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SYNTHESIS OPERATIONS IN SCHWARZHEIDE  
by Dr. Reisst.

During my stay in Schwarzheide, all questions related to synthesis operations, including the gas purification, condensation, and activated charcoal have been discussed. The following is a summary of the information obtained:

1). Quality of the Catalyst  
A). Reduction Value

The reduction value of the catalysts provided by the catalyst factory Ruhrchemie is 40 - 50%. The lower value of reduction in the previously delivered pure cobalt-thorium catalysts was introduced with the magnesium containing catalysts, which produced too great difficulties of starting because of the high starting activity of these catalysts. The only experimental basis for the use of these less reduced catalysts consisted in transferring to large scale operations results obtained in the experimental laboratories. Catalysts made at the Schwarzheide catalyst factory have considerably higher reduction values. They vary between 49 and 61%, and the monthly averages are:

February 1944	56%
January 1944	54%
December 1943	58%
November 1943	58%

Even when considering the difficulties in sampling, the variations in the individual values are smaller than ours. The reduction conditions here in use are kept constant and are as follows:

A trough charge - 210 kg is reduced for 50 minutes with 2700 m<sup>3</sup>/h of 90% hydrogen, with the inlet temperature, measured above the mass at 360°, and the outlet temperature beyond the mass at 367°. These conditions were also kept during the reduction of the catalysts precipitated on burned kieselguhr and of diluted experimental catalysts with a proportion of cobalt: kieselguhr of 1 : 2.7. The reduction values in these cases also were kept within the same limits. They believe in Brabag that a lowering of the cobalt content in the synthesis reactor may in part be equalized by an increase in the reduction values. The results of synthesis in Schwarzheide

and the experimental results with the Brabag catalysts in Rheinpreussen speak in favor of the Brabag catalysts, and it is up to us to find out to what extent our reducing conditions are in accord with those in Schwarzheide and whether an increased reduction value would result in an improvement, especially of catalysts of lowered cobalt contents.

The men at the catalyst factory, of the synthesis and of the experimental departments emphasized in particular the absence of any exact knowledge between the reduction values and the activity of the catalysts, nor between the degree of liquefaction and the age of the reactor. Tests on these relationships are planned. Comparison of about 100 reactor charges, carried over a long period, have failed to give any clear answer. The reduction values have in many cases been redetermined after the emptying of the reactors, and the results were found to be the same as when the reactors were first started.

The experimental department is at present conducting experiments on the effect of the reduction value on the catalyst structure. Dr. Kröpelin is making use in this investigation of X-ray analysis and the electron microscopy.

#### B). Shape of Catalysts.

The abrasion of the Brabag catalysts is affected by the properties of the Kieselguhr. Burned kieselguhr has given particularly favorable results in this connection. With the usual method of preparation of catalysts, the abrasion resistance with unreduced pieces amounts to 15 - 16%/month, with the reduced particles - 22%. They are redetermined for every reactor charge. The screen analysis of the unreduced catalyst particles given to me, and covering about 100 reactor charges, were as follows:

1 mm and finer	about	1%
1 - 2 mm	"	37%
2 - 3 mm	"	48%
over 3 mm	"	24%

The catalyst particles larger than 3 mm are of a suitable size, since they have been obtained with a rectangular screen with holes 1 x 2 mm, and they become 2 - 3 mm particle size after reduction. The management stated that freedom of the catalyst from dust must be particularly emphasized, and that improvements in the screening reduced the resistance of the cold reactor chamber from the original 100 and over mm water column with a 1000 m<sup>3</sup> gas thruput, to 60 mm water column. Resistances of the individual reactor charges are so uniform, that any adjustments of the reactor load are rarely required. The catalyst factory explains, that the two screening plants in operation produce a material with different dust contents. They have no explanation for these differences, which must be the result of small differences in the screen vibrations, because

the two installations have been built exactly alike. The Schwarzheide factory is only used to 60% of its capacity, and almost the whole of the requirements can be met at the one screening installation which works satisfactorily.

Gray catalyst particles, i.e. partially oxidized, have been observed in Schwarzheide as well. Exhaustive studies have shown, that this oxidation takes place when the trough is emptied into the bucket, because the amount of nitrogen in the trough and the bucket was insufficient. After the nitrogen connections at the trough and the bucket had been increased in size, no more gray particles could be observed.

#### C). Double Precipitation

The question of the extent to which the double precipitation of the cobalt solution is effective as means of improving the quality of the catalyst is being studied here at present. A single precipitation had been used here tentatively several years ago and been found then unsatisfactory, but after running some preliminary tests last summer, about 50% of the reactor charge consists now of catalysts which had been precipitated but once from solution. So far no changes in the catalyst activity have been noted, in agreement with the experimental results of the preceding summer. The opposite results found at the earlier date are at present explained by impurities in the synthesis gas and by failures in the hydrogenation, especially towards the end of it, when a greater amount of impurities was introduced into the cobalt solution.

#### D). Density of Cobalt (Proportion of Cobalt?)

Detailed studies are made in Brabag on the effect of the density of cobalt, as has already been communicated at the joint meeting of Jan. 12, 1944. These tests have not yet been completed, but it can already be said that further reductions in the cobalt content, as has been tried by us and by Brabag are not to be recommended on the grounds of the large scale tests in Schwarzheide. The exact information will be placed at our disposal after the completion or repetition of the tests.

#### 2). Synthesis

##### A). Carbon Monoxide - Hydrogen Proportion.

The CO:H<sub>2</sub> ratio must be kept between 1:2.00 and 1:2.05. It has been established, that when the ratio is permitted to drop, the yields are reduced. Experiments have also been run where the CO:H<sub>2</sub> ratio was maintained at 1:1.85 - 1:2.00. While no changes in the specific yields have in this case been observed, as long as operations were always run with the above proportions, higher reactor temperatures had to be maintained with ratios below 1:2.0, than is normal with proportions

above 1:2.0. When the ratio  $\text{CO:H}_2$  is lowered from 2.0 to 1.85, the proportion of the olefines in the gasoline fraction is increased from 35 to 40%.

The  $\text{CO:H}_2$  ratio plays an important role also in the second stage of gasification, and the excess hydrogen (85%) from the hydrogenation, amounting to 700 - 800  $\text{m}^3/\text{h}$  is added to the end gas of the two installations. This hydrogenation hydrogen is also added when no hydrogenation is being done. The daily operating records of Brabag show that the  $\text{CO:H}_2$  ratio is in most cases greater than 2.0, and this by itself would permit a favorable proportion of  $\text{CO}$  to  $\text{H}_2$  in the synthesis gas of the second stage. No increase in the methane or gasol formation was observed in this procedure.

### B). Age of Reactors and Stage Operations.

The age of the reactors, or the average time of operation of the different reactor charges, is no longer used in Brabag for the evaluation of quality of the catalysts. For this purpose is used the so-called "gas age" (Gasalter), which is the thruout of the ideal gas per reactor charge. These values have recently been changed. Schwarzheide I has set the gas age for the first stage as of 3.1 million  $\text{m}^3$ , i.e. the gas age has been raised, while Schwarzheide II has kept the gas age unchanged at 2,800.00  $\text{m}^3$ . The difference must be attributed to 1% greater excess of inerts in the gas in Schwarzheide II, and, while the resin formers are lower, it carries more harmful ingredients. Schwarzheide II uses a Koppers gas as synthesis gas, while Schwarzheide I uses a mixture of Koppers and Didier gas. The second stage of Schwarzheide II had the gas age reduced from 8.3 to 7.0 million  $\text{m}^3$ . This change in the gas age has necessitated also a change in the after treatment of the products of the two stages. To follow the example of Essener Steinkohle, the treatment should be more uniformly distributed over the two stages. This would require an increased number of reactors in the second stage. The conversion in the two stages is kept at about 74%, so that the total conversion of the two stages amounts to 94%. Increasing the conversion to over 94% resulted in no increase in the specific yield, only in an increase on the amount of methane formation, because of the active ingredient concentration of the Schwarzheide gas.

The affect of the catalysts poison (sulfur and condensates) manifests itself in the first stage in the smaller gas thruout, especially in Schwarzheide II. When considering the amount of converted gas or the production, the catalysts of the first stages of the two installations produce more than the second stages. It is not as yet known whether this poorer operation of the catalysts in the second stage must be attributed to the higher content of the inerts in the synthesis gas II, or to the harmful influence of the carbon dioxide and of steam.

Experiments made to clarify this question indicate that steam alone does not affect the catalyst production in the second stage, and that carbon dioxide must be considered particularly harmful. This however is merely a preliminary observation of the large scale experiments conducted in Schwarzhelde, about which a more detailed report will be made later.

C). Intermediate Reactivation of the Catalysts:

Since the end of 1941 Schwarzhelde has introduced, after a long test period on a semi-industrial and industrial scale, an extraction followed by short hydrogenation in the first and second stages. A definite program has been established for the first stage. The catalysts are subjected to an intermediate reactivation after passing 250,000, 500,000, 800,000, 1,100,000, etc.  $\text{nm}^3$  of the  $\text{CO-H}_2$  ideal gas age. The principal effect of this regular intermediate reactivation is a considerable lowering of the reactor temperatures, as can be observed in the monthly average values since the end of 1941. Several of the curves of contraction, loads and temperatures for about 200 reactors changes in the first stage show, that with about 1,100,00  $\text{nm}^3$  ideal gas age, the temperature of the reactors is between 170 and 190°C, with a 57% contraction, with the synthesis gas load of 1200  $\text{nm}^3/\text{hr.}$ , and the synthesis gas containing around 80% of active ingredients. The degree of liquefaction at these low temperatures must be particularly favorable, according to tests made by me in 1974, and it is not therefore surprising that Schwarzhelde has achieved a considerable increase in the specific yield since the introduction of these intermediate reactivations. The possibility of considerable increase in the reactor age may be an additional benefit of this procedure. However, this possibility has not as yet been utilized. Schwarzhelde I has begun only during the last two months raising the gas age without introducing any noticeable increase in the average reactor temperatures.

The reactivation in the second stage is carried out after longer time intervals than in the first stage. Hydrogenation without extraction is here occasionally used.

The extraction for the reactivation is done at 170° C with 16 - 18  $\text{m}^3$  of oil/reactor, the oil having a boiling range of 170 - 230° C. The operation requires about six hours. The oil is supplied through 12 spraying orifices, introduced into the reactor on two orifice manifolds. The final extraction is done with a considerably higher amount of oil. The hydrogenation which follows it effects particularly favorably the paraffin content of the spent catalyst.

D). Emptying:

The filling and emptying weights of the individual reactors have been determined for several years in Schwarzhelde by a filling supervisor, who transmitted these values to the operator. These weights permit one to judge the degree of cleanliness of the reactors. Values submitted to

-6-

me of all of the reactor fillings over the last three years, or a total of about 2,000 reactor fillings, showed definitely that the filling volume of the reactors of 10.7 m<sup>3</sup> may be considered as normal. The amount of catalyst introduced varies at most between 10 and 11.5 m<sup>3</sup>. Filling therefore is done here according to volumes, and the filling volume of the catalyst is always determined by the operating department during filling and emptying. A normal emptying is followed by a blowing out with compressed air the space between the fins of the reactors. If the emptying volume of the converter has become smaller than the volume found during filling, the blowing out is repeated once or twice. During the blowing out, which is done with the reactors open, the dust is sucked off with a steam injector and led through a small washer operated with the products of synthesis. The total dust is here precipitated and recovered from the sump by means of a filter press. This washing installation is very small. It is about equal to that operated by the Essener Steinkohle. The yearly average time required for the emptying in Schwarzhilde were:

Preliminary operations for emptying (Opening the covers, connecting the upright with the bucket).	6 men x 2 hours = 12 hours
Emptying, including the tapping down	6 " x 1 " = 6 "
Disconnecting the bucket and weighing	6 " x 1 1/2 " = 9 "
Blowing out the reactor	3 " x 5 hours = 15 "
Testing the reactor (mainly screen tests)	2 " x 4 " = 8 "
	<hr/> 50 "
Filling:	
Preliminary work (setting the basket, bringing the bucket)	4 men x 1 1/2 hours = 6 hours
Filling with catalyst	4 " x 3/4 " = 3 "
Closing the reactor cover	4 " x 3 " = 12 "
	<hr/> 21 "

The total time required for emptying and filling is therefore about 71 hours. No repair work needed has been here added.

The converter top is tightened with round Buna packing, but a cardboard packing saturated with aluminum soap has been found here equally good.

Brabeg has studied the question of the uncertainties of gas measurements on the individual reactors, which have been observed by other concerns. No great deviations have so far been found with orifice meters, although a fairly large number of reactors have been standardized with a piston measure of the movable experimental converters.

It has already been mentioned that the slight crusts in the upper layers of the catalyst were the only difficulty met in the emptying. These have been principally observed in the unit reactors. It is hoped that these difficulties will be overcome by changing the method of starting, as will be discussed later. Naturally many reactors which have been injured by many years of operation may offer other difficulties, but these do not increase the time for emptying as we ourselves have observed on many reactors used for the usual synthesis.

The regular and frequent reactivation, and chiefly the final hydrogenation has greatly reduced during the last years the paraffin content of the used catalysts. The average paraffin content found in the catalyst factory was 2.13% in 1943, with the maximum content found during that year equal to 4.53%. It is believed that the de-paraffination carried out to a large extent during operations will also remove other undesirable materials deposited during synthesis operation upon the catalyst, and this will also simplify the purification of solutions at the catalyst factory. This extended de-paraffining will however primarily be beneficial in facilitating the labor of emptying.

The production of paraffin at the catalyst factory is so small, that the sale of this "black paraffin" has been suspended. The oil used in extraction is filtered with a yearly recovery of 600 - 700 kg. of cobalt in the dust. It has been found that the catalyst losses were especially great in some of the reactors, which was an indication of poor screening. It is recommended to call the attention of all synthesis factories to this source of losses. Water carried out from the reactors by the extraction oil is precipitated in Schwarzheide with a soda solution, and about 300 kg. of cobalt a year are also recovered from this source.

#### E). Starting the Reactors:

In Schwarzheide I all the reactors must be started with the synthesis gas I. In Schwarzheide II part of the reactors are started with synthesis gas II and kept in the second stage for about 100 - 200 hours, before being turned over to the first stage. In this installation the reactors may be operated as either the first stage, or the second stage. Schwarzheide differs in this respect from the other works where the catalysts are first operated for varying lengths in the second stage and are then connected in the first stage, and in some places again turned over from the first to the second stage towards the end of the operating period. The starting with synthesis gas I is then as follows:

The reactor is heated to 100° under gas pressure, 200 mm<sup>3</sup> of synthesis gas is passed through, while the reactor temperature is raised 10 - 15° hr. The catalysts in most cases begins its activity at about 140°. Up to a few months ago no considerable importance was attached to the increased methane formation at that time. The catalyst was permitted to react further while increasing the gas throughput and temperature. It has however been recently found that this method of starting causes the formation of crusts in the upper part of the reactors, and because these crusts make the emptying more difficult, the starting today is done so-to-say "from the ground up", that is at the start of the action at 140° this initial activity is permitted to die down, and the temperature and throughput only raised after this. This method of starting has produced a good result in some of the reactors recently emptied of their catalyst.

The synthesis reactors were also discussed in this connection. Operations emphasized that the best results were obtained with the formerly installed heat tube reactors except for the great damage to the finned bundle caused by improper operations. The unit reactors delivered in 1939 - 1940, are less well constructed, which resulted in an increased repair bill. Opinions differ on whether the steam is properly led away in the upper rows of tubes of the stall reactors. The operating department gives preference in new orders to the return bend reactors. Attention has been called to the return bend reactors used in the test operations of the Essner Steinkohle, where the upper row of tubes has a separated steam outlet. I shall get information in Bergkamen at the first opportunity on the test results with these reactors.

### 3). Gas Purification

#### A). Electrostatic filters

It has been stated during discussions at the meeting of Jan. 12, 1944 that at present electrode wires with 12% chromium made by Harckert-Hicken are used in all the electrostatic filters. It is no longer possible to make replacements with chromium steel. It is intended to replace them with galvanized iron wires, which have but a short life.

The individual chambers are closed with slides, which show no trace of corrosion, but it must be remembered that the synthesis gas to be purified contains a small amount of tar, which provides some protection against corrosion. The electrical equipment consists of 5 transformers for 4 chambers, with all of the chambers operated at the same potential. The time of operation of the synthesis gas blowers connected in series are not too highly estimated to be one year on the average.



### B). Preliminary Purification (Grobreinigung)

Since the electrostatic filters have been put in operation, the preliminary purification is working perfectly. Even with the small sulfur content in the synthesis gas, they get 40 - 45% absorption referred to the original weight of the gas, and with about 15% water. The single tower operates for two years, the oxygen addition is 0.2 - 0.3%. The towers are filled with pure lanta mass, to a thickness of layer of 450mm. The method especially emphasized by Essener Steinkohle, consisting in repacking the mass after a certain length of use has been successfully used for years in Schwanheide. Before introducing the electrostatic filters, this repacking was forced by the otherwise greatly increased resistance of the installation. This method of operation has been retained today because the gas inputs have been much increased over those originally intended, permitting only low resistances during preliminary purification. There is strong corrosion in both installations, notably of the supports and columns, and they are today first to be replaced in all of the purifiers. A special lacquer (Copalit of the G. Colladin Co. in Schonbach) has been used with very good results.

### C). Final Purification (Feinreinigung).

The highly porous alkaline mass has been again discarded in the final purification. The reason for it given was its short life. They have confirmed therefore the results obtained in our experiments. 12 aggregates are available for a gas thruput of 180 - 190,000  $\text{nm}^3/\text{hr.}$ , 10 of which are equipped with inserted baskets, and 2 with screens. Preference in every respect is given to the aggregates with the insert basket. The load amounts to 15,000  $\text{nm}^3/\text{hr.}$  with a capacity of 20,000  $\text{nm}^3/\text{hr.}$  The emptying requires on an average 140 hours of labor for the operations beginning with pushing in the slides and ending with the opening of the slides; a small pilot light as well as a small burner light are in use during that time in the preheater, so that the fire box is kept at about 300°. I have found out that the Kratag contains only one reserve bundle for the gas preheater for the 12 final purification aggregates. A few additional replacement bundles are on order. The used final purification mass was formerly simply discarded. At present the final purification mass is put for the hydrogen sulfide purification in the preliminary purifiers after being exposed to the air for a certain length of time. Only this one purification tower is filled up with the used final purification mass. It is connected from the very start of operations in the first position, and has so far absorbed 170 tons of sulfur with good efficiency. If we take into consideration the low hydrogen sulfide content of the synthesis gas in Schwarzhede, it represents good results and a useful utilization of the used up final purification mass. It has been emphasized that only when the insert is in the first position can such good results be obtained.

#### 4). Activated Charcoal Installation

The new charcoal has been used for over a year and has considerably increased the gasol yield. We have learned now that the grade of charcoal used was the Supersorbon SK. At the present however, the seasonal variations in the temperature of the cooling water for condensation and the activated charcoal installation have been overcome by the use of a steam jet cooling installation with gasol recovery today at about 98%. The steam jet cooling installation is a three stage installation furnished by Korking-Hannover. Its guaranteed capacity is 1.6 million h.u., i.e. 100 nm<sup>3</sup>/hr. water can be cooled by it from 26 to 10°. This guaranteed performance has at times been reached. The steam consumption at times is 11 to 900 nm<sup>3</sup>/hr of warm circulating water at 40° from the activated charcoal installation is used as cooling water, and is heated to 46°. The price of the installation without the piping and erection is 36,000 M. The erection and piping costs are appreciably higher.

The cooling water so obtained is used principally in the condensation of the upper part of the second stage of Schwarzheide II. Some of it, 10 - 20 nm<sup>3</sup> are sent to the intermediate cooler of the second stage activated charcoal installation II. The introduction of this water has lowered the temperature of the end gas from 25 - 29° to 18 - 20°. It has been found that lowering the temperature of water by 1° increases the Cs recovery by 4%. The addition of water in the intermediate cooler lowered the circulation water temperature by about 2°, permitting the maintenance of 20°. No accurate information could be given on the charcoal temperatures, but it is intended to test them carefully. The values will then be communicated to us.

Schwarzheide has observed corrosions only on the gas side of the final cooler and in the compensators of the cooling and heating gas lines. In the gas preheater the tubes are arranged vertically and it has at present offered no difficulties, aside from a leaky tube on the steep side. It must be remembered in this connection that the steam used in Ruhland is obtained in an evaporator.

#### 5). General

We may say about the further development of the total installation, that the present intake of 125,000 nm<sup>3</sup>/hr of synthesis gas will be increased to 195,000 nm<sup>3</sup>/hr. by increasing the gas production. No further increases in gas production and in the synthesis installation are at present contemplated.

A preliminary purification of synthesis gas by means of activated charcoal is at present being considered, as has already been reported previously, but the rather large amount of piping is not yet available, and one certainly may not count on the starting of it during this year. The required labor supply is as follows:

In principle Schwarzheide does not require over 8 hours of work even from foreign labor. The 8 hour shifts uses the following number of people per shift in the different operations departments:

Electrostatic filtration: 1 woman. 1 man is brought from another department for the rinsing. 1 man is used in the 3 blower houses per shift. The preliminary and final purification installations each require 1 man. For the emptying of the preliminary purification 10 men with 2 shift foremen are available, in the final purification 2 - 3 men for the filling and emptying of the towers (advantage of bucket installation). 1 crane operator per shift is available for the preliminary and final purification. In Schwarzheide I reactor building there are 4 reactor attendants, 1 reactor operator (for starting new reactor charges), 1 man to watch the water levels, and 1 shift foreman, 1 to 2 Hollanders are employed as converter attendants. In the converter building of Schwarzheide II there are 2 women as converter attendants, 1 reactor operator, 1 man on water level and 1 shift foreman. The 2 vaporization installations are serviced by 1 man each per shift. Likewise the condensation unit and the activated charcoal installation are each operated by 1 man per shift. In the hydrogen unit (pressure conversion with carbon dioxide scrubbing and methane splitting) 3 men are used per shift. No information comparable to ours could be obtained on the size of the repair crew, because most repairs were done by the M.T.A.. In this section the engineer Kunze is alone responsible for the synthesis, and possesses five years as operations assistant in the synthesis, and possesses therefore the required experience of this installation. Even with these favorable conditions the operations department is not satisfied with the repairs being done by the M.T.A., because as one might expect any attempt of the operating department to influence the work leads to difficulties. The two protective gas installations are each operated by 1 man per shift.

No definite information could be made about the introduction of residual gas for cracking in the gas producers, because this addition depends at times on the operating conditions and at other times on coke deliveries.

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Operations of Gas Wells No. 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45, 46, 47, 48, 49, 50, 51, 52, 53, 54, 55, 56, 57, 58, 59, 60, 61, 62, 63, 64, 65, 66, 67, 68, 69, 70, 71, 72, 73, 74, 75, 76, 77, 78, 79, 80, 81, 82, 83, 84, 85, 86, 87, 88, 89, 90, 91, 92, 93, 94, 95, 96, 97, 98, 99, 100, 101, 102, 103, 104, 105, 106, 107, 108, 109, 110, 111, 112, 113, 114, 115, 116, 117, 118, 119, 120, 121, 122, 123, 124, 125, 126, 127, 128, 129, 130, 131, 132, 133, 134, 135, 136, 137, 138, 139, 140, 141, 142, 143, 144, 145, 146, 147, 148, 149, 150, 151, 152, 153, 154, 155, 156, 157, 158, 159, 160, 161, 162, 163, 164, 165, 166, 167, 168, 169, 170, 171, 172, 173, 174, 175, 176, 177, 178, 179, 180, 181, 182, 183, 184, 185, 186, 187, 188, 189, 190, 191, 192, 193, 194, 195, 196, 197, 198, 199, 200, 201, 202, 203, 204, 205, 206, 207, 208, 209, 210, 211, 212, 213, 214, 215, 216, 217, 218, 219, 220, 221, 222, 223, 224, 225, 226, 227, 228, 229, 230, 231, 232, 233, 234, 235, 236, 237, 238, 239, 240, 241, 242, 243, 244, 245, 246, 247, 248, 249, 250, 251, 252, 253, 254, 255, 256, 257, 258, 259, 260, 261, 262, 263, 264, 265, 266, 267, 268, 269, 270, 271, 272, 273, 274, 275, 276, 277, 278, 279, 280, 281, 282, 283, 284, 285, 286, 287, 288, 289, 290, 291, 292, 293, 294, 295, 296, 297, 298, 299, 300, 301, 302, 303, 304, 305, 306, 307, 308, 309, 310, 311, 312, 313, 314, 315, 316, 317, 318, 319, 320, 321, 322, 323, 324, 325, 326, 327, 328, 329, 330, 331, 332, 333, 334, 335, 336, 337, 338, 339, 340, 341, 342, 343, 344, 345, 346, 347, 348, 349, 350, 351, 352, 353, 354, 355, 356, 357, 358, 359, 360, 361, 362, 363, 364, 365, 366, 367, 368, 369, 370, 371, 372, 373, 374, 375, 376, 377, 378, 379, 380, 381, 382, 383, 384, 385, 386, 387, 388, 389, 390, 391, 392, 393, 394, 395, 396, 397, 398, 399, 400, 401, 402, 403, 404, 405, 406, 407, 408, 409, 410, 411, 412, 413, 414, 415, 416, 417, 418, 419, 420, 421, 422, 423, 424, 425, 426, 427, 428, 429, 430, 431, 432, 433, 434, 435, 436, 437, 438, 439, 440, 441, 442, 443, 444, 445, 446, 447, 448, 449, 450, 451, 452, 453, 454, 455, 456, 457, 458, 459, 460, 461, 462, 463, 464, 465, 466, 467, 468, 469, 470, 471, 472, 473, 474, 475, 476, 477, 478, 479, 480, 481, 482, 483, 484, 485, 486, 487, 488, 489, 490, 491, 492, 493, 494, 495, 496, 497, 498, 499, 500, 501, 502, 503, 504, 505, 506, 507, 508, 509, 510, 511, 512, 513, 514, 515, 516, 517, 518, 519, 520, 521, 522, 523, 524, 525, 526, 527, 528, 529, 530, 531, 532, 533, 534, 535, 536, 537, 538, 539, 540, 541, 542, 543, 544, 545, 546, 547, 548, 549, 550, 551, 552, 553, 554, 555, 556, 557, 558, 559, 560, 561, 562, 563, 564, 565, 566, 567, 568, 569, 570, 571, 572, 573, 574, 575, 576, 577, 578, 579, 580, 581, 582, 583, 584, 585, 586, 587, 588, 589, 590, 591, 592, 593, 594, 595, 596, 597, 598, 599, 600, 601, 602, 603, 604, 605, 606, 607, 608, 609, 610, 611, 612, 613, 614, 615, 616, 617, 618, 619, 620, 621, 622, 623, 624, 625, 626, 627, 628, 629, 630, 631, 632, 633, 634, 635, 636, 637, 638, 639, 640, 641, 642, 643, 644, 645, 646, 647, 648, 649, 650, 651, 652, 653, 654, 655, 656, 657, 658, 659, 660, 661, 662, 663, 664, 665, 666, 667, 668, 669, 670, 671, 672, 673, 674, 675, 676, 677, 678, 679, 680, 681, 682, 683, 684, 685, 686, 687, 688, 689, 690, 691, 692, 693, 694, 695, 696, 697, 698, 699, 700, 701, 702, 703, 704, 705, 706, 707, 708, 709, 710, 711, 712, 713, 714, 715, 716, 717, 718, 719, 720, 721, 722, 723, 724, 725, 726, 727, 728, 729, 730, 731, 732, 733, 734, 735, 736, 737, 738, 739, 740, 741, 742, 743, 744, 745, 746, 747, 748, 749, 750, 751, 752, 753, 754, 755, 756, 757, 758, 759, 760, 761, 762, 763, 764, 765, 766, 767, 768, 769, 770, 771, 772, 773, 774, 775, 776, 777, 778, 779, 780, 781, 782, 783, 784, 785, 786, 787, 788, 789, 790, 791, 792, 793, 794, 795, 796, 797, 798, 799, 800, 801, 802, 803, 804, 805, 806, 807, 808, 809, 810, 811, 812, 813, 814, 815, 816, 817, 818, 819, 820, 821, 822, 823, 824, 825, 826, 827, 828, 829, 830, 831, 832, 833, 834, 835, 836, 837, 838, 839, 840, 841, 842, 843, 844, 845, 846, 847, 848, 849, 850, 851, 852, 853, 854, 855, 856, 857, 858, 859, 860, 861, 862, 863, 864, 865, 866, 867, 868, 869, 870, 871, 872, 873, 874, 875, 876, 877, 878, 879, 880, 881, 882, 883, 884, 885, 886, 887, 888, 889, 890, 891, 892, 893, 894, 895, 896, 897, 898, 899, 900, 901, 902, 903, 904, 905, 906, 907, 908, 909, 910, 911, 912, 913, 914, 915, 916, 917, 918, 919, 920, 921, 922, 923, 924, 925, 926, 927, 928, 929, 930, 931, 932, 933, 934, 935, 936, 937, 938, 939, 940, 941, 942, 943, 944, 945, 946, 947, 948, 949, 950, 951, 952, 953, 954, 955, 956, 957, 958, 959, 960, 961, 962, 963, 964, 965, 966, 967, 968, 969, 970, 971, 972, 973, 974, 975, 976, 977, 978, 979, 980, 981, 982, 983, 984, 985, 986, 987, 988, 989, 990, 991, 992, 993, 994, 995, 996, 997, 998, 999, 1000

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