the case of control stock paper. Casey (2) has shown that stiffness varies with the cube of the thickness of the paper. The curve shows that after the first nip there was a great reduction in the caliper but the additional nips make only a slight change in the caliper.

Figures fifteen through seventeen show the effect of supercalendering on the caliper in the cases of casein, casein + latex, starch, starch + latex, protein, and protein plus latex respectively. All of the curves show the same type of relationships. This is to say, supercalendering decreases the caliper of the paper.

CONCLUSION

The following conclusions can be drawn from the tables and datas.

1. The stiffness of the paper increased by applying the coating adhesives on the surface of the paper in all cases.
2. The starch gave the higher stiffness followed by protein and casein in the order given.
3. The stiffness of the paper decreased by adding ten percent latex to the adhesive formula.
4. By supercalendering the stiffness and caliper decreased.

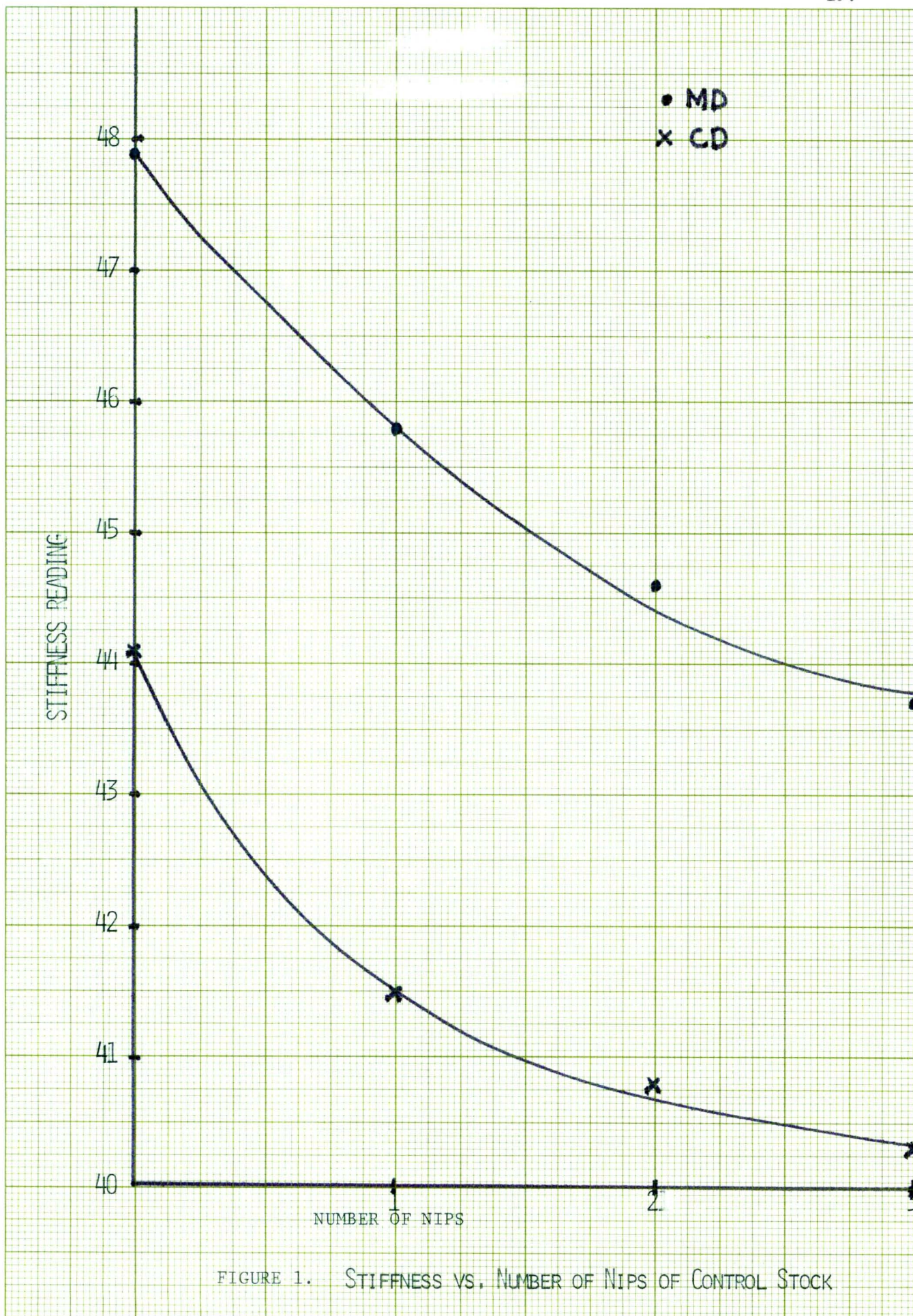


FIGURE 1. STIFFNESS VS. NUMBER OF NIPS OF CONTROL STOCK

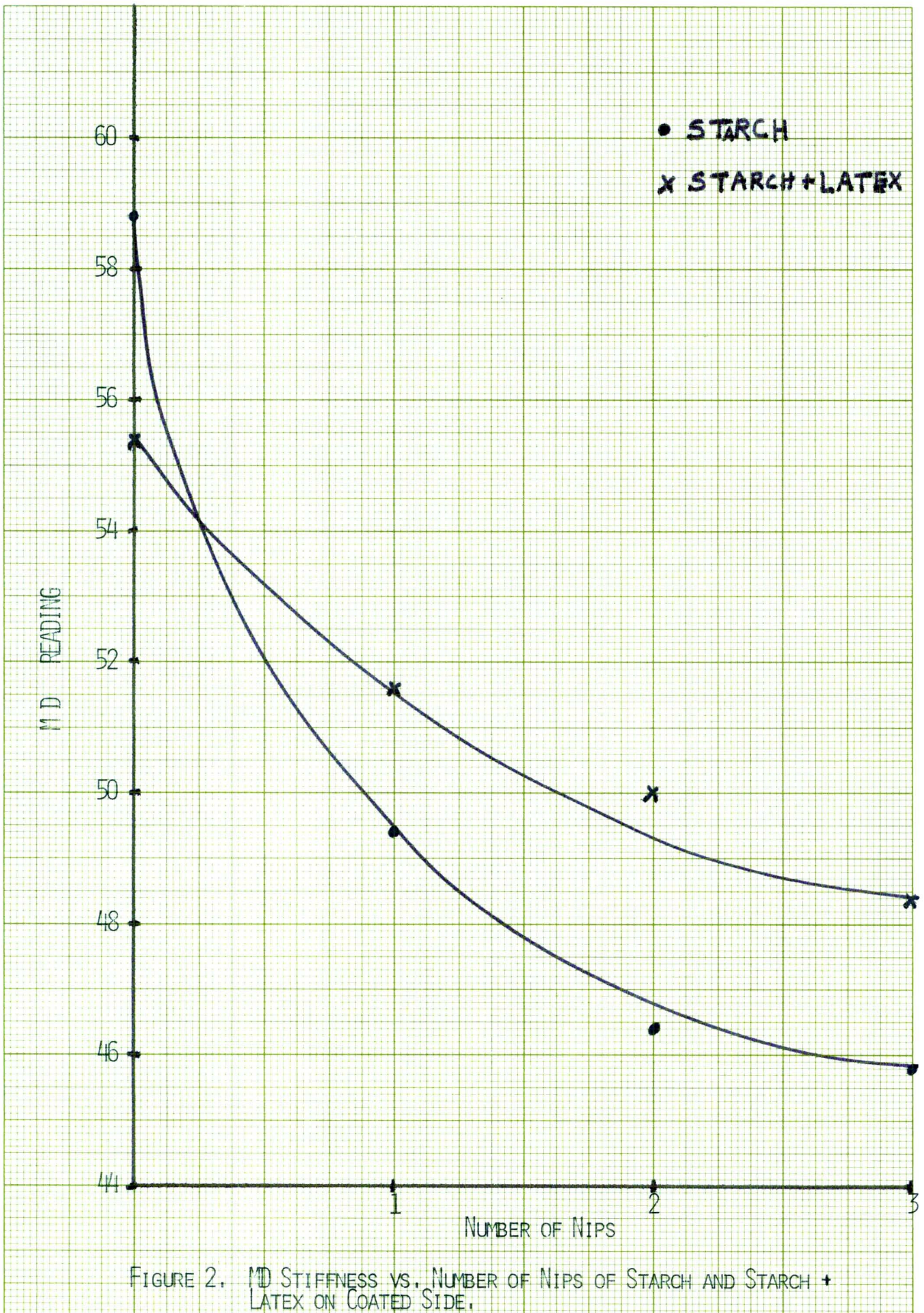


FIGURE 2. MD STIFFNESS VS. NUMBER OF NIPS OF STARCH AND STARCH + LATEX ON COATED SIDE.

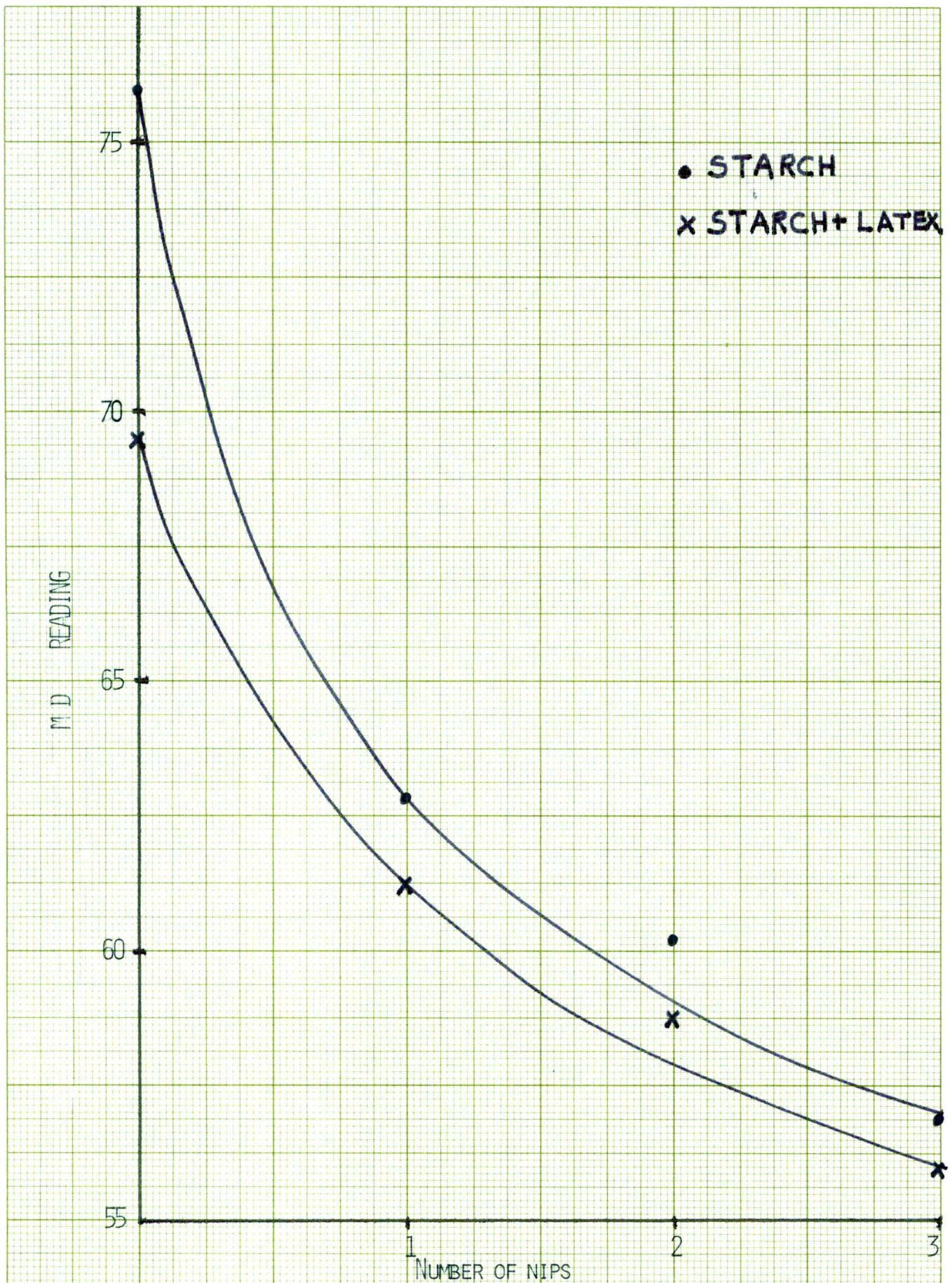
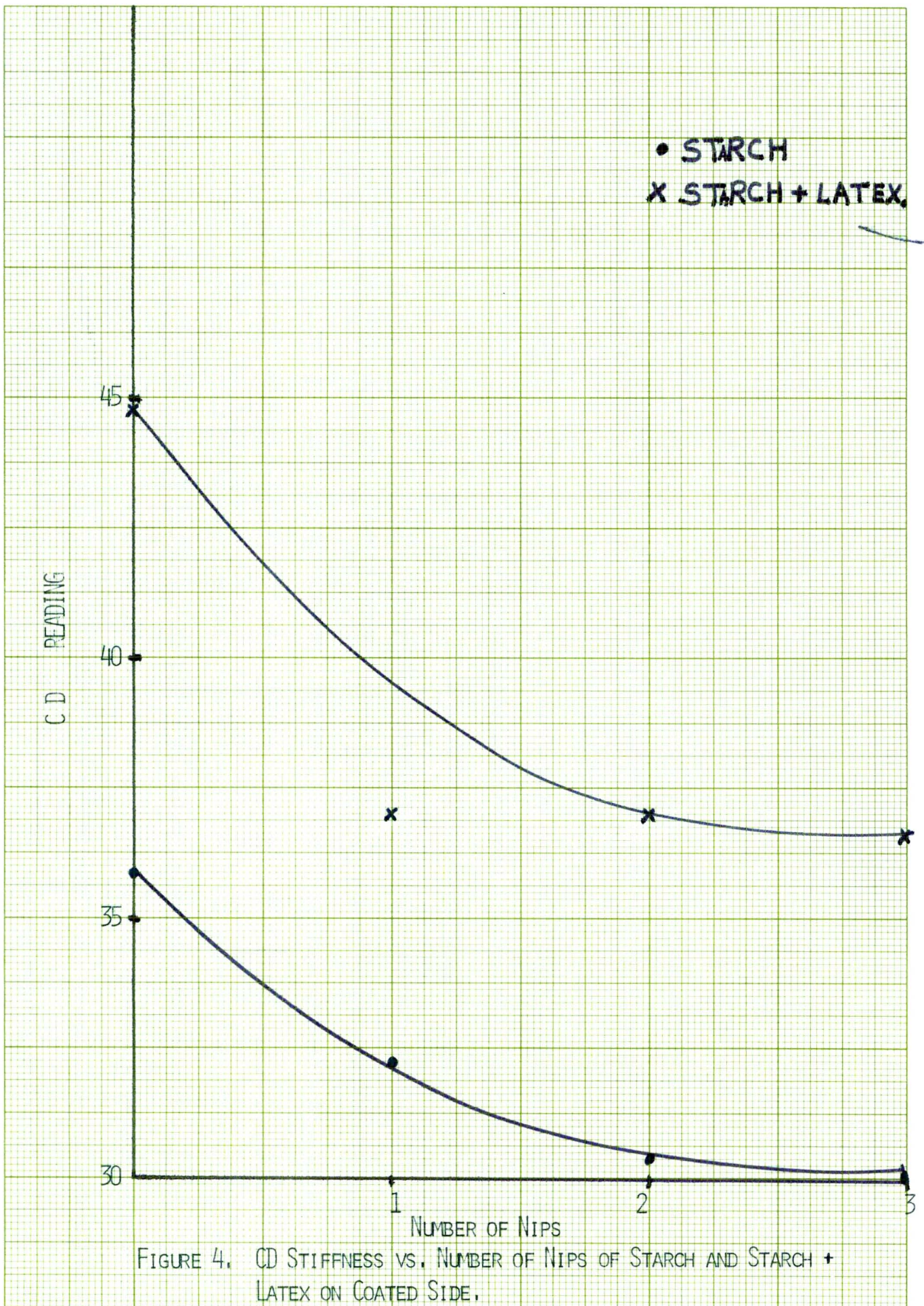


FIGURE 3. MD STIFFNESS VS. NUMBER OF NIPS OF STARCH AND STARCH + LATEX ON UNCOATED SIDE.



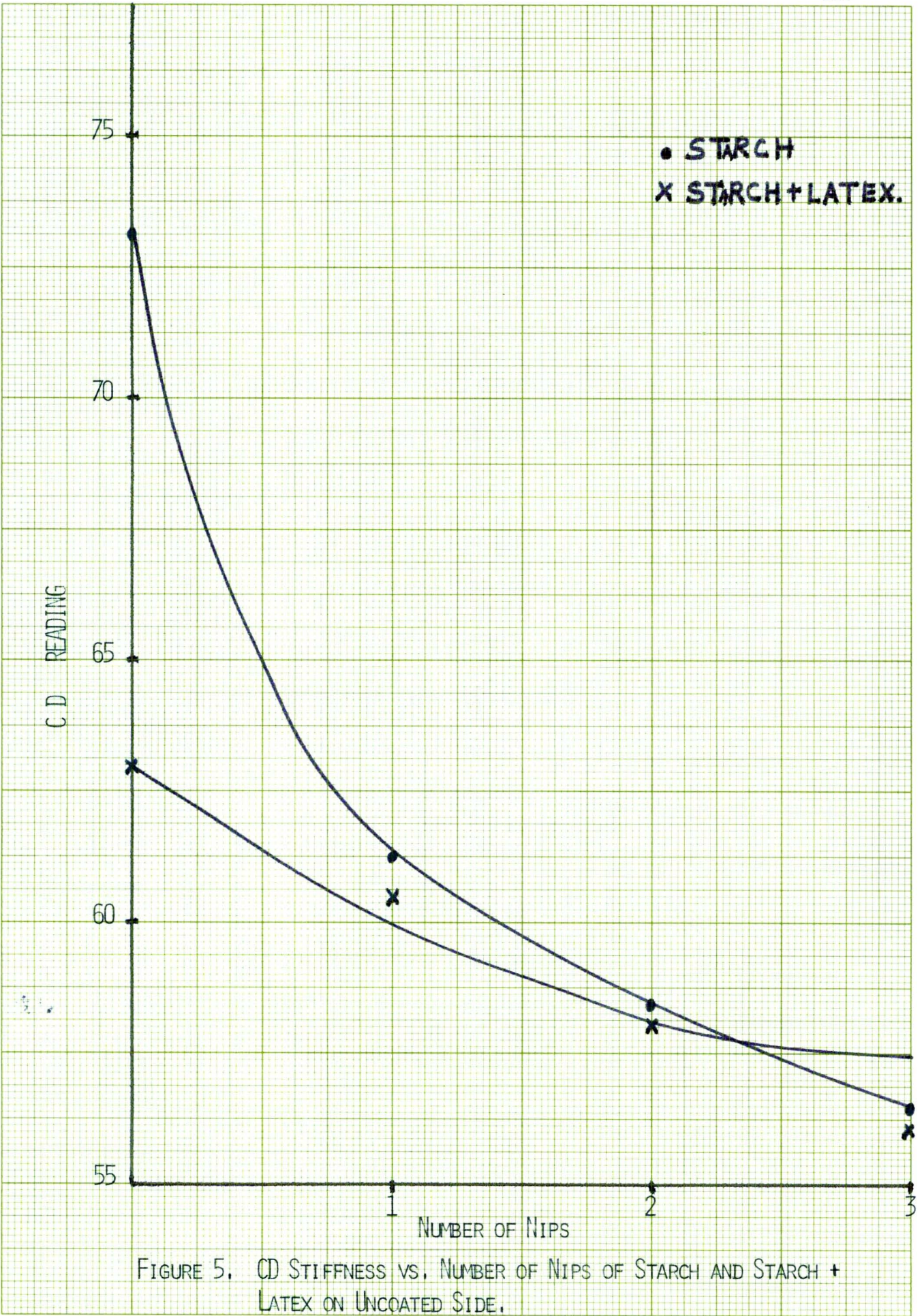


FIGURE 5. CD STIFFNESS VS. NUMBER OF NIPS OF STARCH AND STARCH + LATEX ON UNCOATED SIDE.

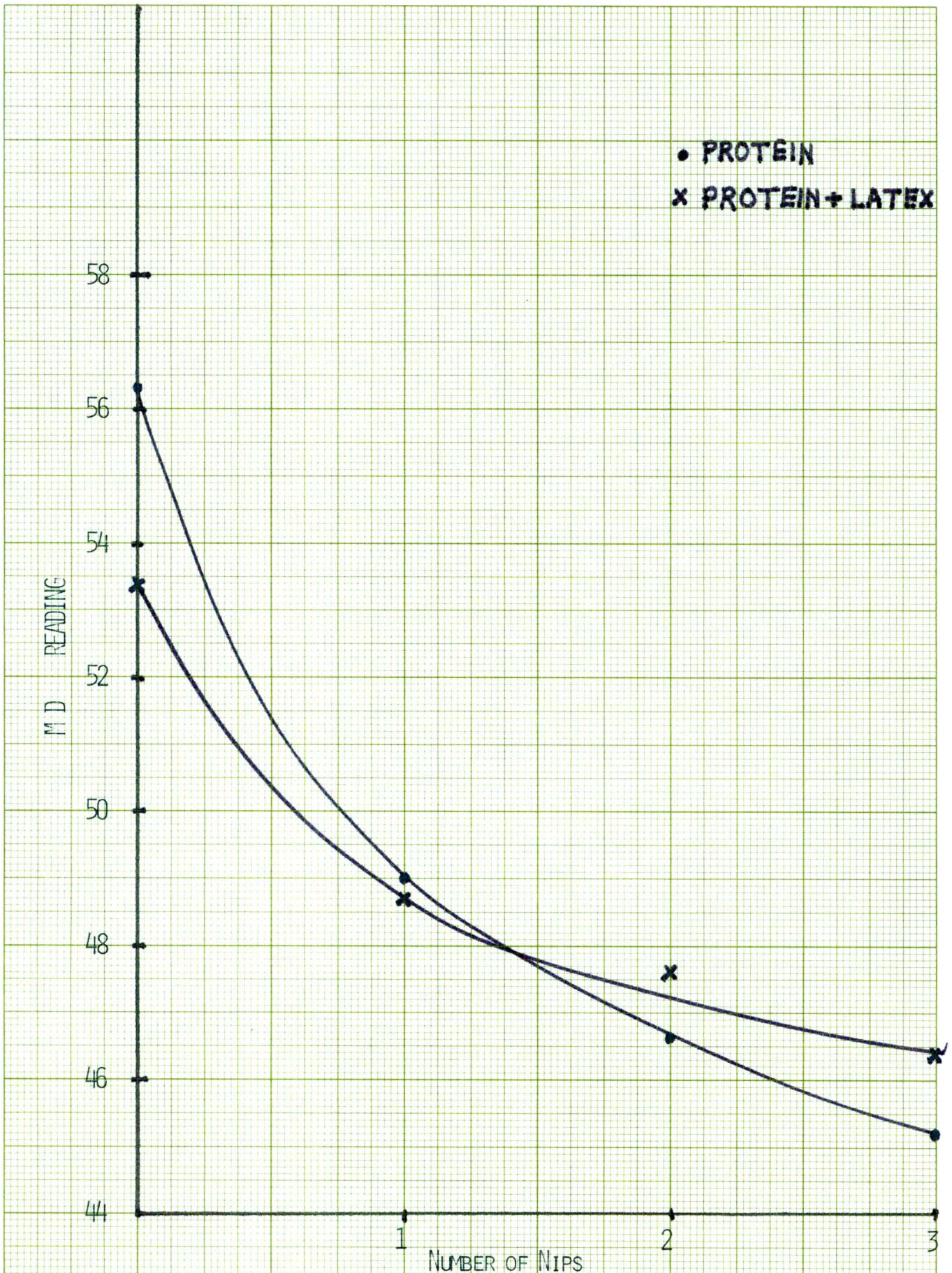


FIGURE 6. MD STIFFNESS VS. NUMBER OF NIPS OF PROTEIN AND PROTEIN + LATEX ON COATED SIDE.

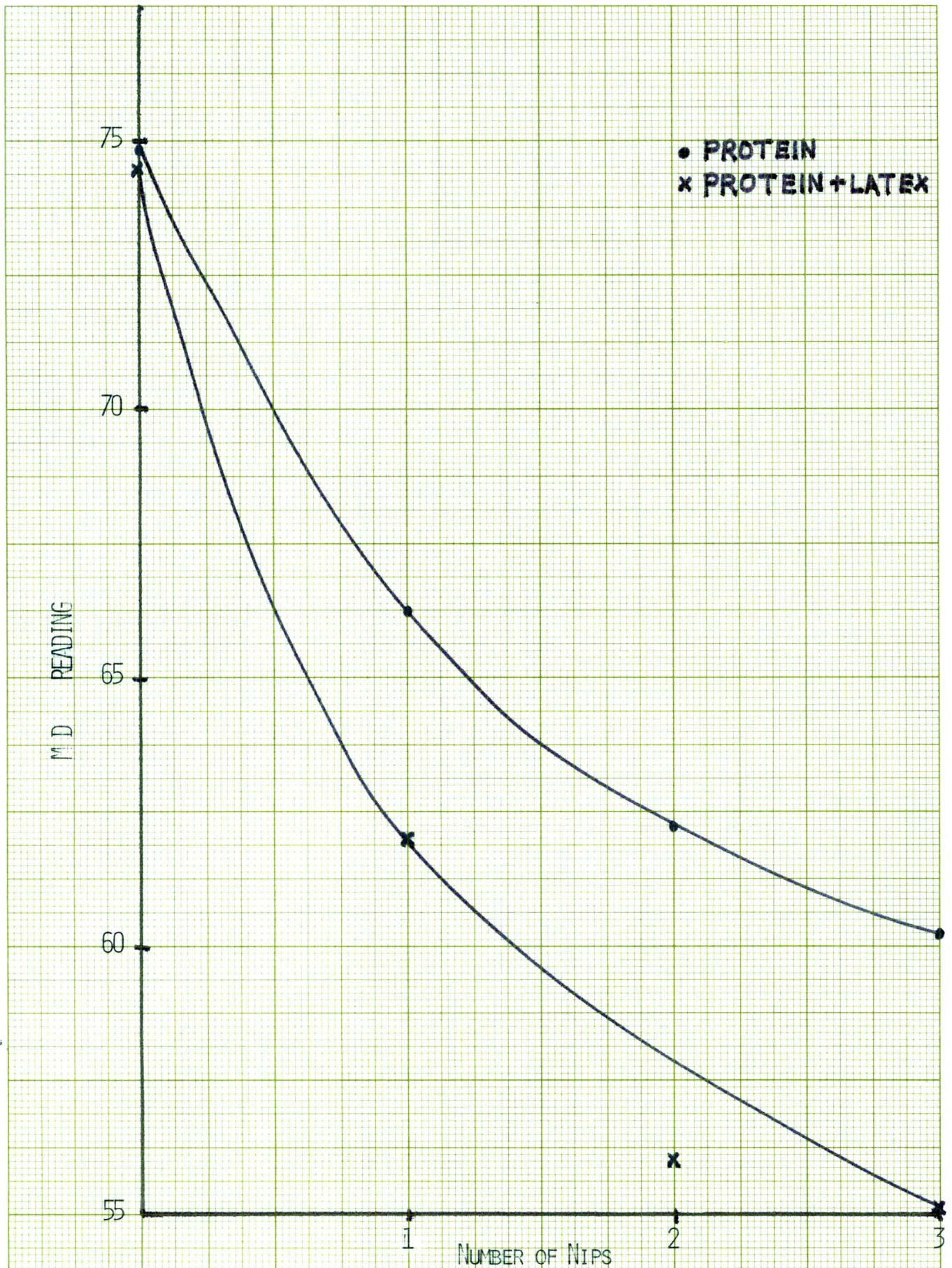
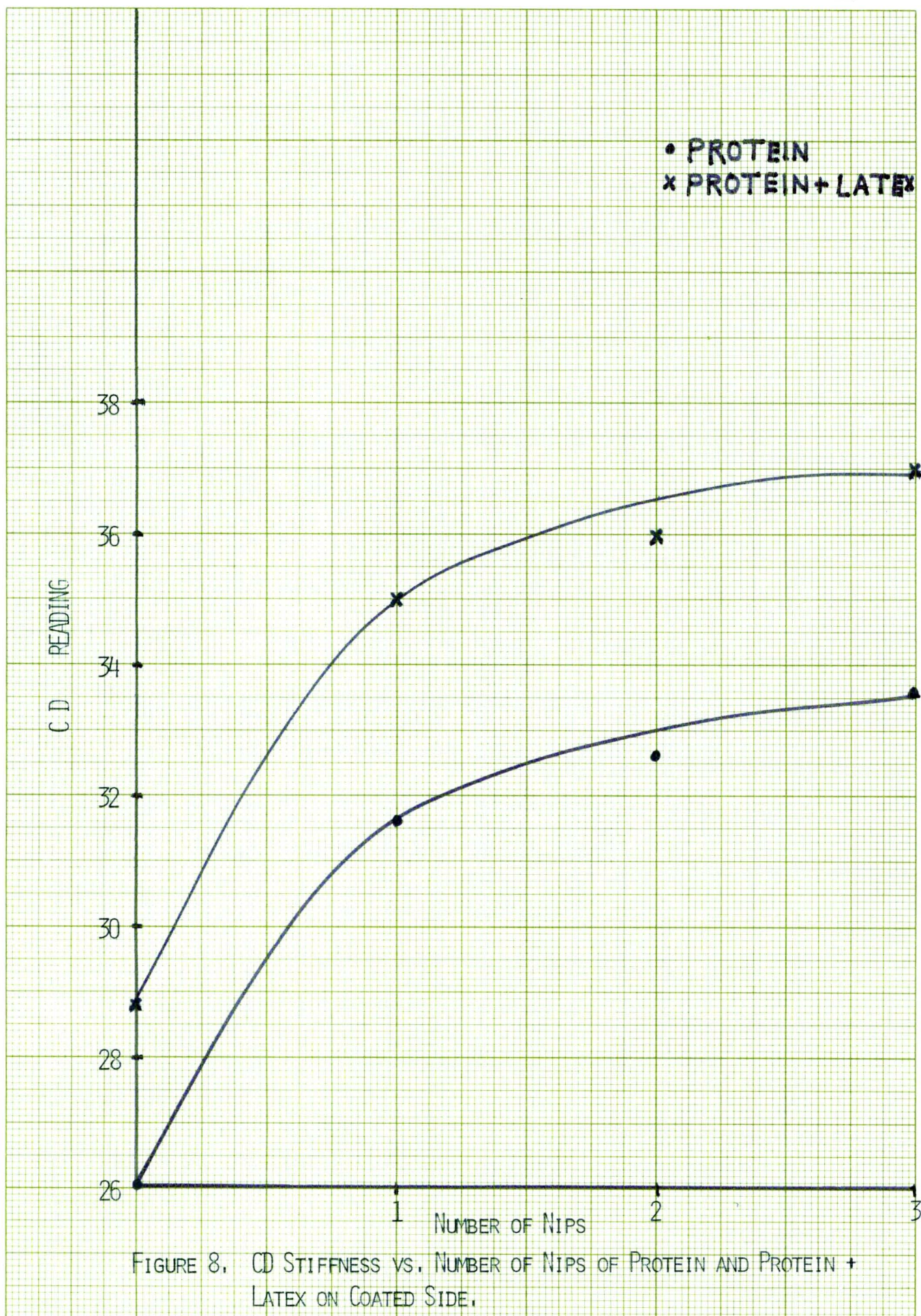
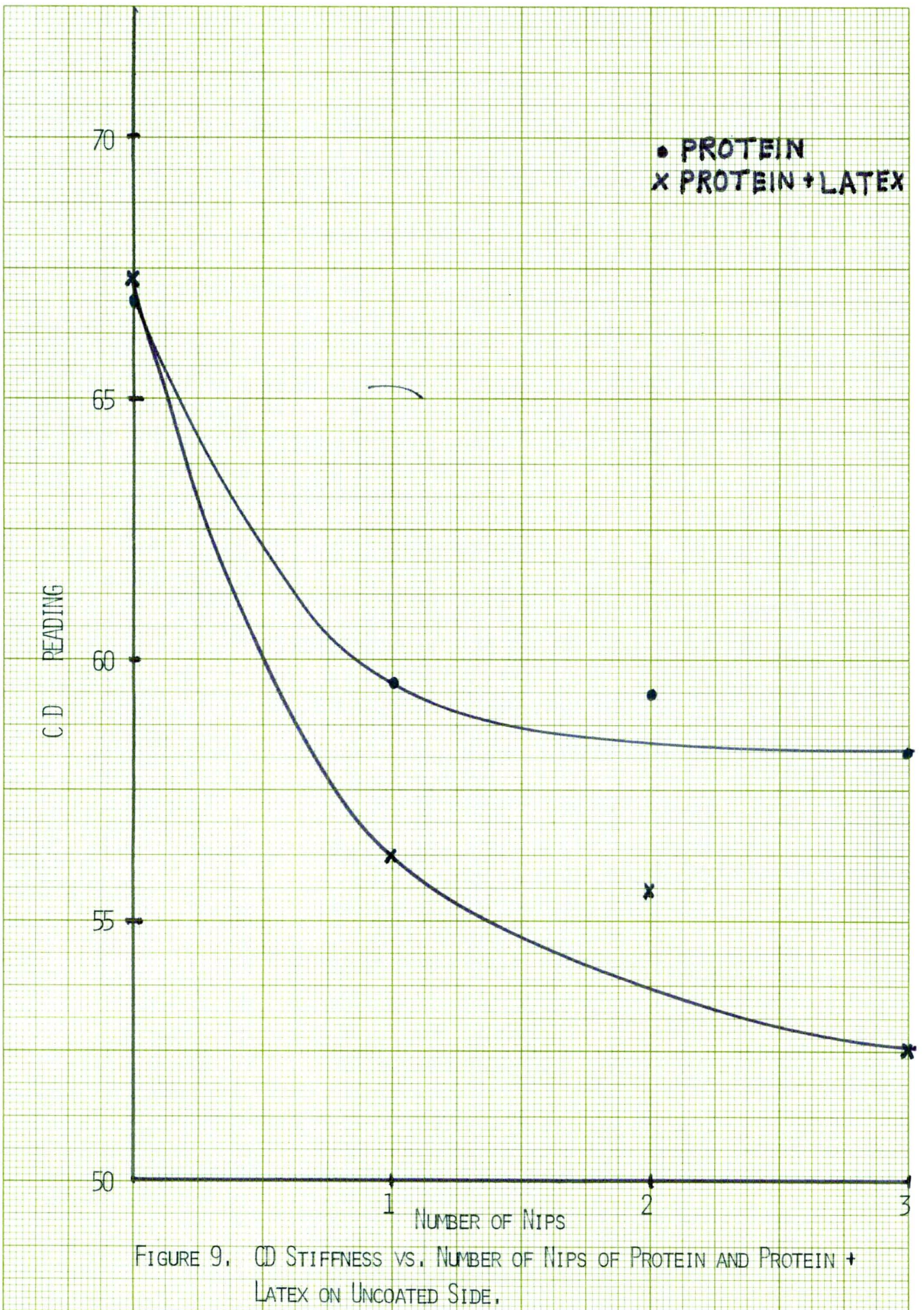


FIGURE 7. MD STIFFNESS VS. NUMBER OF NIPS OF PROTEIN AND PROTEIN + LATEX ON UNCOATED SIDE.





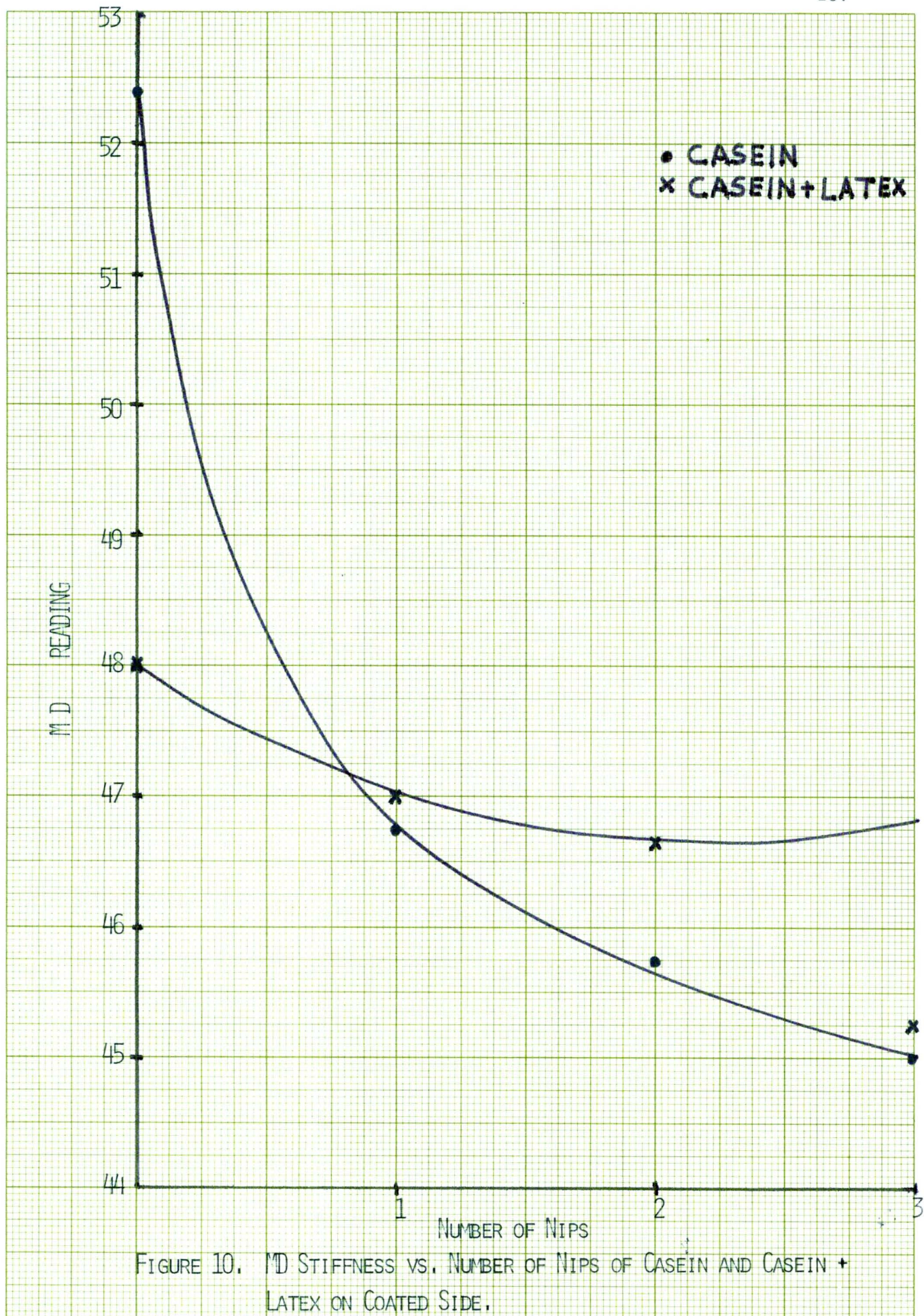
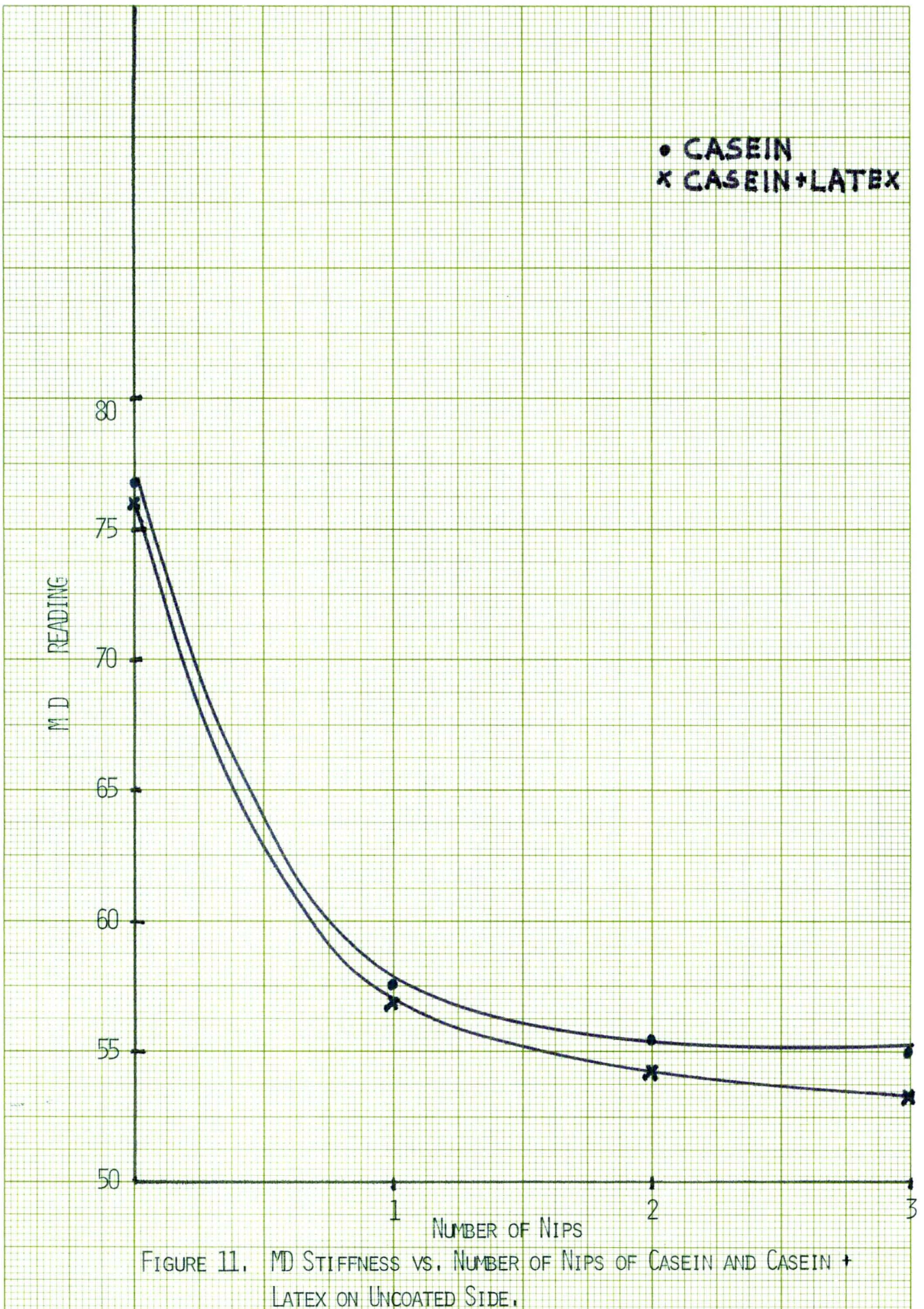
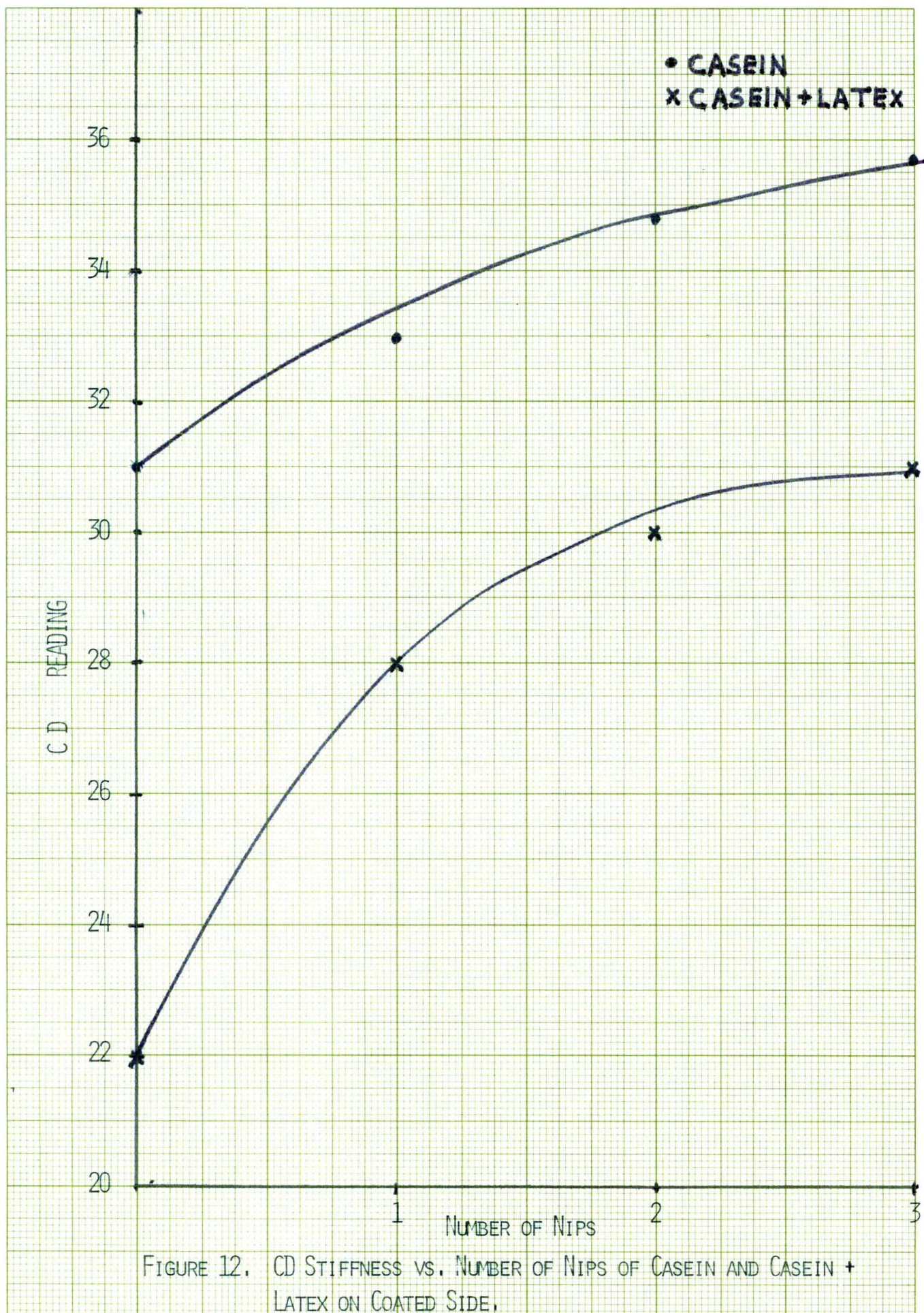


FIGURE 10. MD STIFFNESS VS. NUMBER OF NIPS OF CASEIN AND CASEIN + LATEX ON COATED SIDE.





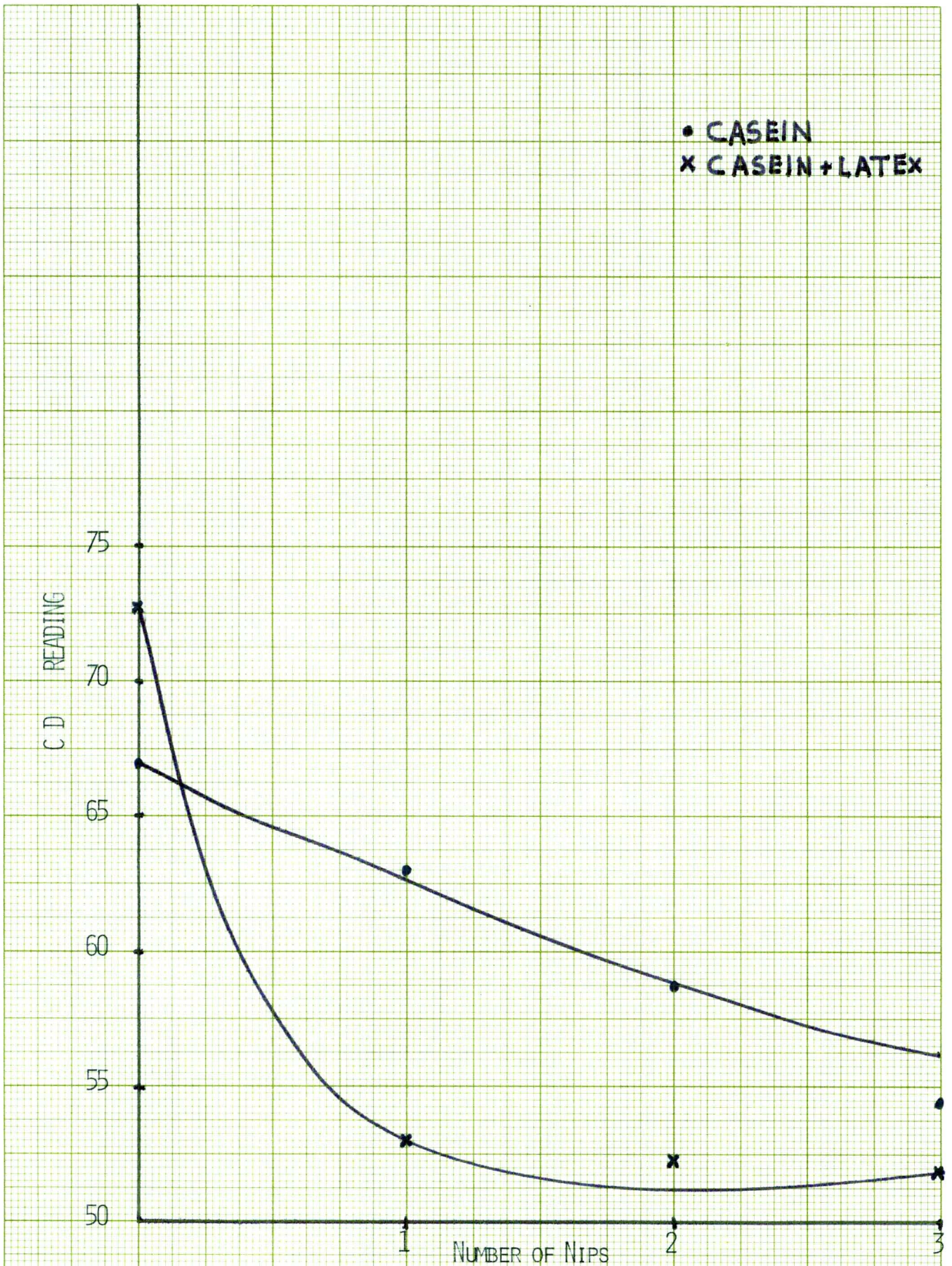


FIGURE 13. CD STIFFNESS VS. NUMBER OF NIPS OF CASEIN AND CASEIN + LATEX ON UNCOATED SIDE.

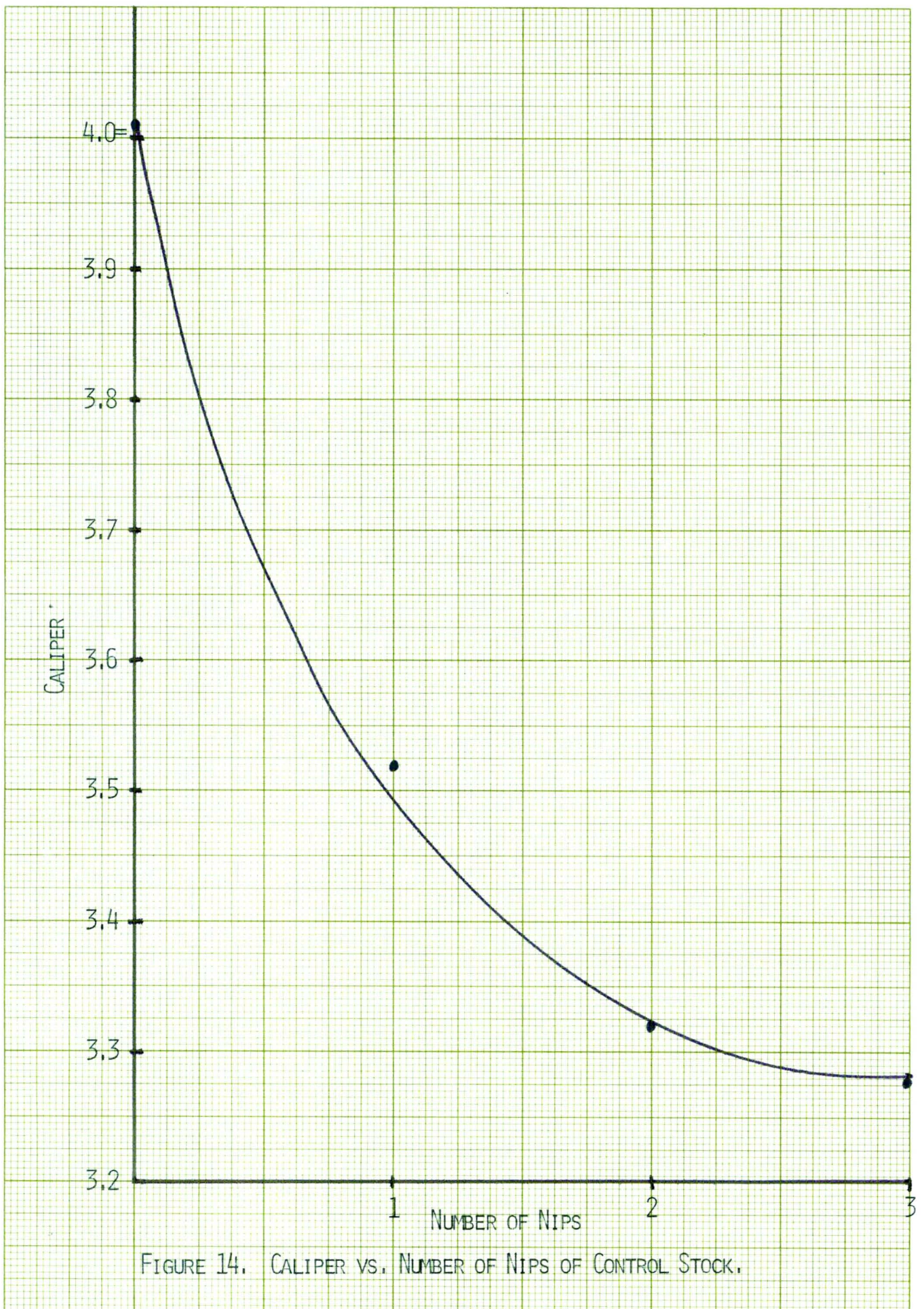


FIGURE 14. CALIPER VS. NUMBER OF NIPS OF CONTROL STOCK.

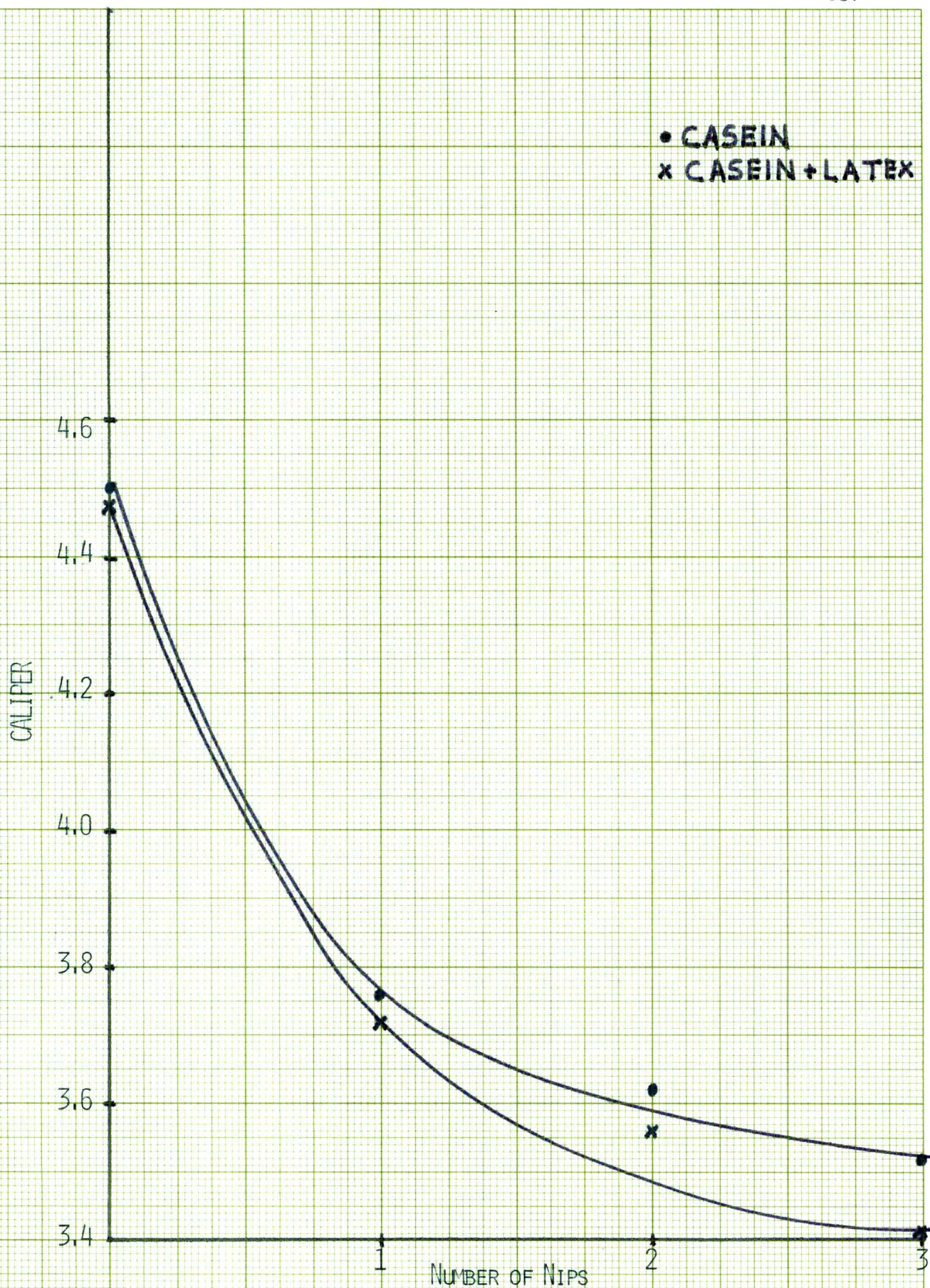


FIGURE 15. CALIPER VS. NUMBER OF NIPS OF CASEIN AND CASEIN + LATEX.

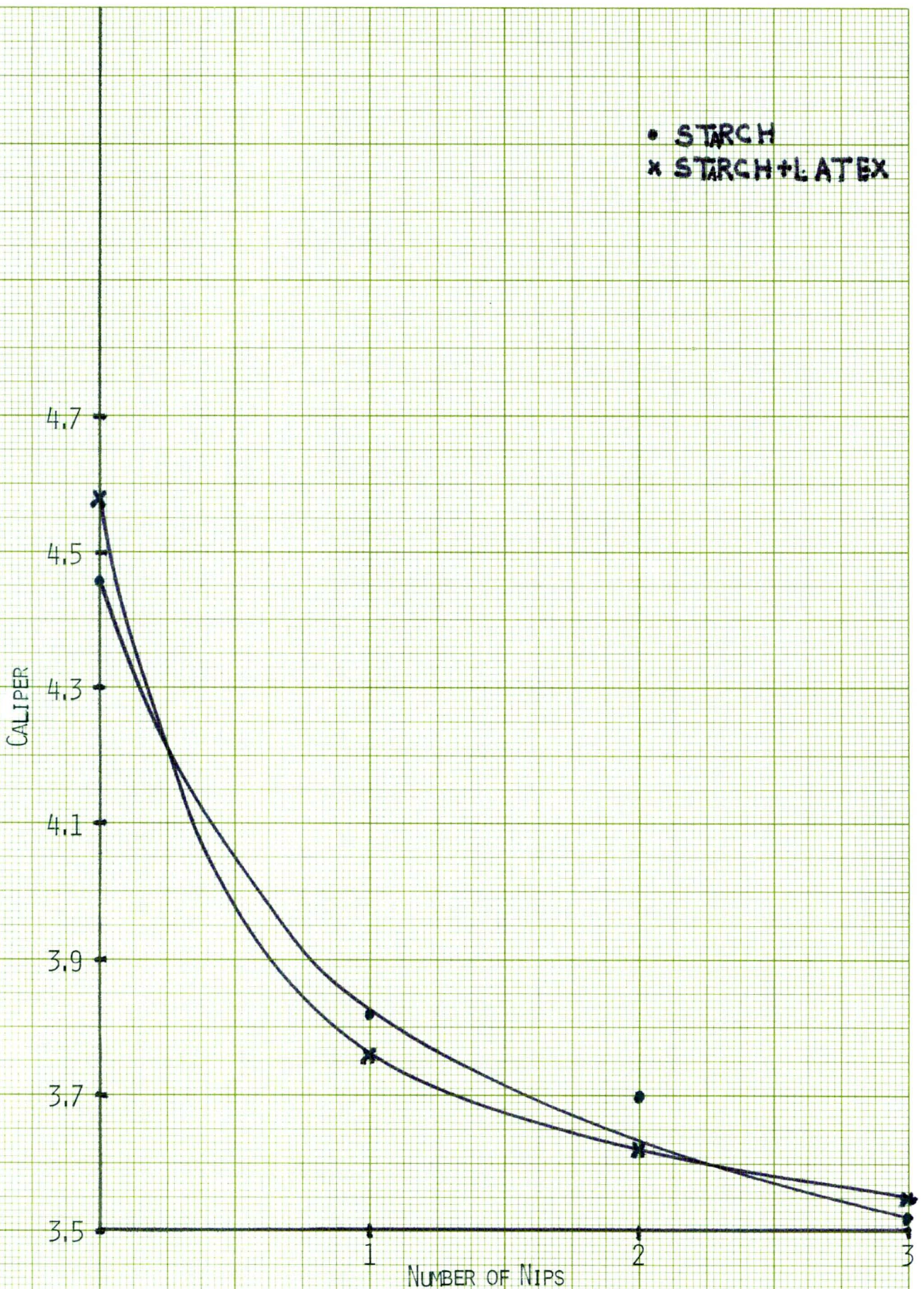


FIGURE 16. CALIPER VS. NUMBER OF NIPS OF STARCH AND STARCH + LATEX.

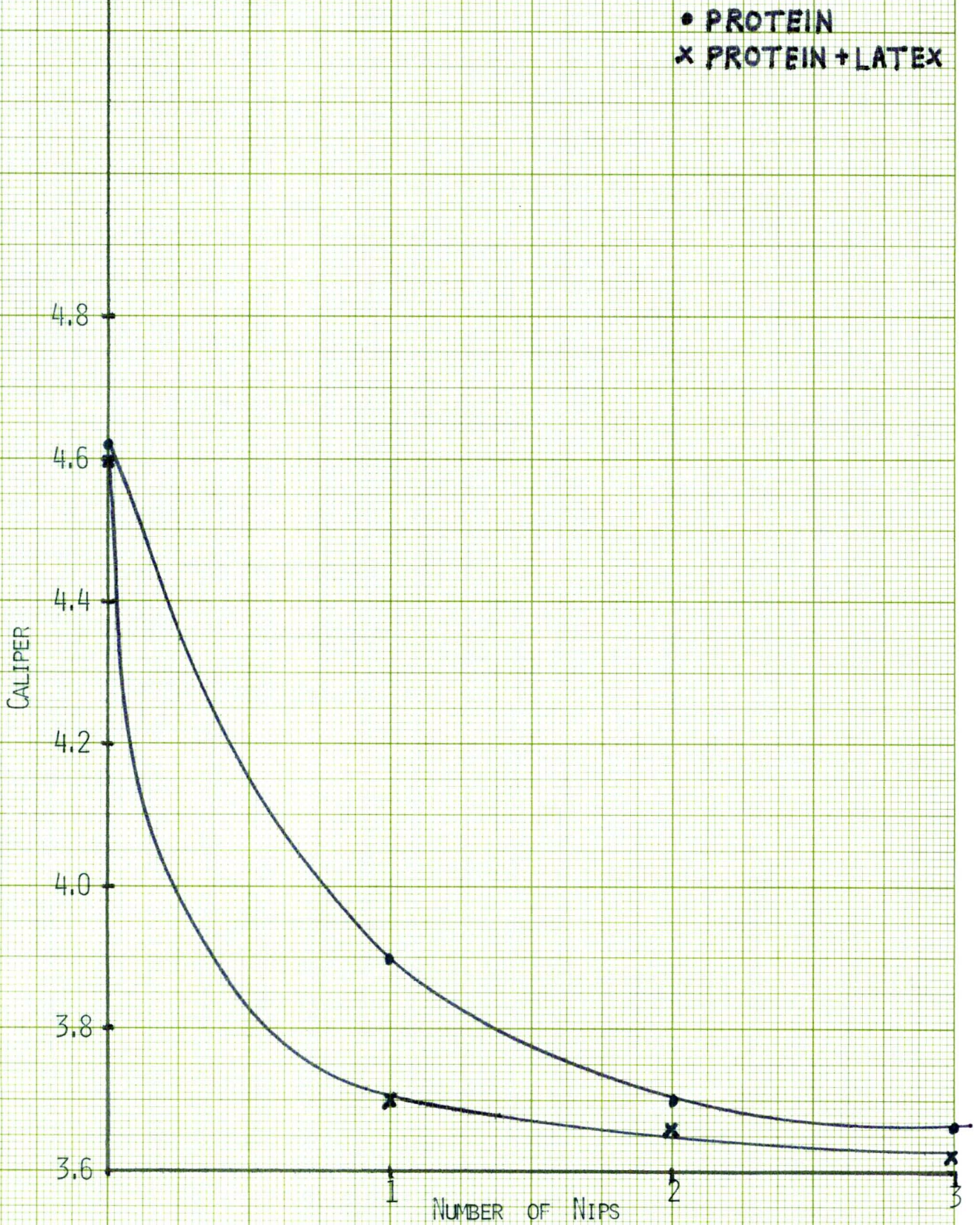


FIGURE 17. CALIPER VS. NUMBER OF NIPS OF PROTEIN AND PROTEIN + LATEX.

NO.	DESCRIPTION
1	UNWIND ASSEMBLY
2A	TRAILING BLADE ASSEMBLY
2B	BACKUP ROLL T. BLADE
2C	T. BLADE TENSION CONTROL
3A	STEEL BACKUP ROLL - SIZE PRESS
3B	PNEUMATIC CYLINDER - SIZE PRESS
3C	NEOPRENE PICKUP ROLL - SIZE PRESS
4	STAINLESS STEEL APPLICATOR ROLL
5	WIRE WOUND ROD HOLDERS
6A	ALUMINUM BACKUP ROLL - AIR KNIFE
6B	AIR KNIFE
7A	HEATED CHROME LAMINATING ROLLS
7B	PNEUMATIC CYLINDER
8	1000 W. INFRA-RED UNITS
9	DRYING AID AIR TUBES
10	ADJUSTABLE ROLL - WEB CONTROL
11	SURFACE REWIND ASSEMBLY
12	CONTROL BOX
13	DRIVE MOTOR - ZERO-MAX VARIABLE SPEED
14A	NEOPRENE BACKUP ROLL - 3 ROLL REVERSE ROLL
14B	CHROME APPLICATOR ROLL - 3 ROLL REVERSE ROLL
14C	CHROME DOCTOR ROLL - 3 ROLL REVERSE ROLL

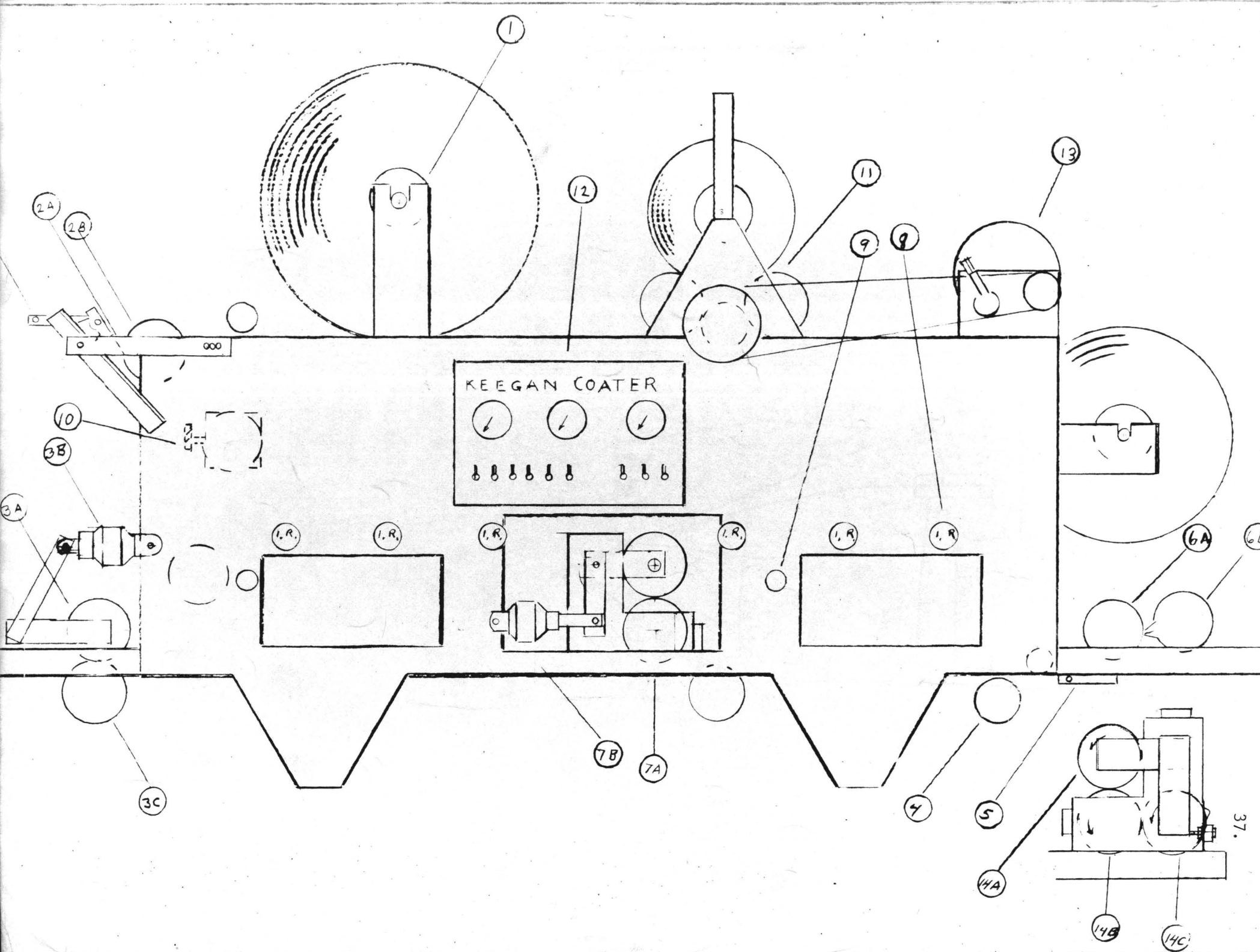
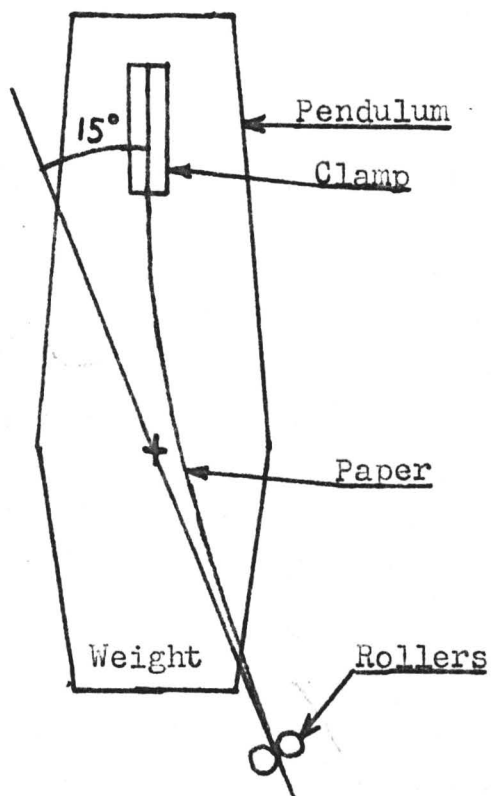


Figure 2

Schematic Drawing of Taber Stiffness Tester



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