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TECHNICAL ARTICLE

Plant-based coagulant of *Theobroma cacao* L. as a substitute for Mercury in Colombian gold beneficiation

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Abstract

The use of mercury in gold mining adversely affects the ecosystems and has raised concerns, especially in specific areas of Colombia. An alternative to the use of mercury involves using the mucilage of plant species for gold separation. Here we present the use of mucilage extracted from *Theobroma cacao* L pod husks. The coagulant agent can be obtained by: (i) cutting and macerating cocoa pod husks to obtain the mucilage, (ii) mixing the mucilage with water, and (iii) thermally treating the mucilage until a coagulant is obtained. This research showed that the coagulant agent from cocoa pod husks waste can replace mercury use in artisanal small-scale gold mining in Colombia. Our coagulant agent collects impurities, leaving the gold completely free, with a recovery percentage of 95%. Consequently, we can eliminate the use of mercury from the gold beneficiation process and thereby reduce water, soil, and air contamination. Additionally, by using solid waste as a coagulant agent, this becomes a successful case of circular economy.

Keywords Artisanal and small-scale gold mining · Circular economy · Cocoa pod husk · Plant mucilage · Pollution · Waste biomass

Introduction

Gold mining is a growing economic activity, but adversely affects communities by environmental deterioration and exposure to mercury (Hg), a toxic element that affects the health of populations (Olivero-Verbel 2011). The gold is recovered by heating the amalgamation in a container, which produces atmospheric emissions of Hg (de Lacerda and Salomons 1998). This use of Hg is not efficient; the average amount of Hg used in gold mining is approximately four times the annual production of gold (Keane et al. 2023). The Hg is amalgamated with the gold to separate the gold from other minerals.

Small-scale gold mining and the associated gold recovery activities present the greatest risk due to the inhalation of toxic Hg vapours (Cordy et al. 2011; Veiga et al. 2006). The released elemental Hg vapor poses both an occupational and environmental threat. Occupationally, those involved in small-scale mining (both directly and indirectly), as well as area residents, may inhale elevated levels of elemental Hg and be exposed to Hg-contaminated inorganics (e.g. soil, food).

In Colombia, Hg is used in artisanal and small-scale gold mining (ASGM) and accumulates in aquatic environments where it has the potential to biomethylate, after which the resulting methylmercury compound can be biomagnified in water bodies and trophic chains, which represent a risk for fish consumers (Baena et al. 2021). The Hg used in ASGM has captured global attention and concern for decades. Approximately 1350 t of Hg per year are released into the

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environment from ASGM, which is considered the largest source of anthropogenic Hg emissions in the world (UNEP – UE Environmental Programme 2013). However, modifications in the practices of miners poses a challenge, given their conviction that gravity concentration or alternative methods prove ineffectual in extracting fine gold particles. For more than five decades, researchers, international agencies, non-governmental organizations (NGOs), and companies have endeavoured to eradicate the use of Hg in ASGM, typically by efforts to develop sustainable gold concentration techniques (UNEP – UE Environmental Programme, 2019).

Colombia is the fifth largest gold producer in Latin America, and Antioquia (46.1%) is the state in Colombia with the largest production (Baena et al. 2021). Many tons of Hg are dumped where gold mining is particularly intensive, so Columbia provides a clear example of the devastating effects of gold mining on natural environments and the human population (Diaz et al. 2020). Colombia has been considered the largest per capita Hg polluter (Cordy et al. 2011).

ASGM is one of the leading causes of Hg release into aquatic ecosystems in Latin America (Agudelo-Echavarría et al. 2020; Veiga 2010). Although this activity significantly contributes to rural employment in Colombia, the apparent economic prosperity resulting from gold mining contrasts with the concurrent loss of ecosystems and the adverse impact on the health and well-being of communities. Despite the existing alternatives for avoiding Hg in Colombian gold extraction, Hg pollution in mining areas has been almost exclusively associated with this activity (Marrugo-Negrete et al. 2008; Olivero-Verbel et al. 2011). The inappropriate use of Hg and the poor technical knowledge on handling and recovering of this metal have facilitated Hg contamination of water bodies and the atmosphere. Hg pollution is a daily threat to human health in towns surrounding these mines. A study of mothers and nursing children in the mining municipalities of Antioquia showed that 11.4% of women had Hg concentrations in breast milk above the permissible limit and 45% of children had high levels of Hg in their hair, indicating transfer during both gestation and breastfeeding (Molina et al. 2018).

In Colombia, a great number of plant species has been proposed for the extraction of mucilage that can replace the use of Hg in ASGM. Among the species identified are yarumo (*Cecropia peltata*), escoba-babosa (*Sida rhombifolia*), escoba negra (*Pavonia fruticosa*), San Juaquín (*Hibiscus rosa-sinensis*), balso (*Ochroma pyramidale*), and guácimo (*Guazuma ulmifolia*) (García-Cossio et al. 2017). However, these species are native to the Chocó region, so they are not available throughout the entire Colombian territory or throughout the world. To eliminate the use of Hg in ASGM, we need to consider species with a wide distribution and that represent an additional benefit for communities that subsist from gold mining.

Theobroma cacao L. (cocoa) is a widely-distributed Amazonian tree from the family Malvaceae that is shade-loving and generally requires abundant rain for its development (ICCO- International Cocoa Organization 2013). Cocoa comes from highly biodiverse complex ecological structures (tropical rain forests) and is an important global crop. 5–6 million farmers in developing countries produce about 90% of the world's cocoa, and 40–50 million people depend on cocoa for their livelihoods (Kongor et al. 2016). Colombia is the tenth most important exporter of cocoa, which is mainly used to make chocolate (USDA (U.S. Dept of Agriculture) 2022). Cocoa pod husk (CPH), bean shells, and mucilage are the three main products generated in chocolate production (Martínez et al. 2012). CPH represents 70% of the fresh weight of the fruit, and after the chocolate components are extracted, it is the greatest source of waste. CPH has high concentrations of natural mucilage, which we hypothesized could be used to produce a coagulant agent that would allow Hg-free gold separation in ASGM.

Replacing Hg amalgamation with other practices seems to be a site-specific issue, which depends on investment capacity, education level, and motivation of miners. Support of local authorities to promote and sustain technological alternatives is critical. Today, no panacea or methodology can be widely promoted as a substitute for Hg. Therefore, an evaluation of the socio-economic context should be conducted before suggesting any alternative process. The aim of this study was to evaluate the technical feasibility and environmental advantages of a potential alternative to using Hg in ASGM, i.e. a plant-based coagulant from cocoa.

Materials and methods

Study area

The municipality of Remedios lies in the northeast of the department of Antioquia, at the coordinates 07° 01′ 21″ North latitude and 74° 41′ 46″ West longitude (Fig. 1). The climate is distributed between warm and temperate with an average relative humidity of 84% and an annual average temperature of 24.9 °C, with July having the highest temperature and November the lowest. The annual precipitation averages 2520 mm (Alcaldía Municipal de Remedios 2019).

The main economic activity of Remedios is mining (Bernal-Guzmán 2018; Molina et al. 2018). Remedios also profits from a planted forest, estimated at 120,000 ha of timber trees, mostly represented by balso (*Ochroma pyramidale)* and Nogal (*Cordia alliodora*). Pastures occupy an area of ≈ 50,000 ha. Agricultural production is scarce, although important areas destined for cocoa cultivation stand out as a phytoremediator of soils previously used by mining. Most of the food is brought in from other places, whereas the fruits generated by cocoa are discarded due to their bioaccumulation of potentially toxic elements. We obtained mucilage from the collection of CPH in the La Bonita farm, located on the periphery of the municipality of Remedios.

Extraction of Mucilage from the Cocoa Pod Husk

To extract the mucilage, pod husks from the mature fruit (when the epicarp is yellow in colour) were cut into pieces with sizes between 1 and 5 cm, preferably with areas of about 3 × 3 cm (Fig. 1C). To obtain the sap from plant material, we macerated the plant sections and extracted the liquid, which corresponds to sap, from the plant material. Subsequently, the chopped and macerated material and the sap was submerged in added water, which caused the plant material to swell (Fig. 2). Also, we extracted the mucilage from the vegetation of other plants such as black cadillo (*Bidens pilosa*) and balso (*O. pyramidale*). The material collected was cut into small pieces for cocoa and balso, whereas the cadillo was cut and macerated in a porcelain capsule with a mortar to obtain the sap.

Samples were submitted to a thermic treatment, where the material was placed in an oven at a temperature between 35 and 50 °C, for 6–10 h. Through the thermic treatment, the sap absorbed water and acquired a high viscosity. The acquired mucilage was observed to maintain its physical properties for at least 10 days after it was produced. To extend the life of the mucilage, other components such as stabilisers, preservatives, or refrigeration were used.

Coagulant agent

The mucilage acquired was mixed with additional water until a coagulant agent was obtained. We used different controlled doses of water from 10 to 70 mL to determine the optimum mucilage: water ratio. The mixture was agitated in a MaxQ 2000 orbital shaker (Thermo Scientific, Iowa, USA) at a speed between 100 and 170 RPM. After introducing the additional water, we obtained the coagulant agent, representing a gelatinous pseudo-solution designed to capture impure particles within the mixture, excluding those without the metals of interest.

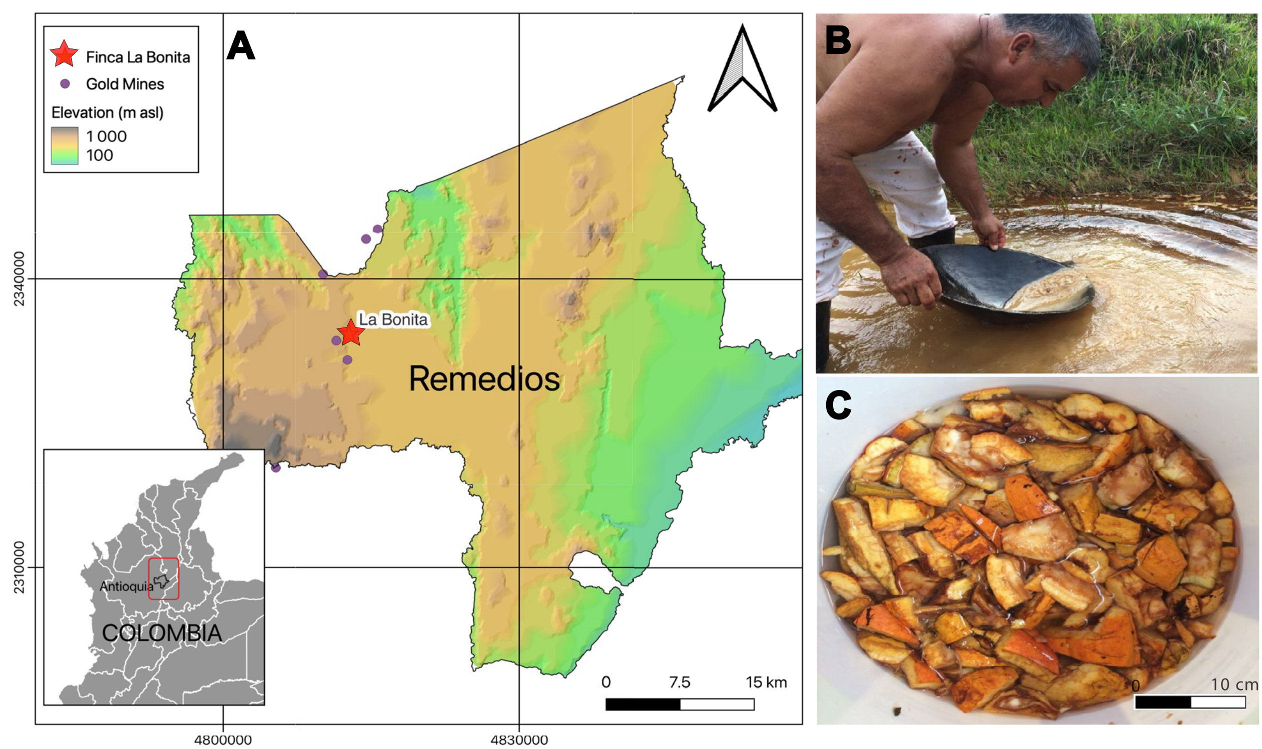

Fig. 1 **A** Location of the farm La Bonita, Gold mines and digital elevation map of Remedios, Antioquia. **B** Artisanal small-scale gold mining in Remedios (courtesy Yaira Rueda Jiménez). **C** Cocoa pod husk in water

Gravity separation of gold from other rocks and minerals

The coagulant agent is useful in the prospecting phase for different metals of interest and minerals, separating the metal from the mine rock. Specifically, we evaluated the coagulant obtained from CPH for gold recovery in ASGM alluvial ores. Once the gold has been crushed and panned, a fine powder known as "jagua" is left. This material contains gold ore and other precious metals, which is commonly treated with Hg and chemical compounds to extract any remaining value. The invention of cocoa coagulant to separate the gold from the jagua was compared with coagulants generated for two other plant species, black cadillo and balso. We applied panning as a traditional form of gravity concentration to separate the materials. The movement of water in the pan flushes out light minerals and sands, enabling heavy minerals such as gold to sink (Veiga and Gunson 2020). A pan can be the main tool used for gravity concentration. In Colombia, the main popular panning equipment is the conical "batea", with a diameter from 0.6 to 1.5 m and edges with an angle of 150–155° (Fig. 1B). Panning with batea demands experience from the operator and the movements are circular, removing the lighter minerals by overflow. Also, a laboratory test was carried out where the panning movements were simulated, using a MaxQ 2000 orbital shaker (Thermo Scientific, Iowa, USA) at a speed between 100 and 150 RPM, for 10 min (Fig. 3).

Results and discussions

Coagulant uses as method of gold separation

Panning test

The mucilage obtained was tested in a traditional wooden pan, adding the filtered substance and material from mining (black sediments with some gold particles) to the tray while making an elliptical movement (Fig. 1B). The pan was continuously shaken with the coagulant and sediment as we added water in dosed amounts. After 1–5 min of agitation in the pan, the impurities with which the gold is found (jagua) were separated on one side and the gold on the other (Table 1).

The added water had a neutral pH, preferably between 7 and 8, an apparent colour range between 5 and 15 UPC, and a turbidity below 2 NTU; levels above this would add impurities to the system, decreasing the gold recovery percentage.

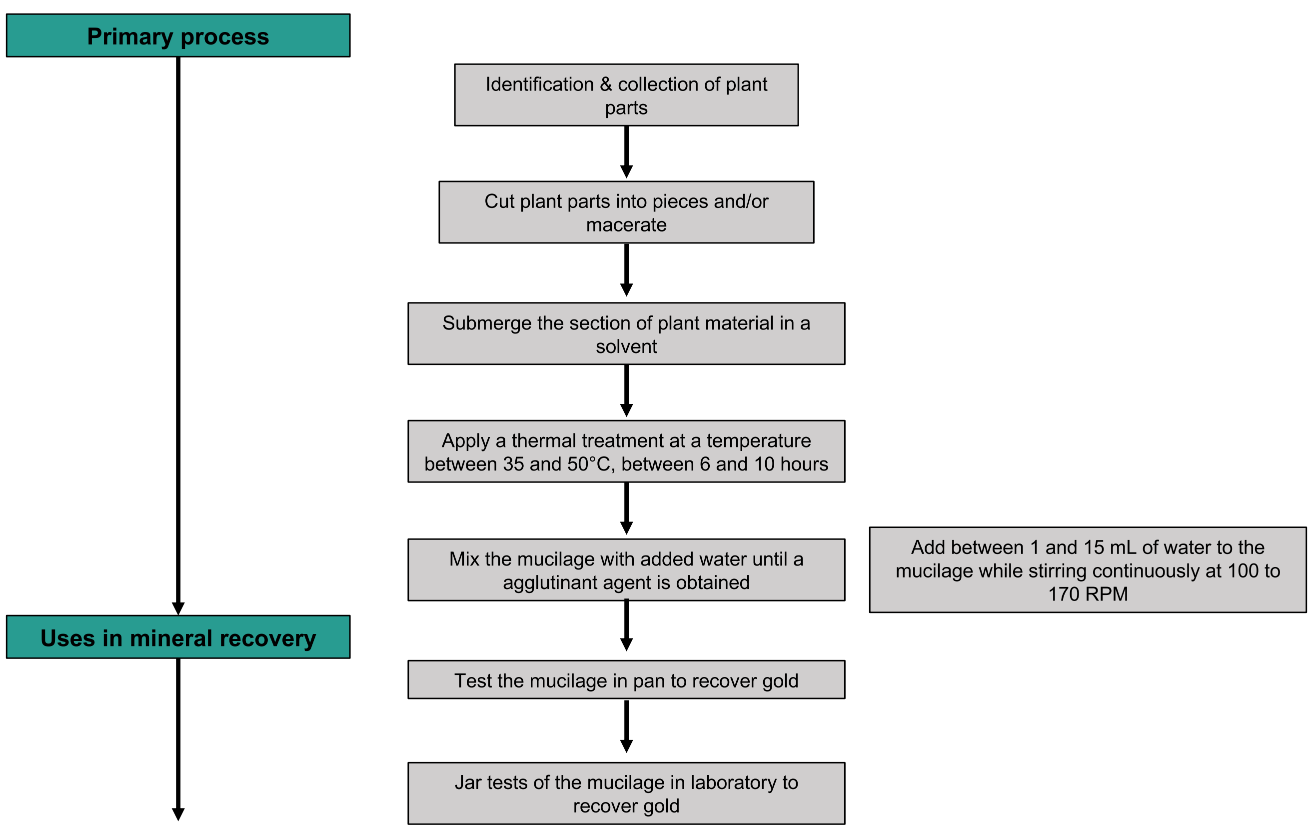

Fig. 2 Method for obtaining a plant-based coagulant agent to separate gold from jagua

The results obtained from the pan experimentation indicate high efficiency in gold recovery using cocoa (96.7%) and cadillo (83.3%) mucilage (Table 1). Higher quantities of cocoa mucilage and added water resulted in the highest percentage of gold recovery (96.7%). Thus, plant-based coagulants demonstrate efficacy in gold recovery, especially for cadillo and cocoa species. However, the cadillo coagulant agent has some limitations, as it must be employed within 72 h after extraction and does not preserve its properties when refrigerated. In contrast, the cocoa coagulant exhibits greater durability and can be refrigerated to preserve its viscosity.

Laboratory test

After carrying out the tests using the pan and verifying that the method and the coagulant agent were indeed functional, we proceeded with the jar test in the laboratory (Table 2). For the jar test, samples of cadillo and balso showed a substantial decrease in viscosity after refrigeration. Gold recovery for those agents identified as optimal was nearly immediate (Table 2). However, when the mucilage was used without added water, it trapped both gold and jagua. We underscore the significance of adding water in measured doses every 2 min to achieve an optimal mucilage concentration. In contrast, when all the water is added at once, it takes at least an additional 10 min longer to observe the results.

According to the results, the cadillo is an efficient mucilage, showing high recovery percentages, possibly because the cadillo has a manual viscosity of 9.94 mPas, while cocoa has a viscosity of 5.30 mPas. However, the mucilage of cadillo should not be refrigerated and preferably used within 72 h after its extraction. In this sense, it does not represent a cost-efficient and durable alternative for the artisanal miner. Also, we observed a percentage of recovery with cocoa reaching up to 90%, with an optimal composition of the coagulant agent (Fig. 4). When the coagulant was optimal for gold recovery, the results were seen in less than two minutes. We also confirmed that cocoa coagulant traps the jagua and does not allow it to move independently; thus, the gold remains at the bottom of the container in such a way that when it is emptied, the gold remains there and just the coagulant comes out with the jagua.

Gold recovery is characterised by concentrated high quantities of iron sands, which occupy 80% of

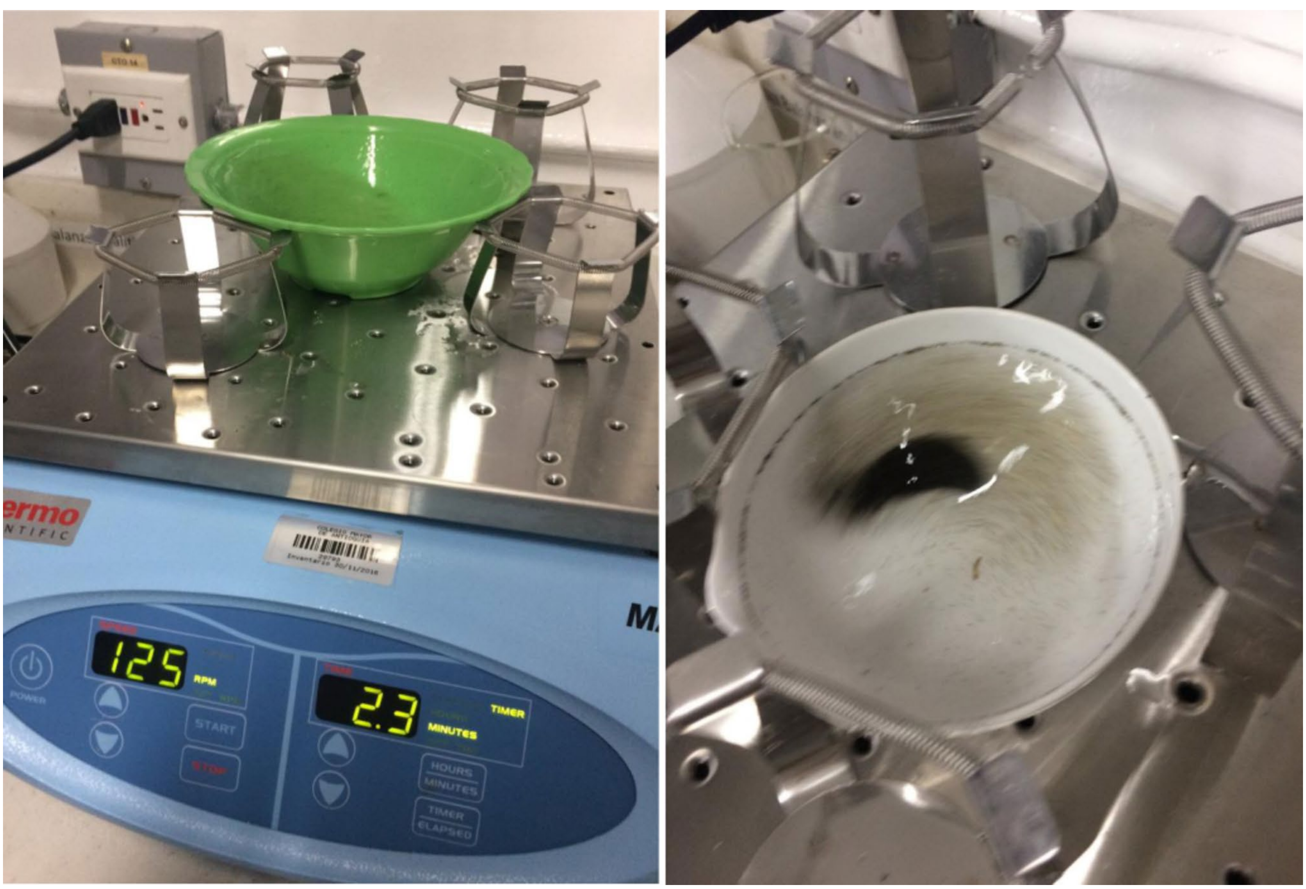

Fig. 3 Agitation in the laboratory to separate the gold from the jagua and sands

Table 1 Panning results, using mucilage from three plants to recover gold from jagua

Plant	Mucilage (mL)	Agitation time (min)	Jagua (oz)	Gold (g)	Water (mL)	Gold recovered (g)	Gold recovered (%)
CD	15	1	2	6	150	5	83.3
BL	30	2	4	6	300	3.5	58.3
CC	15	1	2	6	150	5.8	96.7

CC Cocoa (*Theobroma cacao* L.), *BL* Balso (*Ochroma pyramidale*), *CD* Cadillo (*Bidens pilosa*)

the jagua-gold composition and are difficult to remove (Hylander et al. 2007). However, when we used cocoa mucilage, gold recovery of 95% was achieved (Fig. 4). Like other separation techniques, the efficiency is determined by variables such as solution pH, ionic strength, solvent concentration, and temperature (Somasundaran 1975). Adequate control of these variables offers the opportunity to separate minerals in ASGM.

Coagulant agent of cocoa as alternative for mercury in gold separation

This coagulant is useful for gold separation, offering an environmentally sustainable alternative that does not represent public health risks. Our results showed better gold recovery (95%) for the cocoa coagulant than for the cadillo and balso species. In addition to the durability of cocoa mucilage, this constitutes an ideal coagulant agent to separate minerals in ASGM. Local communities in Colombia have tried using cadillo mucilage (e.g. García-Cossio et al. 2017) instead of Hg. However, the results of this research suggest that cadillo mucilage loses its viscosity in 48 h, unlike cocoa, which can last more than 72 h and is preserved refrigerated. Test 2 represents the optimal combination of variables (10 mL of mucilage, 60 mL of water, and 140 RPM) for the cocoa mucilage, which yielded a 95% gold recovery from the coagulant (Fig. 4). Therefore, CPH provides excellent gold recovery percentage, compared to other techniques, such as cyanide leaching (75%), borax (80%), and gravity (60%) (Davies 2014; Jønsson et al. 2009; Manzila et al. 2022). We highlight that the uptake of Hg-free technologies depends on their local availability, scalability, and adaptability at the local level.

An alternative technique for gold recovery should be affordable and easy to learn, and not require a large capital outlay, complex equipment, nor extensive technical support, and any chemicals used in the extraction of gold should be relatively safe and generate innocuous and biodegradable waste (Esdaile and Chalker 2018). Our cocoa-derived coagulant aligns well with these criteria, making a significant step toward a more sustainable and environmentally conscious ASGM.

Our novel approach not only enhances gold yield but also prioritizes environmental sustainability and economic viability. By focusing on a plant-based, environmentally friendly method, the study provides a significant advancement in the field of ASGM. It challenges the traditional reliance on harmful practices, such as Hg amalgamation, by introducing a sustainable and effective solution (Hinton et al. 2003; Malehase et al. 2017; Zolnikov and Ramirez Ortiz 2018). This innovation is particularly relevant in regions where both ASGM and cocoa cultivation are prevalent, offering a model

Table 2 Test in laboratory using coagulant agent from three plants to recover gold from jagua

Plant	Mucilage (g)	Jagua (oz)	Water (mL)	Agitation (RPM)	Agitation time (min)	Gold recovery (%)	Observation/performance
CC	45	2	5	105	3	60	Good
CC	10	2	60	140	3	95	Optimal
BL	5	2	40	140	3	–	Decrease
CC	5	2	40	140	3	–	Increase
CD	40	2	0	140	3	–	Rejected
BL	10	2	60	140	3	70	Good
CC	10	2	60	140	3	75	Good
BL	10	2	75	145	3	85	Optimal
CC	10	2	70	145	3	90	Optimal

CC Cocoa (*Theobroma cacao* L.), *BL* Balso (*Ochroma pyramidale*), *CD* Cadillo (*Bidens pilosa*)

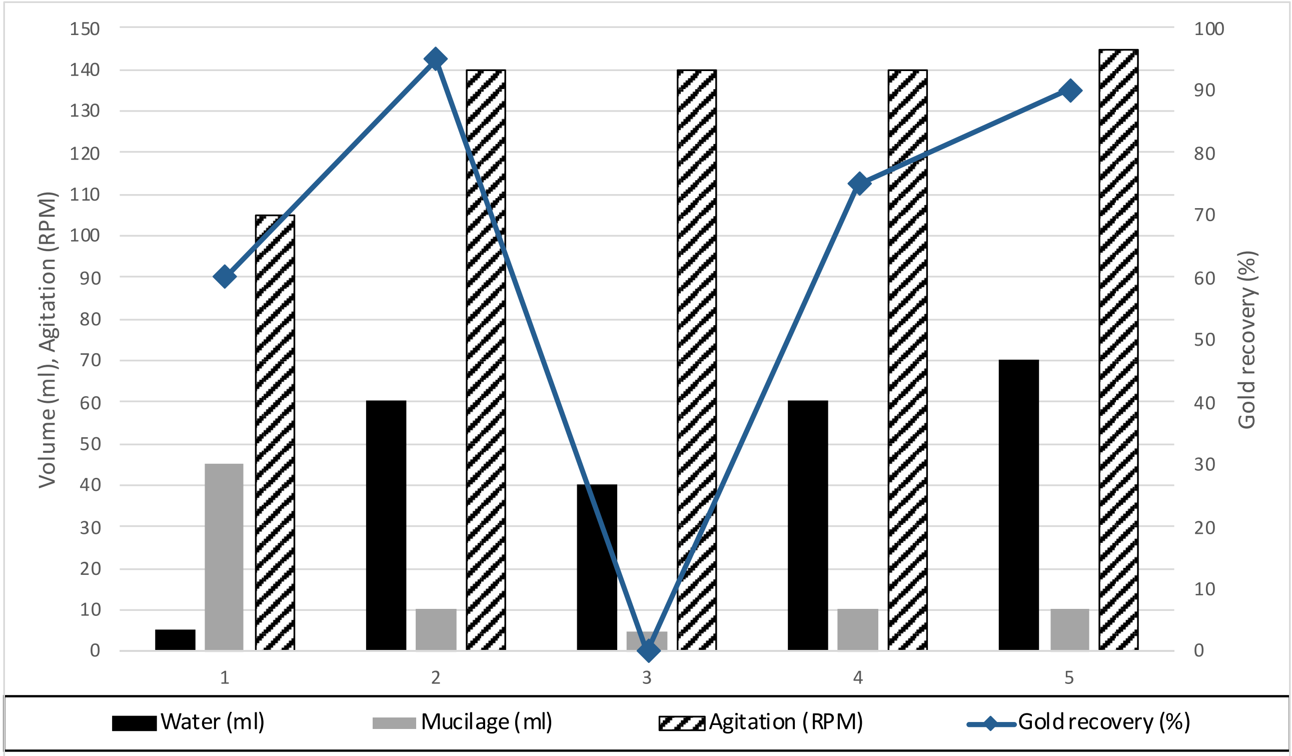

Fig. 4 Jar test for determining cocoa mucilage dosage (mL) in gold recovery percentage (%), revolutions per minute (RPM) and added water (mL) for cocoa coagulant

of local resource utilization that benefits both the environment and the local economy.

In terms of health and environmental impact, this research is pivotal in reducing the toxicological risks associated with Hg use in gold mining. The cocoa mucilage method would minimize environmental contamination and health hazards for mining communities. This aspect is particularly important, filling a critical void in the literature that often neglects the comprehensive assessment of health and environmental safety in mining practices (Bose-O'Reilly et al. 2008; Hsu-Kim et al. 2018; Swain et al. 2007). Moreover, the adaptability and scalability of this method make it a valuable model for various global regions, aligning with international efforts like the Minamata Convention on Mercury to reduce the use of hazardous substances in mining.

The particularities observed in the gold mining industry in Colombia position the country as an instructive case for investigating alternatives to Hg-dependent practices and mitigating ecosystem degradation resulting from mining activities (Diaz et al. 2020; Molina et al. 2018). Our results suggest that Hg-free gold mining is possible in Remedios Antioquia by using a plant-based coagulant agent to separate gold particles. Although our laboratory results exhibit great promise, the potential for scalability and particular utilization with panning techniques must be investigated. However, we must also bear in mind that any alternative that replaces Hg must be appropriate, considering the socio-economic context of the country and its challenges. Colombia has a long tradition of illegal and informal mining, the illegal trade of Hg, the remote location of many mining districts, obstacles for mining formalisation, and the involvement of illegal armed groups (Diaz et al. 2020). Addressing these complex socio-economic factors is crucial for the successful and sustainable transition to Hg-free gold mining in Colombia.

The importance of plant-based coagulant and circular economy

The results demonstrate that CPH mucilage has the potential for gold extraction. Every year, 675 kilotons of CPH accrues as a waste stream in the food processing industry worldwide (Panak Balentić et al. 2018). In this sense, this use of CPH would make it a more sustainable crop, where all parts of the plant are used and residual biomass accumulation is reduced (Villamizar et al. 2017).

Green extraction technologies, including plant-based coagulant agents, are emerging as sustainable methods for preparing and extracting these compounds from CPH. Our cocoa mucilage extraction is entirely eco-friendly and sustainable without affecting the environment. However, the solid waste remaining after the mucilage extraction process is an environmental concern. To meet the demands of a circular economy, not only must the bioproducts be utilized, but the waste (e.g. residue after extraction) must also be processed sustainably. Anaerobic digestion of CPH waste can be used to produce biogas (Montenegro-Orozco et al. 2016).

Mucilage extraction from cocoa generates great quantities of residuals, which are mainly discarded as waste and cannot fully justify the circular economy concept. Therefore, more research needs to be conducted on how to utilise these solid wastes for preparing compost or biochar, which can be used as a plant nutrient/fertiliser (e.g. Bahrun et al. 2018). Also, we need to explore technological advances related to the recovery of enriched extracts. Purified extracts enriched in flavanols and other polyphenols have shown impressive nutraceutical properties and value compared to the traditional use of CPH for agricultural composting (Acciardo et al. 2022).

The coagulant agent is relatively easy to implement and represents a cost-effective technique for communities with a shortage of technology and low economic autonomy. However, large-scale implementation will likely require public policy efforts to encourage the use of Hg-free methods for gold extraction. The complete integrated process could potentially avoid and reduce adverse ecological and economic effects, since valorisation of these wastes will reduce emissions of greenhouse gases and leachates (Sanchez and Stern 2016) and improve soil composition (Quintero Ramírez et al. 2017).

The utilization of CPH, a locally sourced and often discarded agricultural byproduct, is the key contribution of this research. By tapping into this sustainable resource, this study not only provides an eco-friendly alternative to Hg but also supports local economies in ASGM regions. Furthermore, our study extends beyond technical solutions to encompass broader socio-economic and policy implications. It emphasizes the importance of community engagement and governmental support in implementing sustainable mining practices. This holistic perspective integrates technical, social, and political dimensions, addressing a notable gap in existing research, which often isolates technical solutions from their wider contexts. This research, therefore, stands as a comprehensive contribution to the field of ASGM, offering insights and solutions that are environmentally sound, economically viable, and socially responsible, particularly relevant for countries like Colombia where the intersection of mining and agriculture poses unique challenges and opportunities.

Patent

The use of mucilage of *T. cacao* as a coagulant agent instead of Hg in gold mining has been presented in a patent that is in the validation and final phase by the Colombian Superintendence of Industry and Commerce (# NC2022/0007333) under the invention name *"Especies vegetales como alternativa al uso del mercurio en el proceso de beneficio del oro."*

Conclusions

This research reveals that a plant-based mucilage can be used to efficiently separate gold from jagua in ASGM in areas such as Remedios Antioquia, where cocoa plantations abound. The use of cocoa mucilage for gold separation will be very suitable and applicable in ASGM, particularly in rural communities, because it is simple to use, has a low cost, and has the potential to generate a circular economy. Using cocoa mucilage as a sustainable and efficient alternative to Hg can achieve 95% efficient gold recovery, surpassing traditional methods such as cyanide leaching, borax, and gravity concentration. Notably, this method diminishes the environmental and health risks traditionally associated with Hg use, aligning with global initiatives for sustainable mining. Moreover, it fosters a positive economic impact by utilizing locally available agricultural waste, presenting a cost-effective and environmentally responsible solution for ASGM communities.

The future trajectory of this research focuses on refining the application and efficacy of cocoa mucilage in gold recovery. Key areas for further study include optimizing the mucilage extraction process and adapting its application to various mining conditions and scales. Additionally, there is a need for comprehensive long-term studies to assess the environmental and economic sustainability of this method. Integrating this innovative approach with local mining practices, socio-economic considerations, and policy frameworks is also crucial. These future directions aim to solidify the method's practicality and scalability, ensuring it can be effectively implemented in diverse mining contexts.

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Data availability The datasets generated and analyzed during the current study are available from the corresponding author reasonable request.

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