

Investigation of Reduction and Precipitation Rate of Colloidal Gold Particles Obtained in the Process of Electrical and Electronic Waste Recycling

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ABSTRACT: *This paper studies the influence of process parameters: temperature, the weight of the added precipitant agent, pH and reducing agent on the kinetics of the process of reduction and precipitation of gold of 98.44% purity obtained in the process of electrical and electronic waste recycling. The optimal conditions of reduction and precipitation of colloidal gold particles in the process of recycling of different waste types in order to obtain high-purity gold are the following: an optimal process time ($\tau = 11$ minutes), temperature ($t = 5^\circ\text{C}$), the weight of the added precipitant ($m = 6$ g of pure gold) and $\text{K}_2\text{S}_2\text{O}_5$ as the preferred reducing agent. The results have contributed to a better understanding of reduction and precipitation rate of colloidal gold particles in order to manage and control this process. The granulometric analysis confirms the existence of colloidal gold particles. SEM analysis indicates spherical particles and good homogeneity of the sample. XRF analysis shows that gold obtained in this process of recycling is highly pure (99.99%).*

KEYWORDS: *Colloidal gold particles; The reduction and precipitation rate; Recycling, waste.*

INTRODUCTION

The intensive development of the industry of electronic and electrical products has caused the generation of large quantities of this specific waste. The percentage of recycled electrical and electronic components is very low, around 15%, and the remaining 85% of discarded components are disposed of in landfills [1]. Waste reduction, recycling, and reuse are the basic ways of managing such waste [2]. Electronic waste is classified as hazardous waste, which requires special treatment. However, the presence of precious metals, such as gold, silver, platinum, palladium, tantalum,

germanium and selenium in the waste makes it attractive for recycling. Many electronic parts such as resistors, ceramic capacitors, relays, parts of microwave devices, parts of computer devices, connectors and processors contain Ag, Au, Pt, Pb, Ru [3,4]. Recycling is an environmentally and economically efficient method, which has a positive effect not only on the environment but also on the society as a whole. Recycling of precious metals must be based on modern technological solutions.

The aggressive substances (HNO_3 and HCl) are used in the process of recycling that causes the development of

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highly toxic NO_x gases and their neutralization is very important from the aspect of environmental protection. It is possible to significantly reduce the amount of acid used in the recycling process by using the abrasive substance (Al_2O_3). A method of removing gold from electronic and electrical components using Al_2O_3 is not found in the literature. Chemical or electrochemical methods of separation of precious metals which require larger amounts of toxic substances (HNO_3 , HCl , H_2SO_4 , HCN) are only used in the process of recycling [5-7]. The recycling process that was used in this study includes the minimum amount of HNO_3 and HCl , wherein the toxic NO_x gases are converted into nitrates.

Colloidal gold particles may be obtained by reduction of the Au (III) salts using different reducing agents: formaldehyde, boron hydride, hydrazine, tin (II) chloride [8]. Various reducing agents also influence the size of the obtained colloidal gold particles. For example, gold particles sizes from 1.7 to 8.2 nm are formed using NaBH_4 at pH value of 5-9 [9]. The method of synthesis significantly affects the shape and size of colloidal gold particles [10]. In this paper, colloidal gold particles are obtained by reduction of HAuCl_4 with $\text{K}_2\text{S}_2\text{O}_5$ and L-ascorbic acid (vitamin C).

The aim of this research is to define the optimal process parameters: temperature, the weight of added precipitant agent, pH value and a reducing agent on the reduction and precipitation rate of 98.47% pure gold obtained in the process of recycling different types of waste. By reviewing the available literature the results of the impact of these parameters alone or combined with the reduction and precipitation rate of colloidal gold particles in the process of recycling were not found.

EXPERIMENTAL SECTION

Extraction of gold was carried out within the process of recycling gold-plated pins, RAM computer memory, connectors and graphics cards of processors. Plastic bottles with the aforementioned gold-plated parts, aluminum - oxide and distilled water in the ratio 33%: 33%: 33% are placed alternately on the centrifuge and shaker in order to remove a greater amount of gold of by means friction. When the process of removing gold plating is complete, the content of the bottles is transferred to a flask. Concentrated nitric acid is added to the flask in order to dissolve all metals except gold.

Neutralization of the NO_x gases was carried out by introducing a solution of the NaOH into the mixture to form the sodium nitrate. A slight warming is carried out to enhance the dissolution process. The solution is filtered and a sufficient amount of acid ($\text{HCl} : \text{HNO}_3 = 3:1$) is added to the filtrate in order to perform the dissolution of gold, according to the equation:



The obtained solution is then filtered so that the fluid level is not too high in order to allow leaching of residual gold from the filter paper by acid. The filtrate is heated until a yellow vapor becomes white in order to remove the excess of nitric acid which would subsequently impact the dissolution of gold. Neutralization of the solution is done by adding certain amounts of urea according to the equation:



A solution of tin (II)-chloride applied onto a filter paper with a solution of the sample is used as an indicator for gold (a black color appears in the case of a positive reaction to gold).

Reduction of gold ions is carried out simultaneously with potassium-metabisulfite $\text{K}_2\text{S}_2\text{O}_5$ and L-ascorbic acid $\text{C}_6\text{H}_8\text{O}_6$ (vitamin C). Then, the influence of temperature ($t=5^\circ\text{C}$; $t=10^\circ\text{C}$; $t=20^\circ\text{C}$ and $t=30^\circ\text{C}$) on the kinetics of reduction and precipitation of gold is examined. In order to study the deposition rate of gold, the different weights of elemental gold powder (0.5 g; 1 g; 2 g; 3 g; 4 g; 5 g and 6 g) have been added into the same quantity of HAuCl_4 . The reduction and deposition time is measured at a variety of the above conditions.

The influence of the pH value in the range from 3 to 7 on the kinetics of the reduction and precipitation of gold particles is not registered.

The resulting precipitate was then decanted and dried. The dried precipitate of gold was heated using the MAPP gas in order to obtain a solid sample.

Granulometric analysis of the obtained powder gold was done on the instrument "Mastersizer 2000" type immediately after mixing the components, after two, four and twenty-four hours of holding at ambient temperature. The measurement range of the instrument was from 20 nm to 2 mm.

In order to define the shape of the gold particles, SEM analysis was done on the instrument type ZEISS EVO 15 Ma-Oxford (X -Max 50 mm²) with Energy Dispersive X-ray detector. Since the surface of powder gold is conductive, there was no need for vapor deposition patterns by graphite or gold.

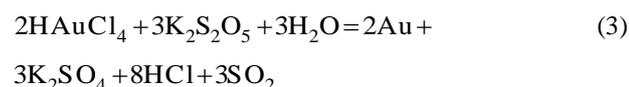
XRF analysis of the solid gold sample was done on the instrument of type M1 MISTRAL Bruker.

RESULTS AND DISCUSSION

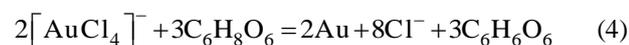
Figs. 1-7 show the dependences of reduction and precipitation time of colloidal gold as a function temperature at different amounts of the added elemental gold (0.5 g; 1 g; 2 g; 3 g; 4 g; 5g and 6g), and the influence of different reducing agents (K₂S₂O₅ and vitamin C) on the rate of said reactions.

From these dependencies, it can be concluded that with increasing amounts of added gold, the reduction and deposition time at all temperatures decreases. In the process of reduction of HAuCl₄ solution colloidal, gold particles are formed. In the presence of gold particles as nucleation centers, they lose properties of colloids, form the larger aggregates and precipitate. It can be also seen that reduction and deposition time increases along with an increase of temperature for both reducing agents. At lower temperatures, precipitation process is faster than at higher temperatures.

The obtained results show a faster process of reduction and precipitation with K₂S₂O₅ at all temperatures, which takes place according to the equation:



In the case of L-ascorbic acid as a lower reducing agent the process takes place according to the equation:



The highest reduction and precipitation rate of gold ions was achieved under the following conditions: $t = 5^\circ\text{C}$; $m = 6 \text{ g Au}$; $\tau = 11 \text{ minutes}$; K₂S₂O₅ as a reducing agent.

The granulometric analysis was done on a sample without addition of elemental gold as an agent for increasing of the reduction and precipitation. Colloid Particles are combined into larger agglomerates which precipitate

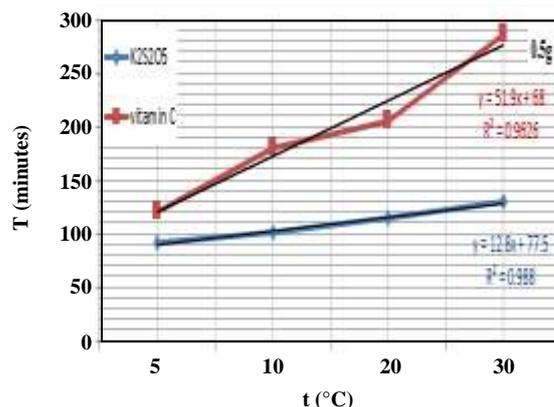


Fig. 1: The dependence of reduction and precipitation time of colloidal gold as a function temperature. The amount of added elemental gold is 0.5 g.

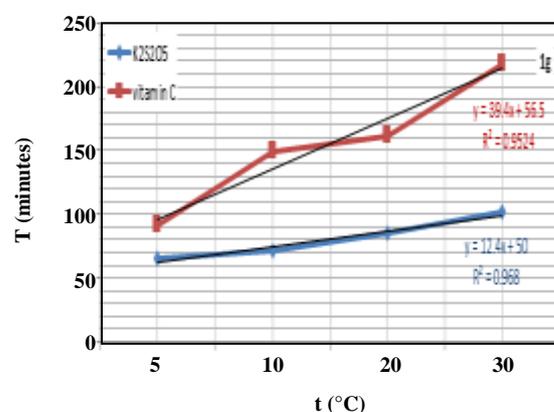


Fig. 2: The dependence of reduction and precipitation time of colloidal gold as a function temperature. The amount of added elemental gold is 1.0 g.

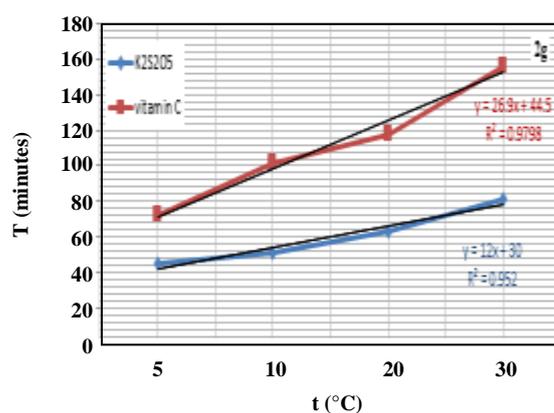


Fig. 3: The dependence of reduction and precipitation time of colloidal gold as a function temperature. The amount of added elemental gold is 2.0 g.

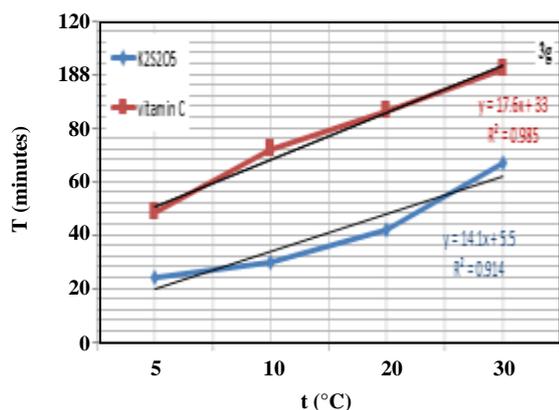


Fig. 4: The dependence of reduction and precipitation time of colloidal gold as a function temperature. The amount of added elemental gold is 3.0 g.

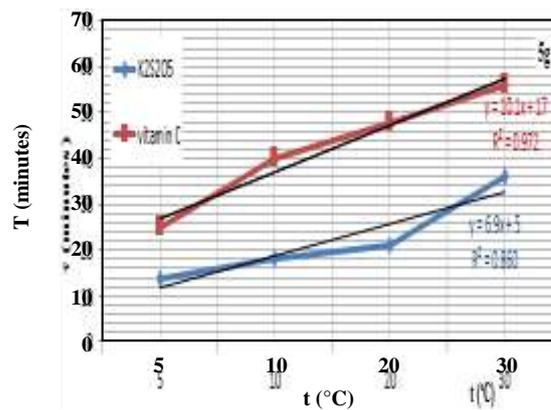


Fig. 6: The dependence of reduction and precipitation time of colloidal gold as a function temperature. The amount of added elemental gold is 5.0 g.

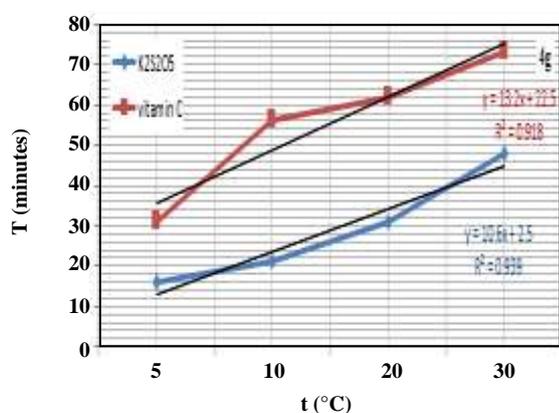


Fig. 5: The dependence of reduction and precipitation time of colloidal gold as a function temperature. The amount of added elemental gold is 4.0 g.

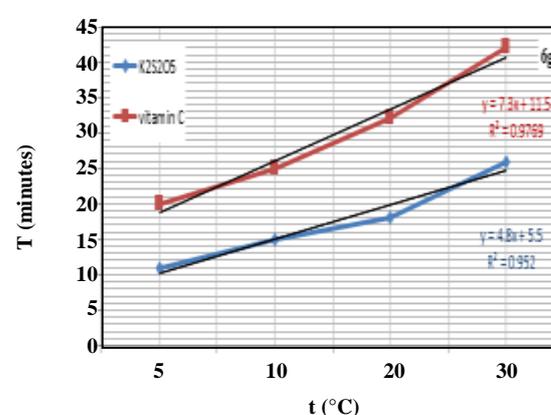


Fig. 7: The dependence of reduction and precipitation time of colloidal gold as a function temperature. The amount of added elemental gold is 6.0 g.

under gravitation after 24 hours (Fig. 8). The following results are obtained: 10% of the particles are smaller than 214 nm, the mean particle size is 274 nm, while 90% of particles are less than 479 nm in size.

It is very important for practical purposes that by adding of gold powder (98.47% purity) as centers of nucleation in a solution of colloidal gold separated in the recycling process, the reduction and precipitation time, which is 24 hours during the process of gravitation, decreases to 15 minutes.

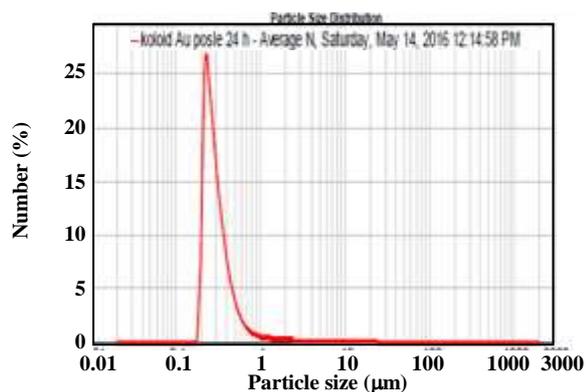
On the basis of the XRF spectrum of a solid sample of gold (Fig. 9), it was found that the content of gold in the tested sample is 99.99%. The presence of other elements which that sample classified as a material of very high chemical purity was not noted.

SEM analysis of a gold powder (Fig. 10) clearly shows the defined spherical shape of particles. The absence of large and small particles, as well as the beginning of the creation of agglomerates can also be noted, as a result of the drying and adhesion process of powder particles.

CONCLUSIONS

This study evaluates and analyses the reduction and deposition rate of gold as a function of temperature, the weight of added gold, pH and a reducing agent. Based on the present experimental results the following can be concluded:

- The reduction and precipitation rate of gold for the same amount of precipitant agent decreases along with the increase of temperature for both reducing agents.



Size (µm)	Number (%)	Size (µm)	Number (%)	Size (µm)	Number (%)	Size (µm)	Number (%)	Size (µm)	Number (%)
0.012	0.00	0.020	0.00	0.036	0.01	0.062	0.00	0.110	0.00
0.015	0.00	0.025	0.00	0.045	0.01	0.075	0.00	0.135	0.00
0.018	0.00	0.030	0.00	0.054	0.01	0.090	0.00	0.165	0.00
0.022	0.00	0.036	0.00	0.063	0.01	0.108	0.00	0.198	0.00
0.027	0.00	0.045	0.00	0.075	0.01	0.135	0.00	0.247	0.00
0.033	0.00	0.054	0.00	0.090	0.01	0.165	0.00	0.309	0.00
0.040	0.00	0.063	0.00	0.108	0.01	0.198	0.00	0.387	0.00
0.048	0.00	0.075	0.00	0.135	0.01	0.247	0.00	0.486	0.00
0.057	0.00	0.090	0.00	0.165	0.01	0.309	0.00	0.609	0.00
0.067	0.00	0.108	0.00	0.198	0.01	0.387	0.00	0.771	0.00
0.079	0.00	0.135	0.00	0.247	0.01	0.486	0.00	0.971	0.00
0.093	0.00	0.165	0.00	0.309	0.01	0.609	0.00	1.230	0.00
0.109	0.00	0.198	0.00	0.387	0.01	0.771	0.00	1.581	0.00
0.127	0.00	0.247	0.00	0.486	0.01	0.971	0.00	2.073	0.00
0.147	0.00	0.309	0.00	0.609	0.01	1.230	0.00	2.766	0.00
0.169	0.00	0.387	0.00	0.771	0.01	1.581	0.00	3.639	0.00
0.194	0.00	0.486	0.00	0.971	0.01	2.073	0.00	4.809	0.00
0.221	0.00	0.609	0.00	1.230	0.01	2.766	0.00	6.351	0.00
0.251	0.00	0.771	0.00	1.581	0.01	3.639	0.00	8.409	0.00
0.284	0.00	0.971	0.00	2.073	0.01	4.809	0.00	11.109	0.00
0.320	0.00	1.230	0.00	2.766	0.01	6.351	0.00	14.709	0.00
0.359	0.00	1.581	0.00	3.639	0.01	8.409	0.00	19.451	0.00
0.401	0.00	2.073	0.00	4.809	0.01	11.109	0.00	25.719	0.00
0.446	0.00	2.766	0.00	6.351	0.01	14.709	0.00	34.119	0.00
0.495	0.00	3.639	0.00	8.409	0.01	19.451	0.00	45.459	0.00
0.548	0.00	4.809	0.00	11.109	0.01	25.719	0.00	60.519	0.00
0.605	0.00	6.351	0.00	14.709	0.01	34.119	0.00	80.119	0.00
0.666	0.00	8.409	0.00	19.451	0.01	45.459	0.00	106.359	0.00
0.731	0.00	11.109	0.00	25.719	0.01	60.519	0.00	142.519	0.00
0.801	0.00	14.709	0.00	34.119	0.01	80.119	0.00	184.519	0.00
0.875	0.00	19.451	0.00	45.459	0.01	106.359	0.00	247.519	0.00
0.954	0.00	25.719	0.00	60.519	0.01	142.519	0.00	328.519	0.00
1.038	0.00	34.119	0.00	80.119	0.01	184.519	0.00	435.519	0.00
1.127	0.00	45.459	0.00	106.359	0.01	247.519	0.00	578.519	0.00
1.221	0.00	60.519	0.00	142.519	0.01	328.519	0.00	761.519	0.00
1.320	0.00	80.119	0.00	184.519	0.01	435.519	0.00	1000.519	0.00
1.424	0.00	106.359	0.00	247.519	0.01	578.519	0.00	1330.519	0.00
1.533	0.00	142.519	0.00	328.519	0.01	761.519	0.00	1780.519	0.00
1.647	0.00	184.519	0.00	435.519	0.01	1000.519	0.00	2380.519	0.00
1.766	0.00	247.519	0.00	578.519	0.01	1330.519	0.00	3180.519	0.00
1.891	0.00	328.519	0.00	761.519	0.01	1780.519	0.00	4250.519	0.00
2.021	0.00	435.519	0.00	1000.519	0.01	2380.519	0.00	5650.519	0.00
2.157	0.00	578.519	0.00	1330.519	0.01	3180.519	0.00	7450.519	0.00
2.300	0.00	761.519	0.00	1780.519	0.01	4250.519	0.00	9800.519	0.00
2.449	0.00	1000.519	0.00	2380.519	0.01	5650.519	0.00	12950.519	0.00
2.605	0.00	1330.519	0.00	3180.519	0.01	7450.519	0.00	17100.519	0.00
2.768	0.00	1780.519	0.00	4250.519	0.01	9800.519	0.00	22700.519	0.00
2.938	0.00	2380.519	0.00	5650.519	0.01	12950.519	0.00	30400.519	0.00
3.115	0.00	3180.519	0.00	7450.519	0.01	17100.519	0.00	40100.519	0.00
3.299	0.00	4250.519	0.00	9800.519	0.01	22700.519	0.00	52100.519	0.00
3.490	0.00	5650.519	0.00	12950.519	0.01	29800.519	0.00	66800.519	0.00
3.689	0.00	7450.519	0.00	17100.519	0.01	39100.519	0.00	84800.519	0.00
3.895	0.00	9800.519	0.00	22700.519	0.01	50500.519	0.00	107000.519	0.00
4.109	0.00	12950.519	0.00	29800.519	0.01	65000.519	0.00	134500.519	0.00
4.340	0.00	17100.519	0.00	39100.519	0.01	84000.519	0.00	170000.519	0.00
4.580	0.00	22700.519	0.00	50500.519	0.01	108000.519	0.00	218000.519	0.00
4.838	0.00	29800.519	0.00	65000.519	0.01	140000.519	0.00	283000.519	0.00
5.115	0.00	39100.519	0.00	84000.519	0.01	183000.519	0.00	371000.519	0.00
5.410	0.00	50500.519	0.00	108000.519	0.01	240000.519	0.00	480000.519	0.00
5.723	0.00	65000.519	0.00	140000.519	0.01	315000.519	0.00	610000.519	0.00
6.055	0.00	84000.519	0.00	183000.519	0.01	405000.519	0.00	760000.519	0.00
6.406	0.00	108000.519	0.00	240000.519	0.01	520000.519	0.00	940000.519	0.00
6.786	0.00	140000.519	0.00	315000.519	0.01	670000.519	0.00	1170000.519	0.00
7.195	0.00	183000.519	0.00	405000.519	0.01	860000.519	0.00	1480000.519	0.00
7.634	0.00	240000.519	0.00	520000.519	0.01	1100000.519	0.00	1890000.519	0.00
8.103	0.00	315000.519	0.00	670000.519	0.01	1410000.519	0.00	2430000.519	0.00
8.603	0.00	405000.519	0.00	860000.519	0.01	1810000.519	0.00	3140000.519	0.00
9.135	0.00	520000.519	0.00	1100000.519	0.01	2340000.519	0.00	4070000.519	0.00
9.700	0.00	670000.519	0.00	1410000.519	0.01	3040000.519	0.00	5280000.519	0.00
10.298	0.00	860000.519	0.00	1810000.519	0.01	3950000.519	0.00	6850000.519	0.00
10.930	0.00	1100000.519	0.00	2340000.519	0.01	5150000.519	0.00	8950000.519	0.00
11.606	0.00	1410000.519	0.00	3040000.519	0.01	6700000.519	0.00	11750000.519	0.00
12.326	0.00	1810000.519	0.00	3950000.519	0.01	8750000.519	0.00	15000000.519	0.00
13.090	0.00	2340000.519	0.00	5150000.519	0.01	11400000.519	0.00	18900000.519	0.00
13.900	0.00	3040000.519	0.00	6700000.519	0.01	14900000.519	0.00	24500000.519	0.00
14.756	0.00	3950000.519	0.00	8750000.519	0.01	19500000.519	0.00	32100000.519	0.00
15.660	0.00	5150000.519	0.00	11400000.519	0.01	25500000.519	0.00	42300000.519	0.00
16.612	0.00	6700000.519	0.00	14900000.519	0.01	33500000.519	0.00	55800000.519	0.00
17.614	0.00	8750000.519	0.00	19500000.519	0.01	44500000.519	0.00	73500000.519	0.00
18.668	0.00	11400000.519	0.00	25500000.519	0.01	59000000.519	0.00	96500000.519	0.00
19.776	0.00	14900000.519	0.00	33500000.519	0.01	77000000.519	0.00	126500000.519	0.00
20.938	0.00	19500000.519	0.00	44500000.519	0.01	100000000.519	0.00	166500000.519	0.00
22.156	0.00	25500000.519	0.00	59000000.519	0.01	131000000.519	0.00	221000000.519	0.00
23.430	0.00	33500000.519	0.00	77000000.519	0.01	172000000.519	0.00	286000000.519	0.00
24.762	0.00	44500000.519	0.00	100000000.519	0.01	228000000.519	0.00	367000000.519	0.00
26.154	0.00	59000000.519	0.00	131000000.519	0.01	305000000.519	0.00	480000000.519	0.00
27.608	0.00	77000000.519	0.00	172000000.519	0.01	400000000.519	0.00	625000000.519	0.00
29.126	0.00	100000000.519	0.00	228000000.519	0.01	525000000.519	0.00	815000000.519	0.00
30.710	0.00	131000000.519	0.00	305000000.519	0.01	685000000.519	0.00	1065000000.519	0.00
32.362	0.00	172000000.519	0.00	400000000.519	0.01	900000000.519	0.00	1395000000.519	0.00
34.084	0.00	228000000.519	0.00	525000000.519	0.01	1190000000.519	0.00	1830000000.519	0.00
35.878	0.00	305000000.519	0.00	685000000.519	0.01	1570000000.519	0.00	2395000000.519	0.00
37.746	0.00	400000000.519	0.00	900000000.519	0.01	2080000000.519	0.00	3215000000.519	0.00
39.690	0.00	525000000.519	0.00	1190000000.519	0.01	2770000000.519	0.00	4255000000.519	0.00
41.712	0.00	685000000.519	0.00	1570000000.519	0.01	3680000000.519	0.00	5600000000.519	0.00
43.814	0.00	900000000.519	0.00	2080000000.519	0.01	4850000000.519	0.00	7300000000.519	0.00
45.998	0.00	1190000000.519	0.00	2770000000.519	0.01	6350000000.519	0.00	9400000000.519	0.00
48.266	0.00	1570000000.519	0.00	3680000000.519	0.01	8350000000.519	0.00	12000000000.519	0.00
50.620	0.00	2080000000.519	0.00	4850000000.519	0.01	11000000000.519	0.00	15300000000.519	0.00
53.062	0.00	2770000000.519	0.00	6350000000.519	0.01	14600000000.519	0.00	19500000000.519	0.00
55.594	0.00	3680000000.519	0.00	8350000000.519	0.01	19500000000.519	0.00	25000000000.519	0.00
58.218	0.00	4850000000.519	0.00	11000000000.519	0.01	25500000000.519	0.00	32300000000.519	0.00
60.936	0.00	6350000000.519	0.00	14600000000.519	0.01	3350000			

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