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The effect of heat aging upon silicone rubber compounds

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THE EFFECT OF HEAT AGING UPON

SILICONE RUBBER COMPOUNDS

by

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ABSTRACT

Silicone rubber compounds are high temperature rubber-like materials consisting of silicone gum, a filler, and a curing catalyst. The filler is added to the rubber to give good molding properties and also good tensile strength and elongation. The data in this thesis shows that the compounds which had the best heat aging properties consisted of 100 parts of silicone rubber and 0.5 parts of benzoyl peroxide as the curing catalyst, with titanium dioxide as the filler.

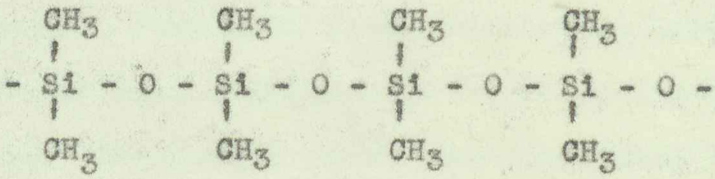
INTRODUCTION

Silicone rubber is a semi-organic synthetic with exceptionally good thermal stability. Due to this thermal stability, silicone rubber has found useful applications for such things as gaskets in turbosuperchargers and searchlights. Silicones have also been used to develop better electrical insulation to raise higher the operating efficiency of electrical apparatus. Other uses are as impregnants and binders. (1)

The silicone structure consists of long chains of alternate silicon and oxygen atoms. Organic groups as methyl, ethyl, vinyl, etc. are attached to the silicon atoms. Silicone rubber owes its thermal stability to the long chains of alternate silicon and oxygen atoms. Two methyl groups attached to each silicon atom impart flexibility. (2)

The original work on silicones was done by Frederick S. Kipping at the University of Nottingham, England. The rubber is made from dimethyldichlorosilane, which is prepared by either reaching methylmagnesium chloride with silicon tetrachloride to form a mixture of methylsilicon chlorides or by reacting elementary silicon directly with methyl chloride using a copper catalyst to form a mixture of methylchlorosilanes. Dimethyldichlorosilane is treated with water to form an oil. In this step the chlorine atoms in the molecules are

replaced with hydroxyl groups which react with one another to form the polysiloxane chain or methyl silicone polymer. The oil is then polymerized to form a long chain-high molecular weight gum (or rubber). This gum may be compounded by milling with fillers and curing agents, and cured to form a synthetic rubber-like material. The theories explaining the mechanism of the curing of silicone rubber state that both oxygen linkages and ethylenic bridges are formed between chains. The following diagrams show the probable silicone structure before and after curing:



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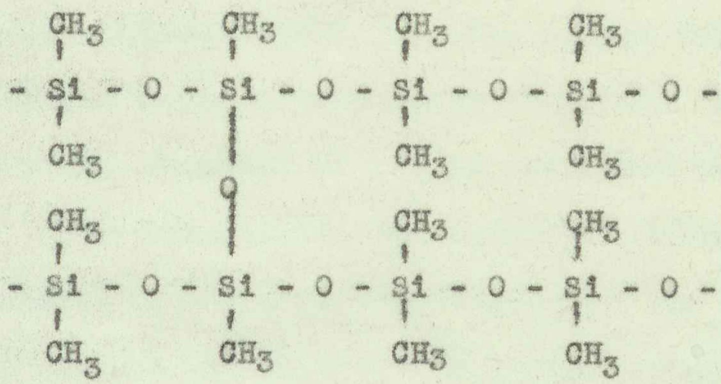

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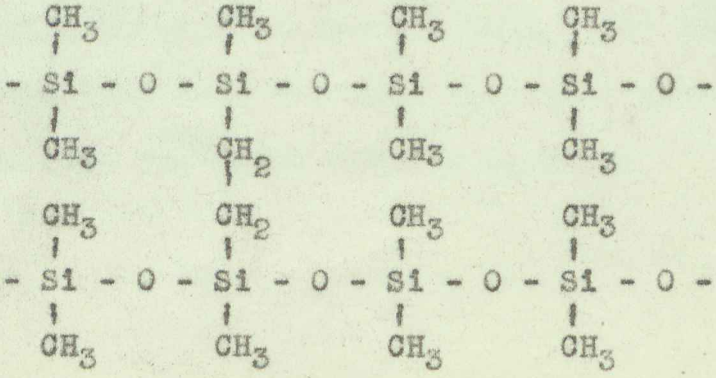


(cured)

When silicone rubber is aged at elevated temperatures some of the gum is converted to volatile cyclic trimers and tetramers.(3) The weight loss and flexibility of silicone rubber after heat aging are functions of the filler, other chemicals,(4) catalysts, and catalyst concentrations.(5) Silicone rubber compounds use many of the common rubber fillers, while benzoyl peroxide is commonly used as the curing catalyst, although other catalysts may also be used.

In addition to high thermal stability silicone rubber has low permanent set, good oil resistance, low tensile strength, good weathering, excellent dielectric properties, and low heat build-up due to internal friction. Chemically polar compounds such as alcohols, phenols, chlorinated compounds, and nitro compounds have minimum swelling action on the rubber, while hydrocarbons cause considerable swelling. Silicone rubber is rapidly attacked by strong acids and bases.(1)

In this thesis the aging of silicone rubber compounds at 200 C is described. It was the idea of the writer to investigate low benzoyl peroxide concentrations in order to determine whether improved heat aging properties would result. The compounds used before this work was done contained two percent to three percent (or more) benzoyl peroxide.

PURPOSE OF INVESTIGATION

The purpose of this investigation is to measure the life (maintenance of flexibility and rubber-like properties) and weight loss of silicone rubber molding compounds when aged at elevated temperatures. This information is needed in order to determine the correct formulas to use when a compound with long life at elevated temperatures is desired. Although silicone rubber does have excellent thermal stability, the long chains do slowly break down to form cyclic trimers and cyclic tetramers when held at sufficiently high temperatures. These trimers and tetramers are volatile, and their evaporation from the rubber may be measured by the weight loss of the rubber or a rubber compound. As the silicone rubber in a compound loses weight, gradual hardening and stiffening of the compound occurs. The weight loss and flexibility of the rubber compound after aging at elevated temperatures may be used as an indication of the expected life of the compound. Since the lower the weight loss, the greater is the flexibility, usually, and the longer the life as a rubber-like material. For this work 200 C has been chosen as the temperature at which the tests are conducted. However, a comparison between weight loss at 175 C and 200 C has also been made for some compounds and this data is also included.

Several of the commonly used fillers for silicone rubber were used with two different catalysts. The amounts of the fillers and the concentrations of the catalysts were varied in the different compounds tested in order to measure the effect of the amount of the filler as well as the effect of the catalyst concentration upon heat aging at 200 C.

Dielectric constant, power factor, hardness, tensile strength, and elongation measurements were also made on some of the compounds prepared for heat aging tests, and this data is also included in this thesis.

The first part of this thesis compares mainly the weight loss of compounds using different fillers when the benzoyl peroxide concentration is in the 2.0-3.0 percent range (based on silicone gum). The following sections describe the test data for low benzoyl peroxide concentrations, followed by a discussion of zirconyl nitrate [6] as a catalyst and a comparison of benzoyl peroxide and zirconyl nitrate catalysts. All weight loss graphs show weight loss data in percent based on the weight of silicone gum originally present in the compounds. The molding temperatures of all the compounds was 150 C. Compounds containing no catalyst, or cured with benzoyl peroxide, were molded for ten minutes. Compounds cured with zirconyl nitrate were molded for twenty minutes.

APPARATUS

Milling Rolls:

The silicone gum, fillers, other chemicals, and catalysts were mixed on milling rolls. After a band or sheet of rubber was formed around the rolls, the filler was added. Any other chemicals may be added with the filler or afterwards. The during catalyst was added last.

Mold:

Sheets of silicone rubber compounds can be molded in any suitable mold. For these tests the sheets were molded 3 in. by 4 in. in area and between 90 mils and 100 mils thick. The molding temperature was 150 C.

Laboratory Balance:

"Chainomatic" type laboratory balance

Laboratory Ovens:

Heat aging was performed by suspending the sheets in a 175 C or 200 C circulating air laboratory oven.

Tensile Tester:

Scott tensile testing machine.

Power Factor Bridge:

General Electric Bridge

Shore "A" Durometer

Used for hardness tests

METHODS OF TESTING

Weight Loss:

The sheets were weighed to the nearest one ten-thousandth of a gram. Weights were taken after the compound was cured, and periodically after aging at 175 C and 200 C. Weight losses are expressed in the weight loss data section both on the basis of the total weight of the compounded sheets, and on the basis of the silicone gum originally present in the compounds. The graphs show weight losses based only on the original silicone gum present in the compounds, since this is the most useful method for comparing the amounts of gum lost between different compounds.

Flexibility:

Flexibility tests were conducted by bending the sheets at the edge back upon themselves and creasing flat by pressing tightly between the forefinger and thumb. If a sheet did not crack, the flexibility was described as excellent. If a

crack occurred, the limit of flexibility was arbitrarily (for this investigation) said to have been reached and a broken line was drawn on some of the weight loss curves (when indicated on the graph) to show that cracking occurred by this test. On the solid lines on the graphs thus marked, no cracking occurred by this test.

Tensile Strength and Elongation:

Tensile strength and elongation tests were made on a Scott tensile testing machine.

Power Factor and Dielectric Constant:

Power factor and dielectric constant measurements were made on General Electric equipment at 25 C and 500 cycles.

Hardness:

Hardness measurements made using a Shore "A" Durometer.

MATERIALS

Silicone Gum: Standard General Electric Silicone gum (not compounded)

Fillers: Ferric oxide (red), celite (mainly diatomaceous silica), zinc sulfide, and titanium dioxide

Other Chemicals: Antimony trioxide, lead oxide (PbO), mercuric oxide (HgO), and boric acid

Catalysts: Benzoyl peroxide and zirconyl nitrate - $ZrO(NO_3)_2 \cdot 3H_2O$

PREPARATION OF COMPOUNDED RUBBER

The silicone rubber compounds used for this thesis were mixed from the several components by the writer on laboratory milling rolls.

DISCUSSION

I. Celite and Ferric Oxide as Fillers for Silicone Rubber. Comparison of Weight Losses at 175 C and 200 C.

For celite filled compound No. 1 the weight loss at 200 C was considerably greater than at 175 C after aging up to 83 days in a circulating air oven. The percent of rubber lost versus time for this compound is shown on Graph I. It is seen that the weight loss for this compound follows practically a straight line for the first 50 days and then levels off slightly. By the flexibility test both the samples aged at 175 C and the samples aged at 200 C cracked after less than ten days. The formula for the compound is given below. Small amounts of antimony trioxide were formerly added to some silicone rubber compounds, presumably to help to neutralize the catalyst used in making the gum. It was later found that celite filled compounds without antimony trioxide were no better or worse than compounds with antimony trioxide.

<u>Compound No. 1</u>

100	Silicone gum
135	Celite
3	Antimony trioxide
2.5	Benzoyl peroxide

A similar test comparing the weight loss at 175 C

and 200 C was made for a ferric oxide filled compound. Silicone rubber compounds can easily take higher amounts by weight of ferric oxide than celite because of the lower bulk factor of ferric oxide. It is to be expected that the weight loss at 175 C would be lower than at 200 C in the same length of time because the rate of forming volatile trimers and tetramers from the silicone gum would probably be lower at 175 C. This was the case for celite filled compound No. 1, but for ferric oxide filled compound No. 2, as given below, the weight loss at 175 C was practically equivalent to the weight loss at 200 C for the first 25 days and thereafter exceeded the weight loss at 200 C. The smaller weight loss at 200 C than at 175 C may possibly be due to oxidation of the silicone gum at 200 C when ferric oxide is used as the filler. The data for compound No. 2 are given on Graph II. This compound also cracks after only a few days at 175 C or 200 C by the flexibility test.

Compound No. 2

100	Silicone gum
250	Ferric oxide
2	Benzoyl peroxide

Examination of the data and curves for compounds Nos. 1 and 2 shows that celite filled compounds have much better heat aging properties from the standpoint

of weight loss than ferric oxide filled compounds.

II. Effect of Lead Oxide (PbO) and Mercuric Oxide (HgO) upon Celite Filled Silicone Rubber

In order to determine whether the addition of small amounts of lead oxide (PbO) is beneficial to the heat aging properties of silicone rubber, celite filled compounds were made with 1 percent and 2 percent PbO added, based on the gum. Compound No. 3 with 1 percent PbO and compound No. 4 with 2 percent PbO are compared to compound No. 1 on Graph III. From this information it is seen that 1 percent PbO has little effect upon the weight loss after 83 days at 200 C, but 2 percent PbO increases the weight loss by almost 20 percent after aging for 83 days. This indicates that little or nothing is to be gained by adding PbO to silicone rubber compounds, or at least to celite filled compounds. A sheet of compound No. 5, which is similar to compounds Nos. 3 and 4, except that it contains 5 percent PbO, became brittle so quickly at 200 C that weight checks were not made, since this compound would not be suitable for high temperature (200 C) use.

Compound No. 3

100	Silicone gum
135	Celite
1	PbO
3	Antimony trioxide
2.5	Benzoyl peroxide

Compound No. 4

100	Silicone gum
135	Celite
22	PbO
3	Antimony trioxide
2.5	Benzoyl peroxide

Compound No. 5

100	Silicone gum
135	Celite
5	PbO
3	Antimony trioxide
2.5	Benzoyl peroxide

If 2.4 percent of mercuric oxide is added to a celite filled silicone rubber compound replacing the antimony trioxide used in compound No. 1, although the initial weight loss is higher, a lower weight loss results after aging for about 70 days at 200 C. This is shown on Graph IV comparing compounds Nos. 1 and 6. However, even though mercuric oxide seems slightly beneficial, it is not permitted to be used in many factories because of its toxic and poisonous properties and, therefore, no further tests using mercuric oxide were made.

Compound No. 6

100	Silicone gum
150	Celite
2.4	Mercuric oxide
2.5	Benzoyl peroxide

III. Zinc Sulfide and Titanium Dioxide Fillers

A compound made using zinc sulfide (cpd. No. 7) as the filler lost more weight than celite compound No. 1 in the same length of time at 200 C, but the zinc sulfide filled compound passed the flexibility test after 20 days at 200 C, while the celite filled compound failed in less than 10 days, even though the celite filled compound contained less filler by weight than compound No. 7. These results are shown on Graph V. Although the weight loss of the gum differs only a little between the two compounds, zinc sulfide makes more useful silicone rubber molding compounds for use at high temperatures than celite filled compounds, not only because they retain their rubber-like properties longer, but also because they have better elongation and tensile strength and may, therefore, be removed from molds easier without tearing. (3)

Compound No. 7

100	Silicone gum
200	Zinc sulfide
2	Benzoyl peroxide

In titanium dioxide filled compounds the weight loss is lower than when celite, ferric oxide, or zinc sulfide are used as fillers. The flexibility test also shows titanium dioxide to be superior to all the other fillers tested. Titanium dioxide filled compound No. 8

is compared in weight loss and flexibility on Graph V to celite (No. 1), ferric oxide (No. 2), and zinc sulfide (No. 7) filled compounds.

Compound No. 8

100	Silicone gum
200	Titanium dioxide
2	Benzoyl peroxide

Graph V shows that titanium dioxide filled compounds are especially suitable as molding compounds for high temperature use, when compared to the other compounds, especially where shrinkage and other factors which are governed by weight loss are concerned. It might also be added that titanium dioxide filled compounds are considerably easier to mold than celite filled compounds, which are the second lowest in weight loss on Graph V.

IV. The Effect of Catalyst Concentration upon Titanium Dioxide Filled Compounds

Silicone rubber compounds Nos. 9, 10 and 11 were made up in order to determine the effect of benzoyl peroxide concentration upon the heat aging properties of titanium dioxide filled compounds. The weight loss and flexibility of these compounds is compared to compound No. 8, the formula for which has already been given.

Compound No. 9

100	Silicone gum
200	Titanium dioxide
1	Benzoyl peroxide

Compound No. 10

100 Silicone gum

200 Titanium dioxide

0.5 Benzoyl peroxide

Compound No. 11

100 Silicone gum

200 Titanium dioxide

0.25 Benzoyl peroxide

The data for compounds Nos. 8, 9, 10, and 11 are plotted on Graph VI (a). This graph shows the great effect of the benzoyl peroxide concentration upon the useful life of the titanium dioxide filled compounds. Furthermore, it shows that as the weight loss is decreased, the flexibility is increased. By the flexibility test compounds Nos. 8, 9, 10, and 11 will withstand bending without cracking for the maximum number of days listed in the following table.

Cpd. No.	% Benz. Peroxide	Max. days at 200 C without cracking
8	2.0	23
9	1.0	83
10	0.5	140
11	0.25	105
12	0	110

The above table indicates that for maximum flexi-

bility after aging at 200 C, 0.5 percent benzoyl peroxide should be used in compounds containing 100 parts of silicone gum to 200 parts of titanium dioxide. In addition, the minimum weight losses were also attained when the benzoyl peroxide concentration was 0.5 percent.

In a compound prepared as compound No. 12, using no catalyst, the weight loss after aging at 200 C for over 200 days is greater than if 0.5 percent or 1.0 percent benzoyl peroxide had been added, but is less than if 0.25 percent or 2.0 percent had been used. The flexibility of the compound in which no catalyst had been used is only about 110 days, however, so it is seen that nothing from the standpoint of weight loss or flexibility is to be gained by leaving out the curing catalyst entirely. On Graph VI (b) compound No. 12 is compared to compound No. 10, which had 0.5 percent benzoyl peroxide and was the best in flexibility and lowest in weight loss on Graph VI (a).

Compound No. 12

100	Silicone gum
200	Titanium dioxide
	no catalyst

V. The Effect of the Amount of Filler upon Weight Loss

The effect of a filler such as titanium dioxide upon the weight loss of the gum at 200 C may be deter-

mined by comparing compound No. 13 with compound No. 10. Both compounds contain 0.5 percent benzoyl peroxide, but compound No. 13 has only 100 parts of titanium dioxide while compound No. 10 has 200 parts of this filler to 100 parts of silicone gum. The data show that a smaller percentage of the gum is lost from the compounds after aging at 200 C when 100 parts of titanium dioxide is used than when 200 parts of the filler is used. This is shown by plotting the data on Graph VII.

Compound No. 13

100	Silicone gum
100	Titanium dioxide
0.5	Benzoyl peroxide

When no catalyst is used the same results as above are observed. The compound containing the lower amount of titanium dioxide has a lower weight loss based on the gum. This is shown on Graph VIII, which compares compound No. 14 to compound No. 12.

Compound No. 14

100	Silicone gum
100	Titanium dioxide
	no catalyst

In addition to the above two examples, silicone rubber compounds catalyzed with zirconyl nitrate also show a lower weight loss for the compound containing

the smaller amount of filler, as is shown later. Compounds containing less than 100 parts of titanium dioxide to 100 parts of gum were not tested, because a lower amount of this filler gives compounds with poor molding qualities.

Furthermore, the flexibility is also improved when measured by the flexibility test as the amount of filler is decreased. This is shown in the following table.

Cpd. No.	Parts TiO_2 to 100 parts gum	% Benzoyl peroxide	Max. days at 200 C without cracking
10	200	0.5	140
13	100	0.5	400 plus
12	200	0	110
14	100	0	400 plus

VI. Zirconyl Nitrate Catalyzed Compounds

Zirconyl nitrate was found by Mr. J. G. E. Wright (6) of the General Electric Research Laboratory to be a curing catalyst for silicone rubber. In testing zirconyl nitrate catalyzed compounds (2.0 percent zirconyl nitrate) the writer found that zirconyl nitrate cured compounds retain their flexibility much longer than similar compounds when 2.0 percent benzoyl peroxide is used as the curing catalyst. Also, as the

data show, the relative weight loss of zirconyl nitrate cured compounds is very low. However, it requires between fifteen and twenty minutes at 150 C to mold zirconyl nitrate cured compounds, while ten minutes is sufficient to mold compounds cured with benzoyl peroxide.

It was found that 2.0 percent zirconyl nitrate, based on the gum, is approximately the correct percentage to use, since a smaller amount would probably increase the molding time, while a larger amount often causes some puffing or blistering of the compound.

In comparing the weight losses of compounds containing celite and titanium dioxide as fillers, using zirconyl nitrate as the curing catalyst, it was found that the percentage of rubber lost upon heat aging at 200 C was less for the titanium dioxide filled compounds than for the celite filled compounds. This fact was also shown when benzoyl peroxide was used as the catalyst on Graph V. Compounds Nos. 15 and 16 show this same fact. The data are plotted on Graph IX. In comparing the weights of fillers in compounds Nos. 15 and 16 it must be remembered that celite has a higher bulk factor than titanium dioxide, so a smaller weight of celite is

compared to the larger weight of titanium dioxide in compound No. 16 for testing purposes. These compounds are made up to give approximately the same hardness after molding.

Compound No. 15

100	Silicone gum
135	Celite
2	Zirconyl nitrate

Compound No. 16

100	Silicone gum
200	Titanium dioxide
2	Zirconyl nitrate

By the flexibility test compound No. 16 did not fail until 53 days at 200 C, while the celite filled compound No. 15 failed in only 19 days. This is also shown on Graph IX.

Another comparison of the lower weight loss and greater flexibility after aging at 200 C for titanium dioxide filled compounds than for celite filled compounds is observed in comparing compounds Nos. 18 and 19, which are the same as compounds Nos. 15 and 16 except that 1.0 percent boric acid has been added to each compound. These tests also show that boric acid is not beneficial to the compounds, as was hoped, since it affects the weight loss but little, but lowers the flexibility after aging at 200 C for the titanium

dioxide filled compound. The data for compounds Nos. 17 and 18 are not plotted on a graph, but are listed in the weight loss data section of this thesis.

Compound No. 17

100	Silicone gum
135	Celite
1	Boric acid
2	Zirconyl nitrate

Compound No. 18

100	Silicone gum
200	Titanium dioxide
1	Boric acid
2	Zirconyl nitrate

In compounds Nos. 17 and 18 the flexibility test showed that compound No. 17 failed in 19 days, and compound No. 18 failed in 50 days, instead of the 110 days attained in compound No. 16 without the boric acid.

For benzoyl peroxide cured compounds it was seen that, using titanium dioxide as the filler, a lower amount of filler in the compound gives a lower weight loss of gum. This has also been found to be true using zirconyl nitrate as the curing catalyst, and may be observed by comparing compounds Nos. 16 and 19. The data for this comparison are plotted on Graph X.

Compound No. 19

100	Silicone gum
100	Titanium dioxide
2	Zirconyl nitrate

The flexibility test shows that compound No. 19 has not failed in over 400 days.

Compound No. 20 was catalyzed with both benzoyl peroxide and zirconyl nitrate. Both the flexibility and weight loss data show that nothing is to be gained by mixing the two catalysts. Compound No. 21 gives similar results. The data for these latter two compounds is not plotted, but is included in the weight loss data tables.

Compound No. 20

100	Silicone gum
200	Titanium dioxide
0.25	Benzoyl peroxide
1.5	Zirconyl nitrate

Compound No. 21

100	Silicone gum
100	Titanium dioxide
0.5	Benzoyl peroxide
1.5	Zirconyl nitrate

Compound No. 22 was made using zirconyl nitrate to cure a ferric oxide filled compound. Since this compound did not cure in the usual length of time, no heat aging tests were conducted.

Compound No. 22

100	Silicone gum
200	Ferric oxide
2	Zirconyl nitrate

VII. Hardness Tests

As silicone rubber compounds are aged at 200 C, the hardness increases as the compound loses weight, and as its flexibility decreases. An example of the hardness changes as measured using a Shore "A" Durometer before and after aging compounds Nos. 8, 9, 10, and 16 at 200 C may be seen on Graph XI. It is observed that when 0.5 percent or 1.0 percent benzoyl peroxide is used as the curing catalyst, the change in hardness is small. These two compounds had an initial hardness of about 30, and reached a hardness of only 58 to 60 after 133 days at 200 C. Compounds containing 2.0 percent benzoyl peroxide had an initial hardness of 30 and reached a hardness of 90 after the same aging, while compounds catalyzed with 2.0 percent zirconyl nitrate, also having an initial hardness of 30, attained a hardness of 77.

VIII. Power Factor and Dielectric Constant Data for Silicone Rubber Compounds

Although this thesis discusses the effect of heat aging at 200 C upon the physical properties of silicone rubber in detail, some of the electrical properties of the rubber are also included, since silicone rubber compounds may be used as electrical insulation. When measured at 60 cycles and 500 volts, celite filled compound No. 1 shows a

decrease in power factor and an increase in dielectric constant after aging for about 170 days at 200 C. Zinc sulfide compound No. 7 remains the same in power factor, but increases in dielectric constant after 150 days at 200 C. The values for power factor and dielectric constant data before and after aging are given in the following table.

Cpd. No.	Filler	Power Factor Unaged	Power Factor Aged	Dielectric Constant Unaged	Dielectric Constant Aged
1	Celite	.0076	.0020	3.4	7.0
7	ZnS	.0032	.0033	4.2	7.0

The power factor data for titanium dioxide filled compounds Nos. 8 and 16 show that the power factor and dielectric constant are lower for benzoyl peroxide catalyzed compound No. 8 than for zirconyl nitrate catalyzed compound No. 16. The power factor for ferric oxide filled compound No. 2 is quite high in comparison with the other silicone rubber compounds.

Cpd. No.	Filler	Catalyst	Power Factor Unaged	Diel. Const. Unaged
8	TiO_2	Benz. Per.	0.0037	6.3
16	TiO_2	Zirc. Nit.	0.0076	7.7
2	Ferric oxide	Benz. Per.	0.11	3.4

IX. Effect of Aging at 200 C upon Tensile Strength and Elongation

The effect of short time aging at 200 C upon the

tensile strength and elongation of silicone rubber compounds is as follows. Compound No. 8 shows that the tensile strength increases and elongation decreases during short time aging at 200 C.

Cpd. No.	Filler	PSI after 48 hrs at 200 C	PSI after 96 hrs at 200 C	% elong. after 48 hrs at 200 C	% elong. after 96 hrs at 200 C
8	TiO_2	270	345	210	200
2	Fer. Ox.	370		80	
1	Celite	415		95	

CONCLUSIONS

The conclusions concerning the heat aging of silicone rubber compounds derived from the date presented and discussed in this thesis and plotted on the graphs may be summarized as follows:

1. When comparing the weight loss and flexibility of silicone rubber compounds using ferric oxide, celite, zinc sulfide, and titanium dioxide as fillers and 2.0 - 2.5 percent benzoyl peroxide as the curing catalyst, titanium dioxide gives the lowest weight loss and best flexibility after aging at 200 C. Ferric oxide is the poorest of the four fillers, since it gives the highest weight loss and poorest flexibility after aging at 200 C.

2. The silicone gum weight loss is considerably greater at 200 C than at 175 C for celite filled compounds. For the ferric oxide filled compound tested, the weight losses were about equal for 28 days, then the loss at 175 C was somewhat greater than at 200 C.

3. Lead oxide (PbO) increases the weight loss of silicone gum when aged at 200 C in increasing amounts as the amount of lead oxide is increased.

4. Mercuric oxide affects the weight loss of celite filled compounds very little upon aging at 200 C.

5. As the concentration of benzoyl peroxide is decreased in titanium dioxide filled compounds, the weight loss decreases and the flexibility increases upon aging at 200 C. until 0.5 percent benzoyl peroxide is reached. Catalyzing below 0.5 percent with this catalyst gives increased weight loss and poorer flexibility.

6. Silicone rubber compounds containing equal parts of gum and titanium dioxide lose a smaller percentage of the rubber after aging at 200 C than compounds containing twice as much titanium dioxide as gum by weight.

7. Silicone rubber compounds cured with zirconyl nitrate have lower weight loss when aged at 200 C than identical compounds cured with benzoyl peroxide.

8. The flexibility of zirconyl nitrate cured compounds (200 parts of titanium dioxide to 100 parts of gum) is better than identical compounds cured with 2.0 percent benzoyl peroxide, but poorer than compounds cured with 1.0 percent, 0.5 percent, and 0.25 percent benzoyl peroxide.

9. The flexibility of silicone rubber compounds improves upon aging at 200 C as the amount of filler to gum is decreased in the compounds, regardless of the

filler used.

10. Lower weight loss or better flexibility after aging at 200 C are not attained by mixing benzoyl peroxide and zirconyl nitrate curing catalysts.

11. As the weight loss of gum increases in a compound, the flexibility becomes poorer and the hardness increases. These changes in flexibility and hardness depend upon the filler and curing catalysts.

12. When measured at 60 cycles and 500 volts, the power factor of a celite filled compound decreases after aging at 200 C, and the dielectric constant increases. For a zinc sulfide filled compound the power factor remains the same, but the dielectric constant increases after aging at 200 C.

13. Aging at 200 C improves the tensile strength of silicone rubber compounds, but decreases the elongation. (5)

WEIGHT LOSS DATA

Cpd.	% Gum in Cpd.	Days Aged	% Wt. Loss of Cpd. 175 C	200 C	% Gum Loss 175 C	200 C
1	41.6	3	0.4	0.6	0.9	1.4
		10	1.4	1.9	3.4	4.6
		19	2.1	3.9	5.1	9.4
		21	2.4	4.7	5.8	11.3
		38	4.0	8.7	9.6	21.0
		50	4.8	11.8	11.5	28.4
		83	6.4	16.1	15.4	38.8
2	28.4	3	1.3	1.7	4.6	6.0
		10	5.8	6.0	20.4	21.1
		19	8.8	9.4	31.0	33.1
		21	10.5	11.0	37.0	38.8
		38	15.8	13.7	55.6	48.3
		50	17.6	14.6	62.0	51.5
		83	20.2	15.5	71.2	54.6
3	41.5	3		4.4		10.6
		6		6.6		15.8
		25		8.8		21.1
		38		10.9		26.2
		50		11.1		26.7
		67		12.3		29.6
		83		13.6		32.7

Cpd.	% Gum in Cpd.	Days at 200 C	% Wt. Loss of Cpd.	% Gum Loss
4	41.3	15	7.2	17.4
		30	11.8	29.6
		42	14.4	35.0
		58	18.1	43.8
		83	23.1	56.0
6	39.2	15	6.8	17.3
		30	9.6	24.4
		42	11.6	29.5
		105	15.0	38.1
7	33.2	17	6.2	18.6
		30	9.2	27.7
		42	11.4	34.4
		58	13.1	39.5
		75	14.8	44.6
		105	16.6	50.0
8	33.1	4	4.8	14.5
		13	5.8	17.5
		25	7.5	22.6
		42	8.7	26.3
		58	9.7	29.3
		83	10.6	32.0
		105	11.4	34.5

Cpd.	% Gum in Cpd.	Days at 200 C	% Wt. Loss of Cpd.	% Gum Loss
9	33.2	9	3.6	10.8
		30	5.1	15.4
		42	5.7	17.2
		58	6.0	18.0
		75	6.6	19.9
		125	7.9	23.8
		145	8.4	25.3
		165	9.2	27.7
		210	10.6	32.0
		220	11.2	33.8
		250	12.4	37.4
10	33.3	9	3.6	10.8
		30	5.1	15.6
		42	5.6	16.8
		58	5.9	17.7
		75	6.5	19.5
		125	7.7	23.1
		145	8.4	25.2
		165	8.9	26.7
		210	10.7	32.0
		220	10.8	32.4
		250	11.9	35.8

Cpd.	% Gum in Cpd.	Days at 200 C	% Wt. Loss of Cpd.	% Gum Loss
11	33.3	10	4.2	12.6
		25	5.5	16.5
		50	6.4	19.2
		67	7.5	22.5
		105	8.7	26.1
		125	9.8	29.4
		145	11.0	33.0
12	33.3	65	4.5	13.6
		110	5.8	17.5
		210	12.1	36.5
13	50.0	65	6.5	13.0
		110	8.7	17.4
		210	9.6	19.2
14	50.0	65	4.8	9.6
		110	6.4	12.8
		210	9.0	19.0
15	42.2	4	4.9	11.6
		10	5.5	13.0
		19	5.9	14.0
		25	6.2	14.7
		33	6.5	15.4
		50	7.2	17.0
		67	7.5	17.8
		105	8.3	19.7
		125	8.7	20.6

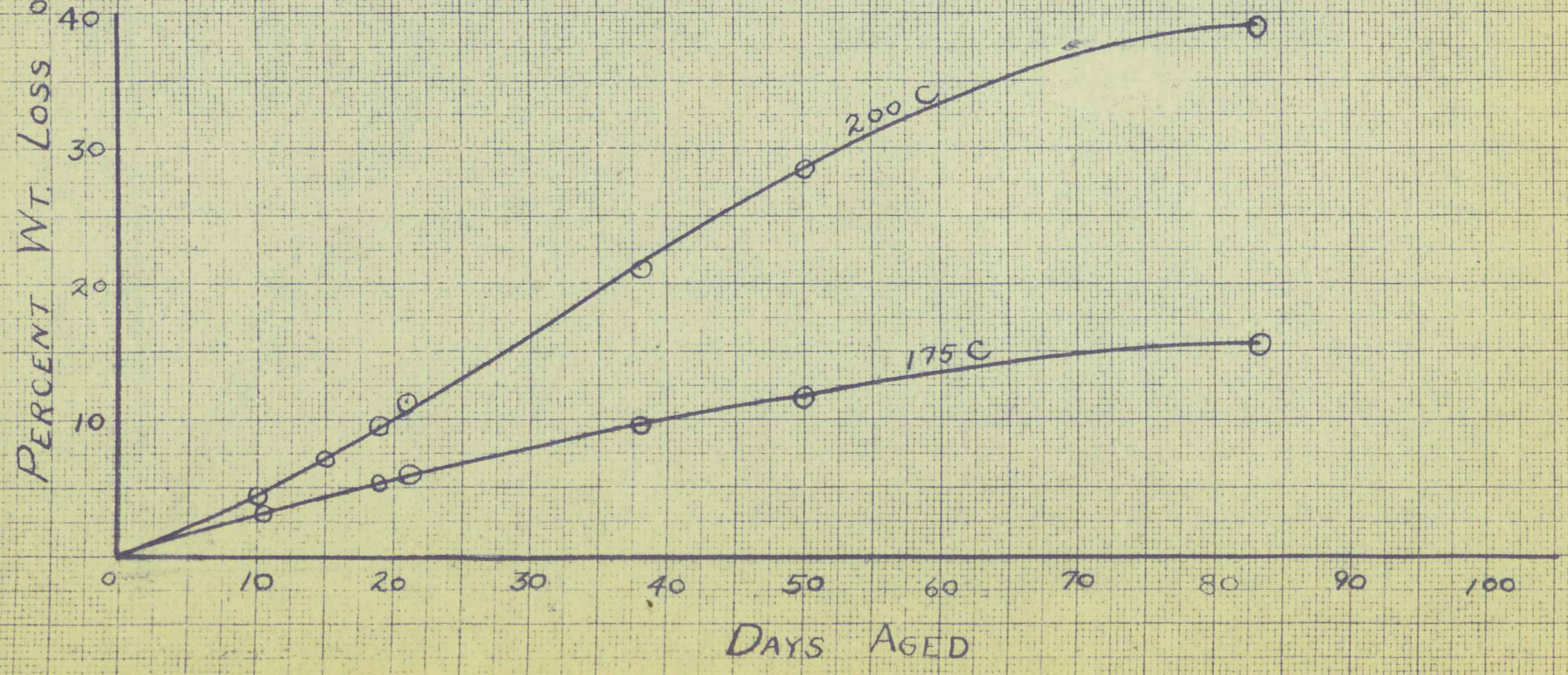
Cpd.	% Gum in Cpd.	Days at 200 C	% Wt. Loss of Cpd.	% Gum Loss
15 (Cont'd.)		145	9.1	21.6
		165	9.7	23.0
		210	10.2	24.2
16	33.1	65	4.3	13.0
		110	5.4	16.3
		210	7.2	21.7
17	42.0	4	5.0	11.9
		10	5.6	13.3
		19	6.2	14.8
		25	6.6	15.7
		50	7.7	18.3
		67	8.2	19.5
		105	9.0	21.4
		145	9.6	22.8
		210	10.7	25.5
		250	12.0	28.6
18	33.0	4	3.0	9.1
		10	3.4	10.3
		19	3.7	11.2
		25	4.0	12.1
		50	4.6	14.0
		67	4.9	14.8
		105	5.4	16.4
		145	5.8	17.6
		210	6.6	20.0

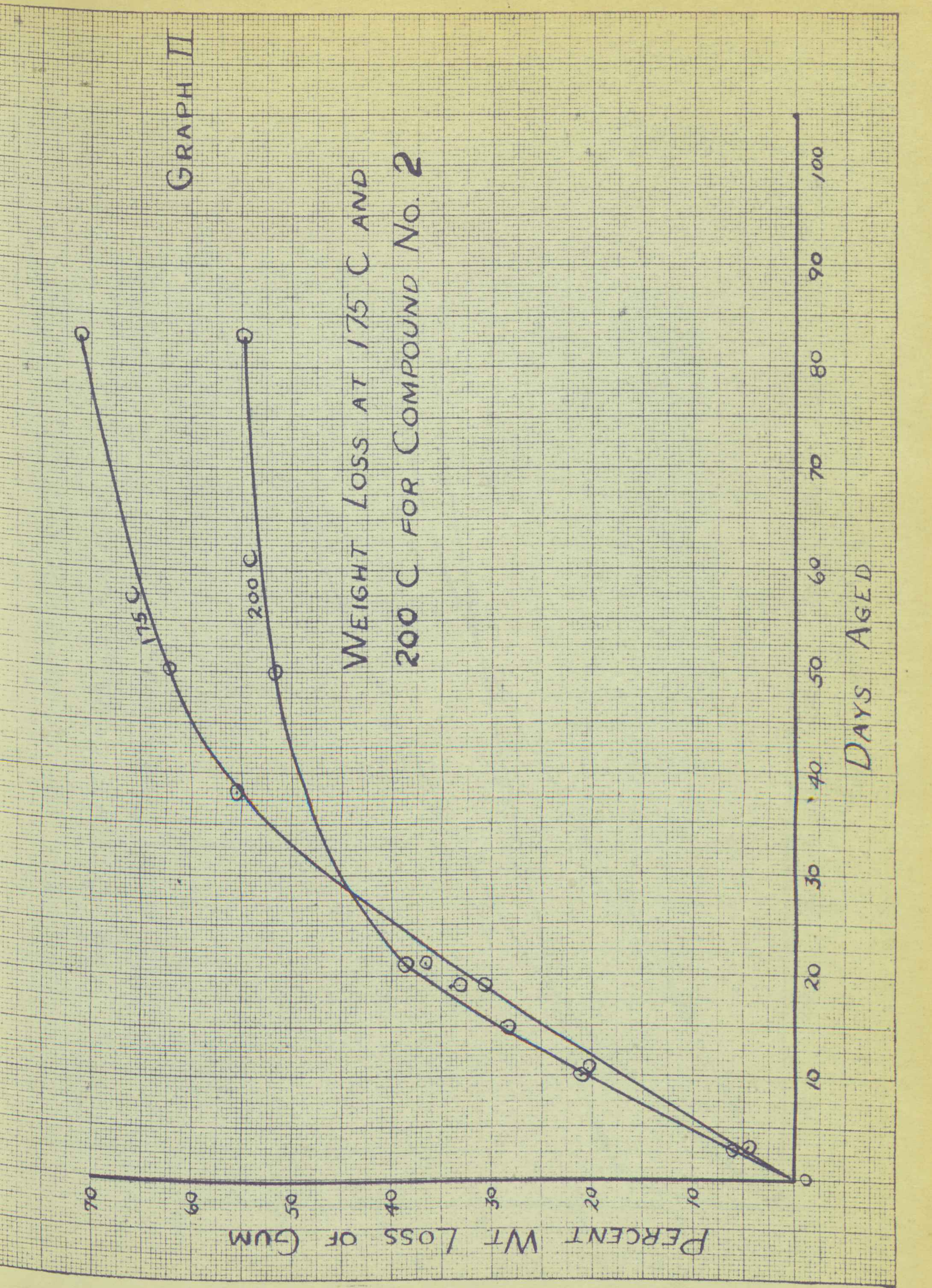
Cpd.	% Gum in Cpd.	Days at 200 C	% Wt. Loss of Cpd.	% Gum Loss
18 (Cont'd.)		250	7.4	22.4
19	49.5	65	4.6	9.3
		110	5.8	11.7
		210	7.8	15.8
20	33.1	65	5.2	15.7
		110	6.8	20.5
		210	9.5	28.6
21	49.5	65	5.8	11.7
		110	7.3	14.7
		210	7.6	15.3

Weight Loss at 175 C and 200 C

for Compound No. 1

Graph I

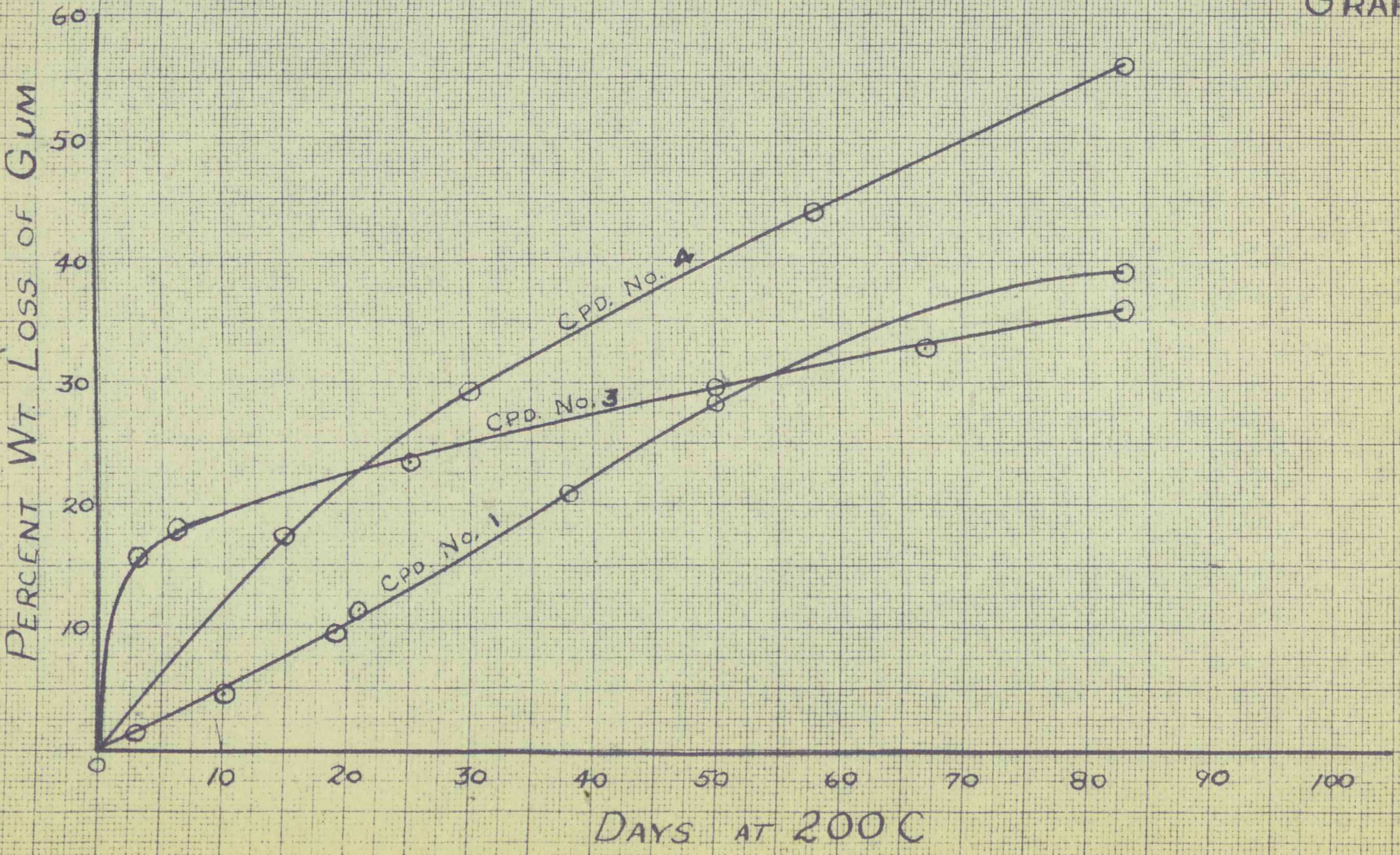


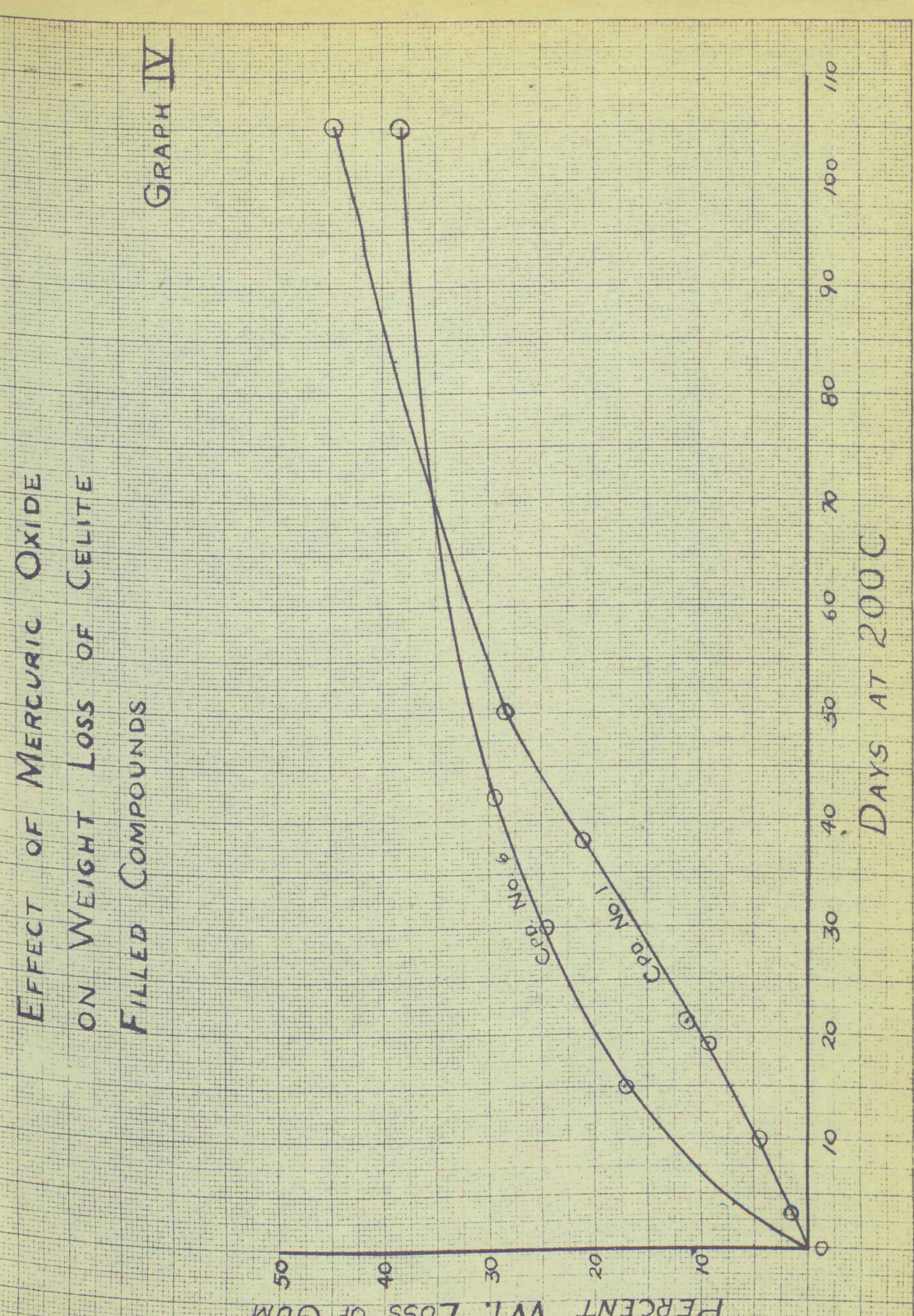


GRAPH II
WEIGHT LOSS AT 175 C AND
200 C FOR COMPOUND NO. 2
175 C
200 C
DAYS AGED
PERCENT WT LOSS OF GUM
0
10
20
30
40
50
60
70
80
90
100

EFFECT OF LEAD OXIDE (PbO) ON WEIGHT LOSS OF CELITE FILLED COMPOUNDS

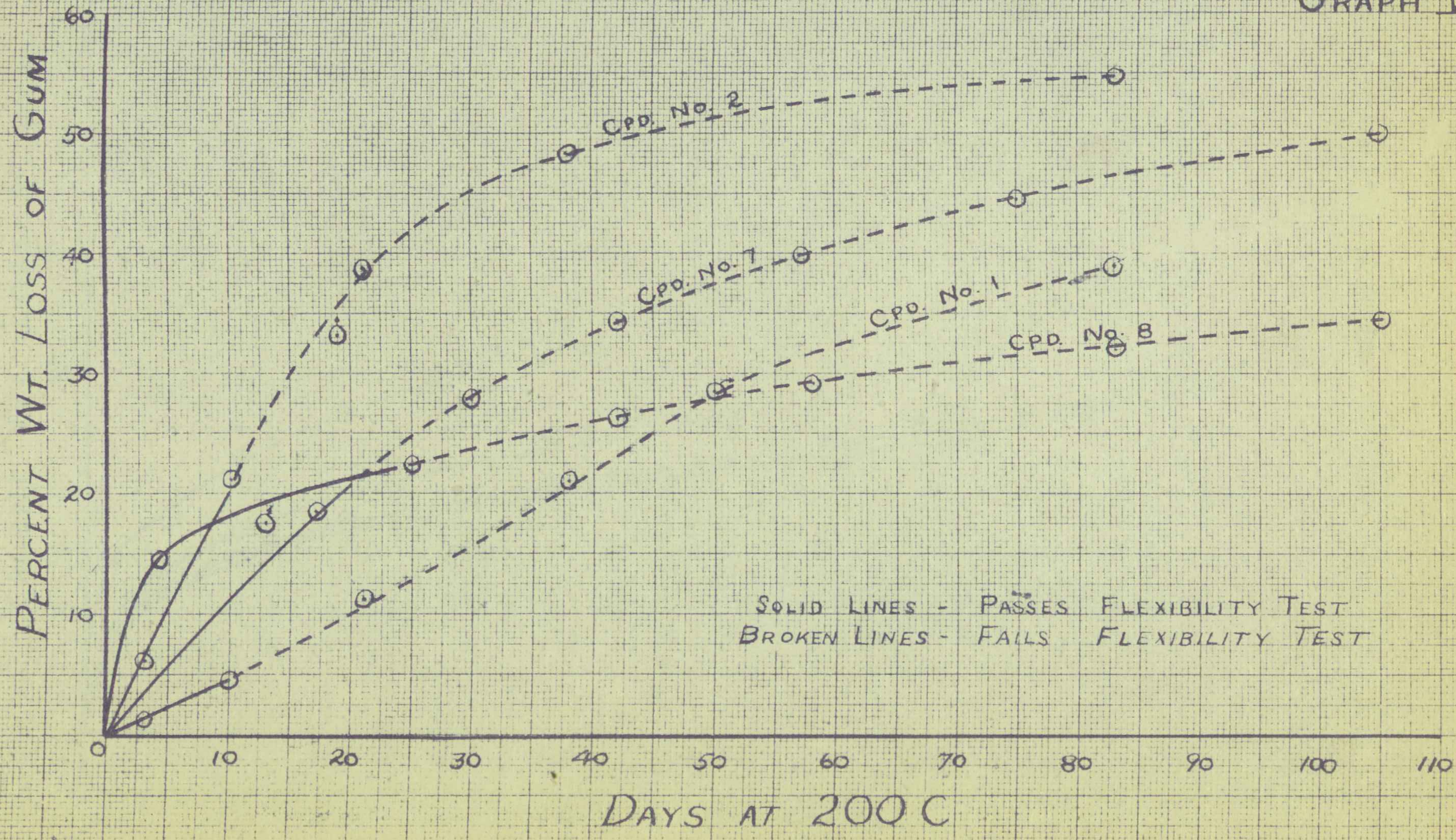
GRAPH III




Effect of Mercuric Oxide
on Weight Loss of Celite
Filled Compounds
Graph IV
Cpd. No. 6
Cpd. No. 1
Days at 200C
Percent Wt. Loss of Gum
0
10
20
30
40
50
60
70
80
90
100
110

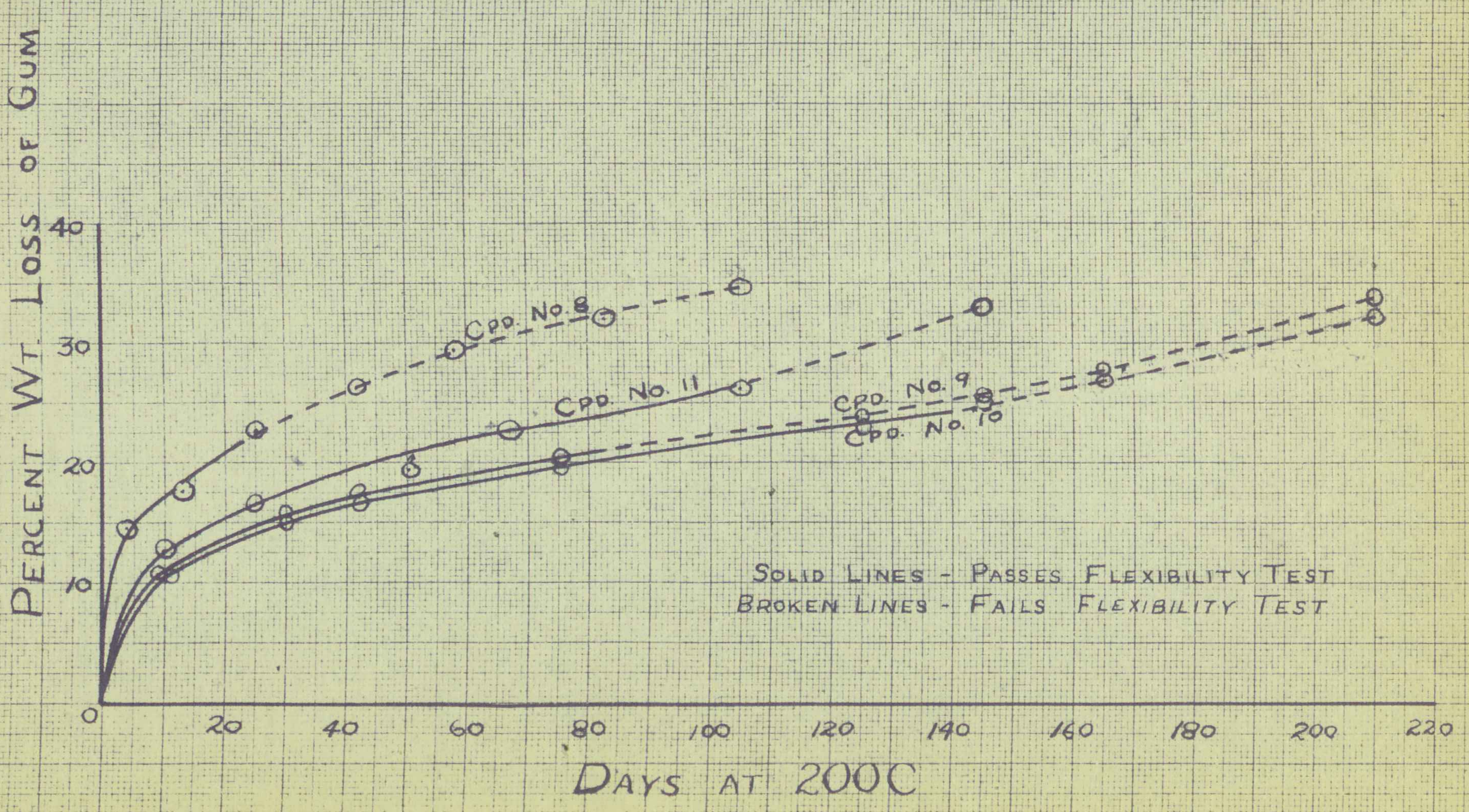
Effect of Different Fillers upon Weight Loss and Flexibility of Silicone Rubber Compounds

Graph V



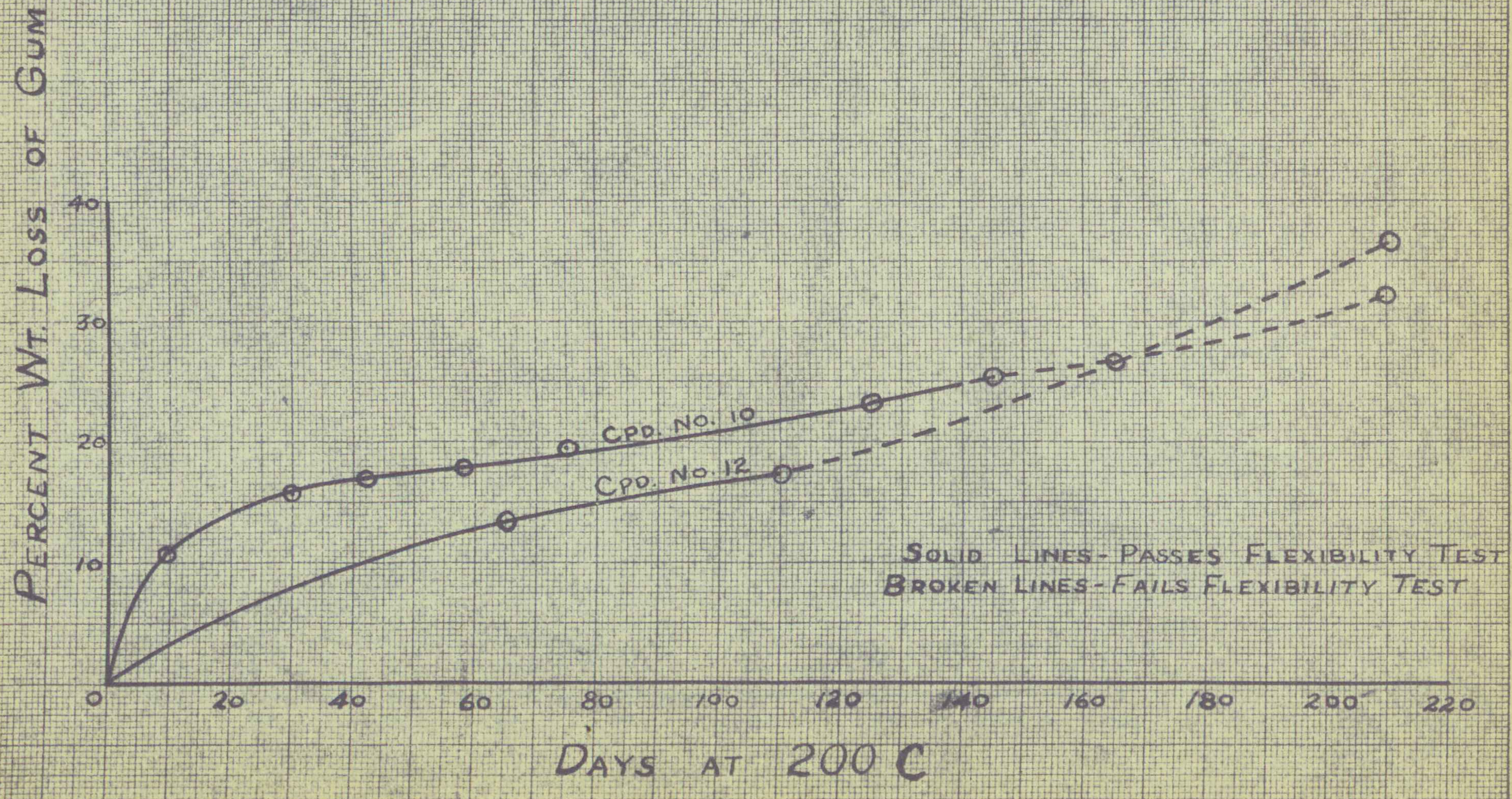
EFFECT OF BENZOYL PEROXIDE CONCENTRATION UPON WEIGHT LOSS AND FLEXIBILITY OF TiO_2 FILLED COMPOUNDS

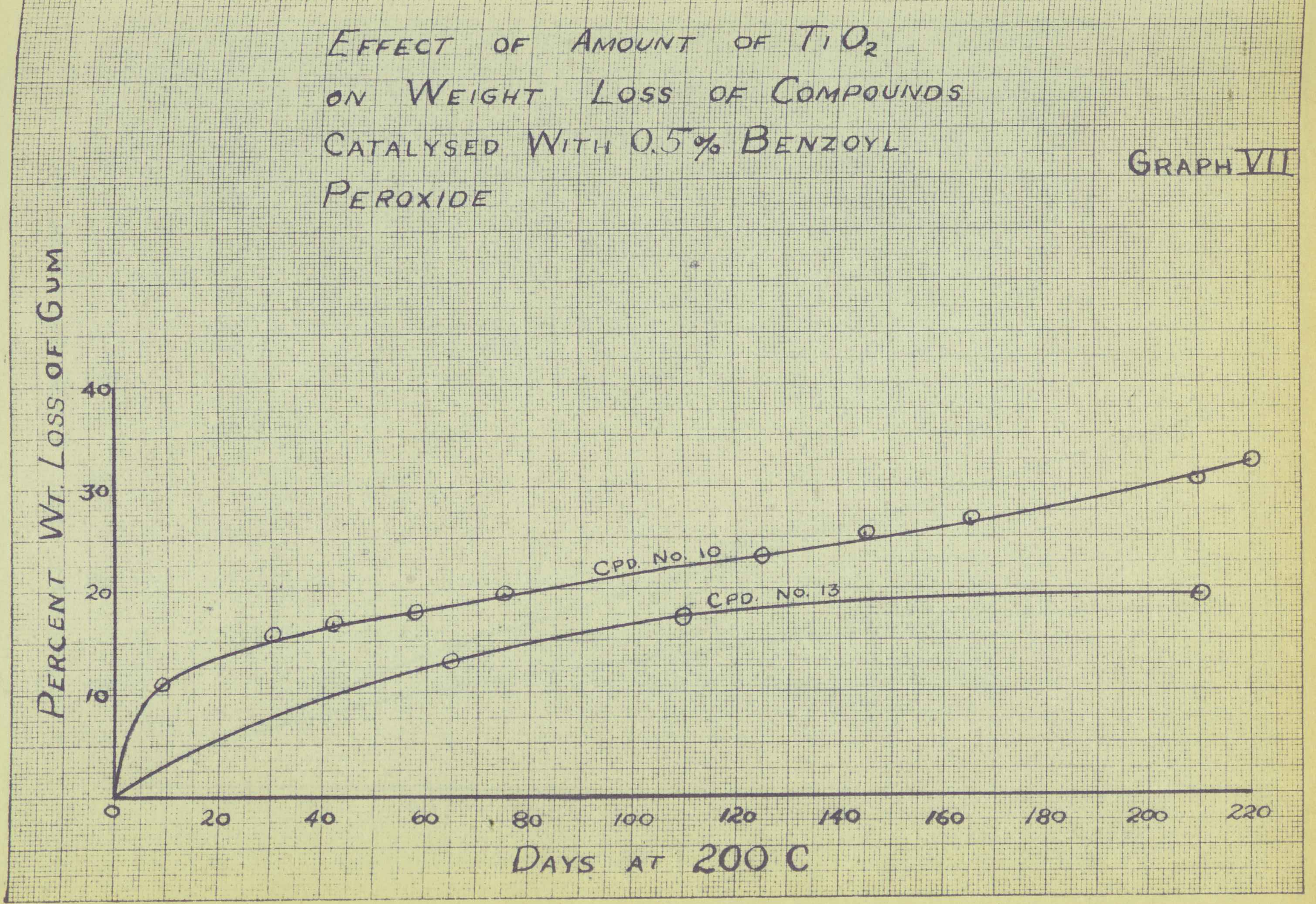
GRAPH VI(a)



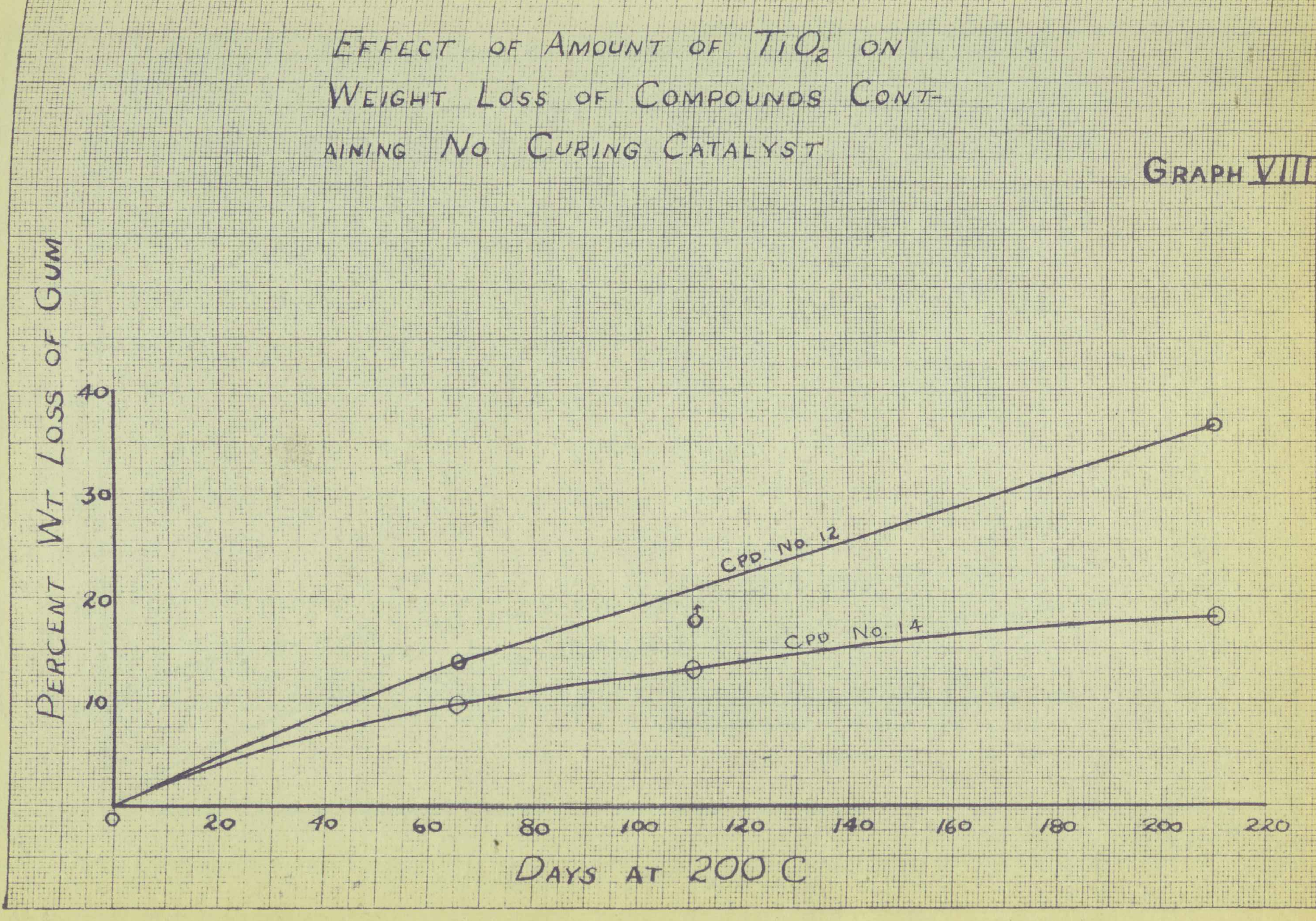
Comparison of Best Compound (0.5% Benzoyl Peroxide) on Graph VI (a) to Similar Compound Containing No Catalyst

Graph VI(b)



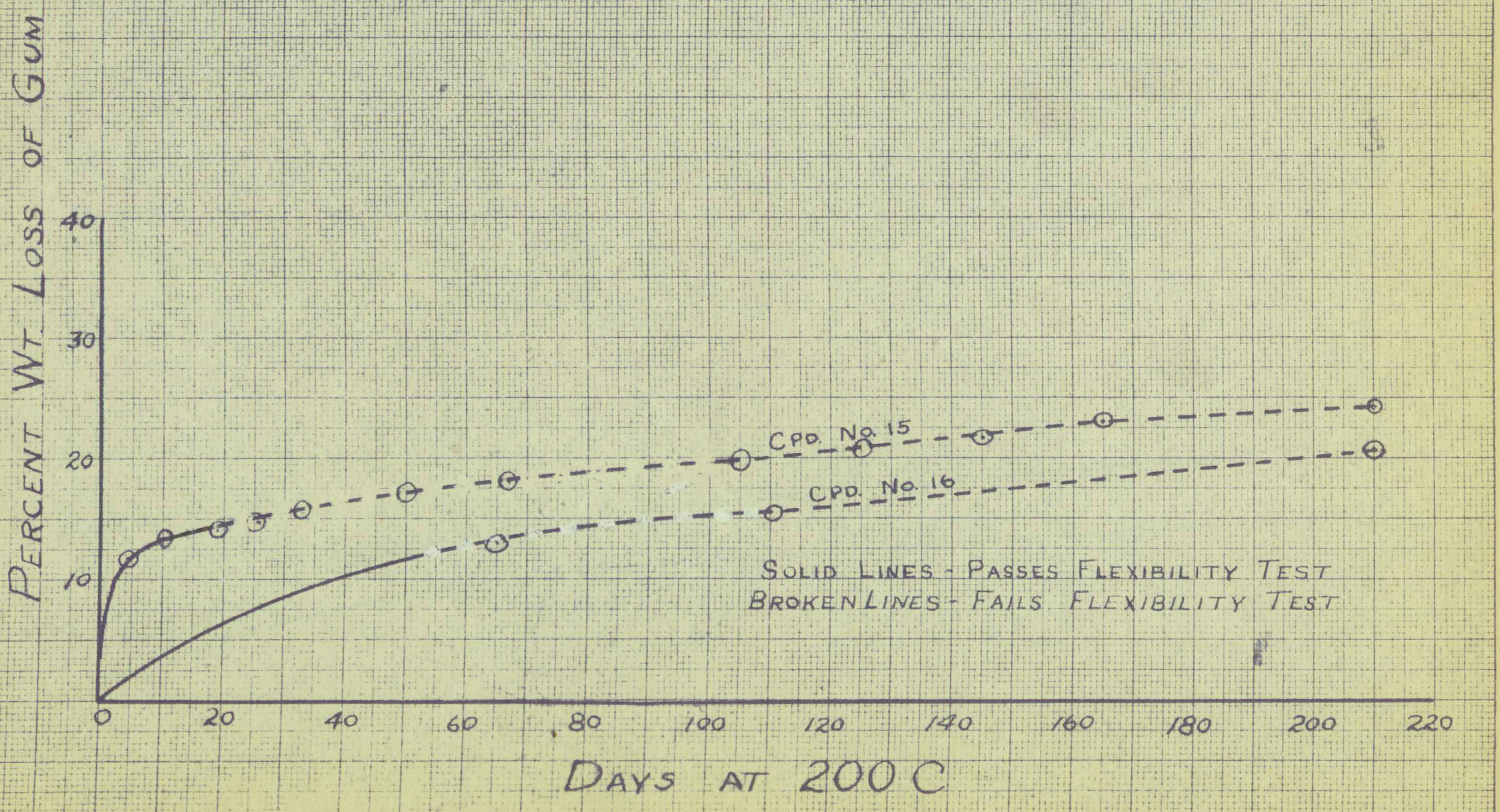


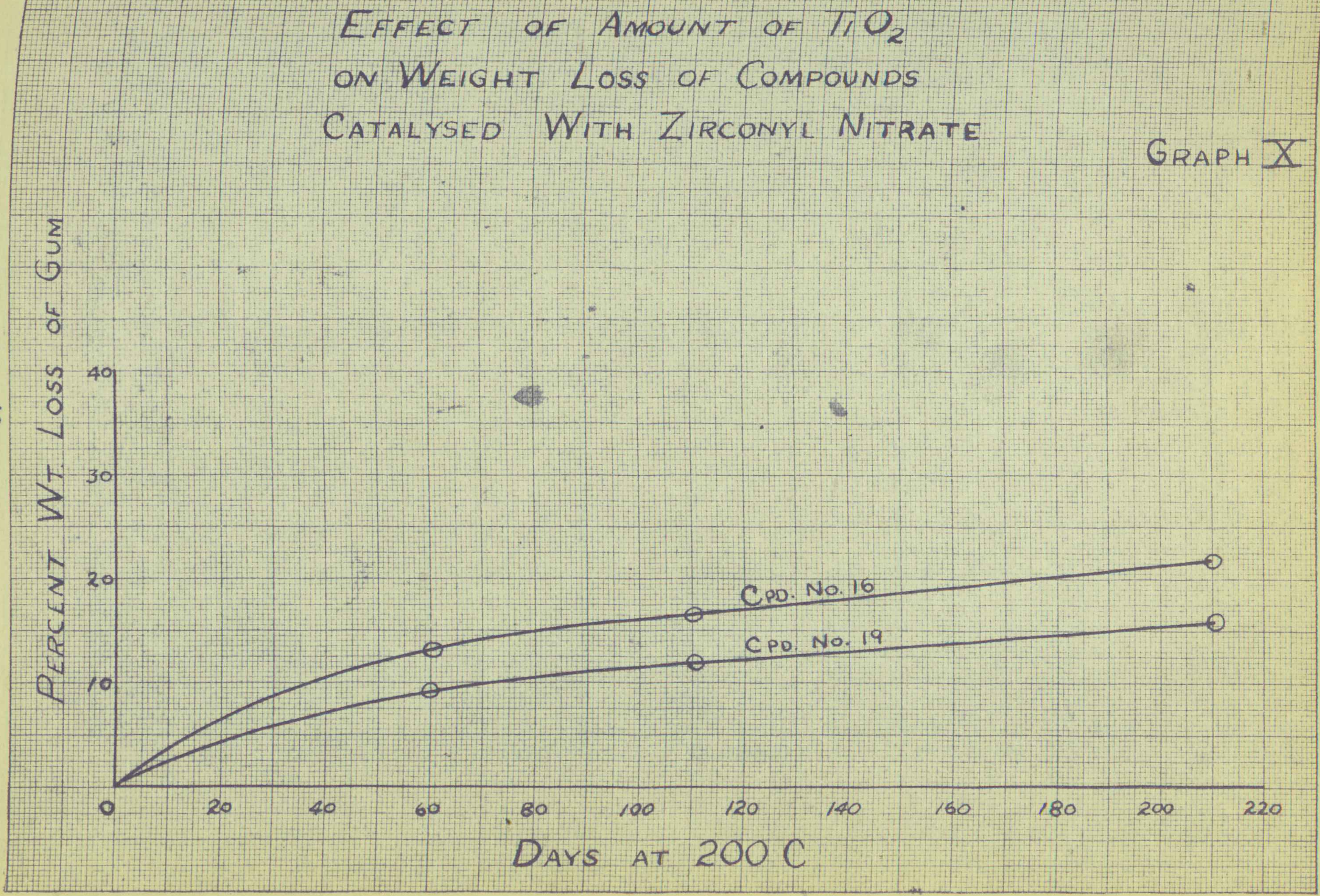
EFFECT OF AMOUNT OF TiO_2
ON WEIGHT LOSS OF COMPOUNDS
CATALYSED WITH 0.5% BENZOYL
PEROXIDE
GRAPH VII
PERCENT WT. LOSS OF GUM
40
30
20
10
0
20
40
60
80
100
120
140
160
180
200
220
CPD. NO. 10
CPD. NO. 13
DAYS AT 200 C


EFFECT OF AMOUNT OF TiO_2 ON
WEIGHT LOSS OF COMPOUNDS CONT-
AINING NO CURING CATALYST
GRAPH VIII
PERCENT WT. LOSS OF GUM
40
30
20
10
0
20
40
60
80
100
120
140
160
180
200
220
DAYS AT 200 C
CPD. No. 12
CPD. No. 14

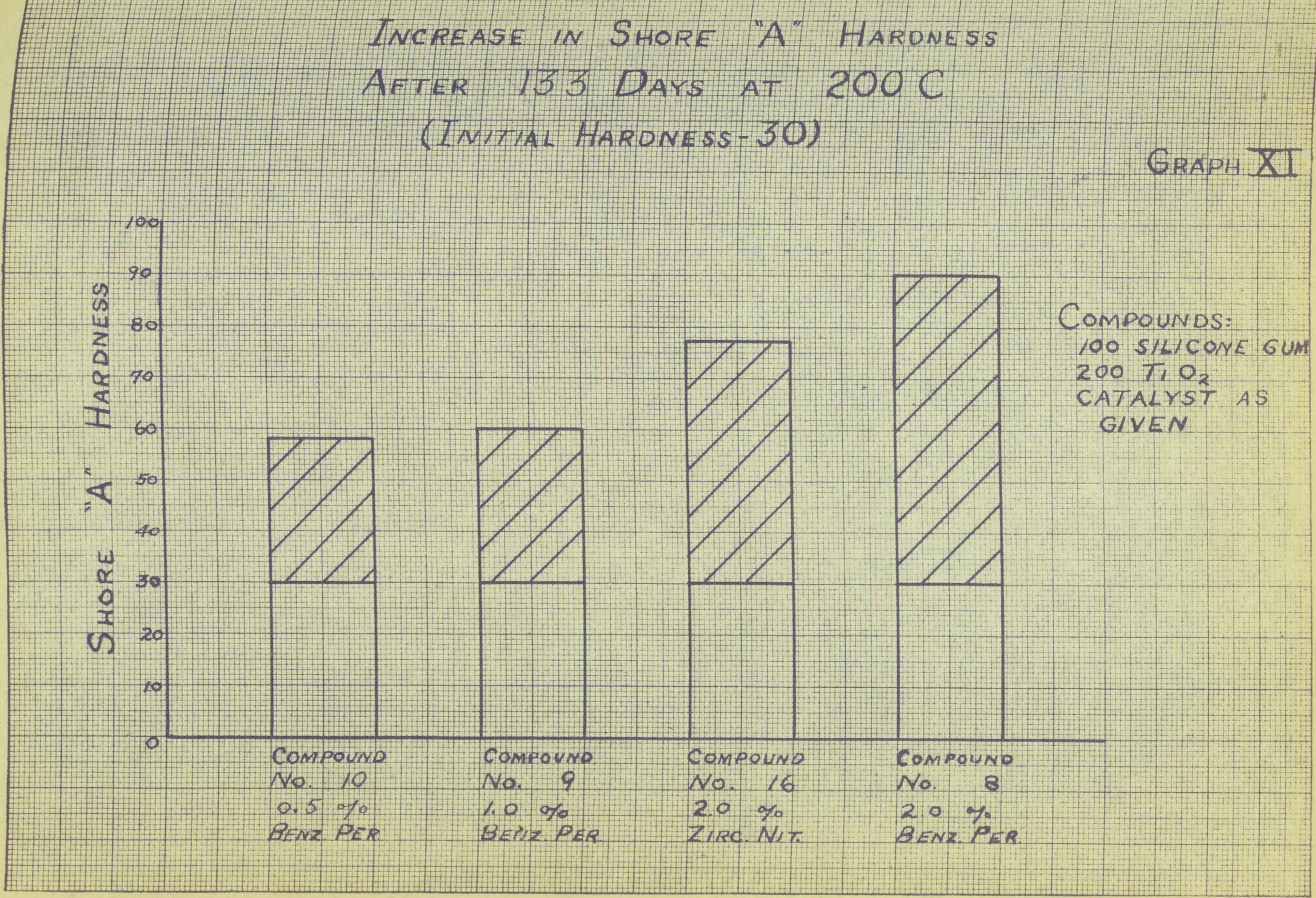
Comparison of Weight Loss and Flexibility of TiO_2 and Celite Filled Compounds Cured With Zirconyl Nitrate

Graph IX





EFFECT OF AMOUNT OF TiO2
ON WEIGHT LOSS OF COMPOUNDS
CATALYSED WITH ZIRCONYL NITRATE
GRAPH X
PERCENT WT. LOSS OF GUM
40
30
20
10
0
20
40
60
80
100
120
140
160
180
200
220
CPD. NO. 16
CPD. NO. 19
DAYS AT 200 C



INCREASE IN SHORE "A" HARDNESS
AFTER 133 DAYS AT 200 C
(INITIAL HARDNESS - 30)
GRAPH XI
SHORE "A" HARDNESS
100
90
80
70
60
50
40
30
20
10
0
COMPOUND No. 10 0.5 % BENZ PER
COMPOUND No. 9 1.0 % BENZ. PER
COMPOUND No. 16 2.0 % ZIRC. NIT.
COMPOUND No. 8 2.0 % BENZ. PER
COMPOUNDS:
100 SILICONE GUM
200 TiO2
CATALYST AS GIVEN

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