Alternatives to PFAS in Floor Polish Formulations

Yuning Xu, Tessa Wardle, and Jenna Tan

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Abstract

Per- and polyfluoroalkyl substances (PFAS) are used in a wide range of consumer products due to their strong resistance to oil, water, and stains. However, PFAS are highly persistent in the environment and are associated with a myriad of irreversible negative health outcomes such as cancer, adverse developmental effects, and neurotoxicity. In 2021 the Maine Legislature required the removal of PFAS from "avoidable" use in products sold in the state by 2030. Floor polish is under consideration as one possible product category since nearly every floor polish on the market contains PFAS.

In this report, we examine potential alternatives to PFAS in floor polish, based on both technical metrics and health and environmental considerations. PFAS alternatives must facilitate the spread of floor polish across a floor surface as a wetting agent and concurrently level the polish so that it dries without application marks, dips, or dents. The metrics that correspond with this performance criteria include surface tension, contact angle, critical micelle concentration, and the octanol-water partition coefficient. To avoid regrettable substitutions, we assess our alternatives for persistence, carcinogenicity, reproductive and developmental toxicity, skin and eye irritation, among other hazard categories.

We offer two biosurfactants as potential alternatives to PFAS in floor polish: rhamnolipids and amino acid surfactants, specifically sodium lauroyl glutamate. Both alternatives demonstrate strong technical potential as leveling agents with surface tension values less than 30 mN/m at very low surfactant concentrations. Rhamnolipids achieve good wettability on a polyvinyl chloride surface, while sodium lauroyl glutamate has good recoatability potential. Both surfactants greatly reduce hazard concerns across human health endpoints and can degrade in the environment. However, further studies are required to clearly understand their impact on aquatic toxicity and long term effects on human health.

In conclusion, we suggest that there is high potential for rhamnolipids and sodium lauroyl glutamate to act as alternatives to PFAS in floor polish products with rhamnolipids as a strong contender. We recommend further investigation into improving the production efficiency of both substances, assessing their actual performance in a formulation on various floor textures, and replacing other potentially hazardous substances in floor polish formulations.

Background

Per- and polyfluoroalkyl substances (PFAS) are ubiquitous in our environment. PFAS form a class of substances made up of over 9,000 chemicals distinguished by a strong chain of carbon atoms bonded to fluorine.¹ This strong chain makes PFAS very difficult to break down.¹ These compounds are used in a wide range of products like nonstick pans, raingear, firefighting foam, and food packaging due to their exceptional ability to repel oil and water, and enhance product durability. The wide range of products with PFAS and their use have introduced large amounts of PFAS into the environment, where they have the potential to significantly affect surrounding ecosystems as well as enter drinking water supplies. Based on biological testing, it is estimated that 99% of Americans have PFAS in their bodies.² More studies have provided toxicological evidence linking PFAS with many adverse health outcomes such as cancer, neurotoxicity, and adverse reproductive, developmental and immunological outcomes in animals and humans.³ Thus, government agencies, corporations and the general public are realizing the importance of limiting the production and use of PFAS.³

The prevalence of PFAS in drinking water systems has emerged as a major concern in the United States and around the world. PFAS in drinking water is particularly alarming because the complete removal and destruction of PFAS is very difficult and often expensive. Municipal drinking water systems in the United States typically use filtration and chemical treatment (e.g. chlorine), but PFAS evade this treatment because they are highly soluble in water and are highly chemically stable.⁴ Options for filtering out PFAS include activated carbon treatment, ion exchange, and high pressure membranes (e.g. reverse osmosis) but may not remove all types of PFAS or work for high volume waste streams.⁴ Meanwhile, water sources not connected to a municipal supply, such as residential wells, may have no treatment mechanisms at all, leaving them particularly vulnerable to contamination by undetectable chemicals like PFAS.

Opportunities for PFAS exposure exist on the other end of water treatment as well. Municipal wastewater treatment systems across the country send dried and treated sewage sludge to agricultural fields for use as fertilizer, and this concentrated sludge has been found to contain high levels of PFAS and other pollutants like heavy metals and microplastics.⁵ Leachate from agricultural fields with this fertilizer can contaminate both groundwater and surface water with PFAS. A panel of experts found this to be the case in Maine, where recent testing revealed high levels of PFAS in drinking water, particularly residential well water and some school water sources.⁶ In part due to this concerning discovery, the Maine Legislature passed laws in 2021 requiring the removal of PFAS from "avoidable" use products sold in the state by 2030.⁷

We are working with Defend Our Health, a nonprofit based in Maine that works to protect residents from the harmful effects of toxic chemicals. We seek to explore whether viable alternatives to using PFAS in floor polish exist, therefore making it "avoidable" to add PFAS to floor polish formulations. PFAS in floor polish is a relevant concern for drinking water contamination because it is typically applied with a mop and bucket, and washing out these tools could lead to leftover floor polish going down the drain.⁸

PFAS uses in floor polish

Potassium N-ethyl perfluoro alkane sulfonamidoacetate (N-EtFOSAA) is a specific fluorochemical that represents fluorosurfactants commonly used in floor polish. N-EtFOSAA is highly persistent, can bioaccumulate, and is associated with aquatic toxicity, mammalian toxicity, and overall systemic toxicity.⁹ While there are still a number of data gaps regarding how N-EtFOSAA affects other important endpoints, we know that its degradation products include perfluorooctane sulfonic acid (PFOS) and perfluorobutanesulfonic acid (PFBS), which are better studied fluorochemicals. PFOS is highly stable in the environment and does not degrade. Various harmful health effects are associated with PFOS, including endocrine disruption, carcinogenicity, organ toxicity, immunotoxicity, neurotoxicity, and more.¹⁰ PFBS shares many of these concerns and presents high concern as eye and skin irritants, reproductive toxicants, and are associated with multiple types of cancer.¹¹

Current large scale users of floor polish include but are not limited to schools, hospitals, and large retailers including grocery stores. Floor polish primarily protects and extends the lifespan of floors, and secondarily enhances the visual appearance of floors and facilitates easier cleaning.¹² Floor polish is also marketed to residential consumers for home floors.

Fluorosurfactants aid in spreading the floor polish out across the floor, a characteristic called wetting, and help ensure no visual impurities are present when the polish dries, a characteristic called levelling (Figure 1).



floor finish without Capstone" fluorosurfactant shows clearly visible orange peel and significant surface defects.



Floor finish dosed with FS-60 at 75 ppm has drastically improved leveling and surface appearance of the floor finish on the vinyl tile.

Figure 1. Close up image of a vinyl tile without any added fluorosurfactant (left) compared to what it looks like with a commonly used fluorosurfactant in floor polish, Capstone FS-60 (right). This demonstrates the enhanced levelling performance that can be achieved when a floor polish contains PFAS. Source: Chemours.¹³

Fluorochemicals further stand out as a floor polish additive when it comes to recoatability. While PFAS replacements have been identified and tested successfully for the first one or two coats, their performance tends to decline when more than four coats are applied.¹⁴

Fluorosurfactants allow for the application of 4-5 coats without compromising gloss and high levelling performance while resisting blackheel scuffs and dents.¹³ Fluorosurfactants are effective in small doses (usually 100-275 ppm), which helps ensure that other properties of the floor polish do not start to degrade. Likely due to this small concentration, fluorosurfactants are extremely versatile and work well in many different formulations and under variable conditions.¹⁴

Approach



Figure 2. Overview of our approach toward finding a PFAS replacement. Image credits: Biolin Scientific, Water Quality Products, Phil Roeder.

Finding an alternative that can completely replicate PFAS performance in floor polish will be challenging given the many strengths of PFAS compounds. However, we strive to find viable candidates that have a strong baseline performance with improved human and environmental health outcomes. The scope of our work will focus on alternative surfactants that are commercially available. We will assess the technical performance of our proposed solutions alongside the potential health and environmental impacts.

Technical assessment

PFAS serve multiple functions in floor polish and can overperform, so we first narrowed our scope by identifying the essential functions of a replacement surfactant as its wetting and levelling performance. Both wetting and levelling agents help the floor polish spread evenly over a substrate but influence different interfaces. Wettability is related to the liquid-substrate interface and whether a liquid droplet will spread (Figure 3).^{15,16} This phenomenon can be measured by the contact angle (θ), where a contact angle that is 90° or higher indicates low wetting. When the contact angle is less than 90°, that means there is partial or high wetting which leads to spreading.



Figure 3. Adapted from Jarray, A. et al.¹⁶ Schematic of a droplet wetting on substrate illustrating low and partial wetting. Also distinguishes surface tension from wettability.

Levelling occurs at the liquid-air interface and can be measured by surface tension (Figure 3). Surface tension refers to the energy required to increase the surface area of a liquid, or more generally, the force required to keep a liquid together as a droplet.^{17,18} A surfactant's ability to reduce surface tension is measured by how well it reduces the surface tension of water, which has one of the highest surface tensions for a liquid at 72 mN/m.¹⁹

The various interactions involved in the ability of a liquid droplet to spread over a surface can be described by the Young equation: $cos(\Theta) = \frac{\gamma_{sv} - \gamma_{sL}}{\gamma_{LV}}$. The contact angle (θ) shown in Figure 3 is actually affected by three interfaces: liquid-surface (γ_{SL}), liquid-vapor (surface tension, γ_{LV}), and solid vapor (γ_{SV}).²⁰ However, since it can be difficult to reliably measure the individual γ_{LV} and γ_{SL} , we approximate that wettability at the surface-liquid interface is best described overall by contact angle, and we can use surface tension as a proxy for levelling performance.¹⁶ Ultimately, a good surfactant will be able to reduce the energy difference between the liquid-substrate interface to achieve good wetting, and the liquid-air interface to achieve good levelling.

Additional technical metrics that will help us assess surfactant candidates are the critical micelle concentration (CMC) and the octanol-water partition coefficient (K_{ow}). CMC is the concentration at which surface tension stops changing with surfactant concentration. It will be a helpful benchmark for us to compare the amount of surfactant that is needed to achieve minimum surface tension, where a small CMC value indicates that less surfactant is needed.^{21,22} The K_{ow} measures whether a substance has greater affinity to octanol, an oil or fatty phase, or water, which can tell us about a substance's ability to repel oil, known as oleophobicity.²³ According to an expert in the floor polish industry, the strong oleophobicity of fluorochemicals potentially drives a floor polish formulation's tolerance for extensive recoating, where it is typical to apply 4-5 coats of floor polish in one application.¹² Thus, we will use K_{ow} as a proxy for recoating potential.

Although we gathered information on the technical performance of PFAS, given that they often outperform other types of surfactants to an extreme degree, we will use sodium dodecyl sulfate (SDS) to represent a baseline performance for non-fluorinated surfactants. SDS is a commercially available anionic surfactant commonly used in cleaning and hygiene products, which will provide a helpful point of reference for our proposed alternative surfactants. We use the following color scheme and internal standards to summarize our technical assessment:

High Efficacy (H)	Moderate Efficacy (M)	Low Efficacy (L)
Water contact angle < 45°	45° ≤ Water contact angle ≤ 90°	Water contact angle ≥ 90°
Surface tension < 36 mN/m	36 mN/m ≤ Surface tension ≤ 54 mN/m	Surface tension > 54 mN/m
CMC: 2 - 7 mM	7 mM ≤ CMC ≤ 14 mM	CMC > 14 mM
LogK _{ow} < 2	2 ≤ LogK _{OW} < 5	LogK _{OW} > 5

We designated good wettability as less than 90°, and the best wettability occurs at less than 45°. Excellent leveling agents need to reduce the surface tension of water by more than 50%. Since most floor polishes use between 100-275 ppm, or 2 - 7 mM, of PFAS, a competitive surfactant should have a CMC within that range. K_{ow} is a unitless value commonly reported in logarithmic form (logK_{ow}) since K_{ow} spans many orders of magnitude. LogK_{ow} spans from -3 (very hydrophilic) to +10 (very hydrophobic).²⁴ We want an oleophobic, or essentially hydrophilic, compound, which means that smaller values indicate better recoating potential. In particular, we set a logK_{ow} of 5 as a threshold since that is a common designation for oleophilicity.²³ Although our internal standards may not completely predict an alternative surfactant's performance in floor polish, we use them as a framework to evaluate their potential.

GreenScreen score assignment for health and environmental endpoints

We evaluated the health and environmental endpoints for both existing PFAS used in floor polish and our proposed alternatives by first gathering information from authoritative lists on Pharos and filling in gaps with literature review and CompTox calculations. We standardized this initial evaluation using the Globally Harmonized System (GHS) of Classification and Labelling of Chemicals. Then we converted the GHS scores into GreenScreen assignments using a procedure adapted from GreenScreen for Safer Chemicals. Based on the risks from PFAS, we focused on the human and environmental endpoints that were most relevant in the entire lifecycle of our alternatives. When considering the disposal of our ultimate solutions, we wanted solutions that are biodegradable and will not persist in the environment, especially in an aquatic environment. We use the following color scheme to rate the hazard potential of the endpoints:

1	2	3	4	5	LC	PC	DG
Very					Low	Potential	
Hazardous	Hazardous	Moderate	Low	Very Low	Concern	Concern	Data gap

Low concern and potential concern meant we found very few studies on an endpoint and could not converge on a more specific assignment. A data gap corresponded with a complete lack of studies that directly related to the endpoint.

Health and environmental performance of PFAS in floor polish

We researched and analyzed the potential hazard concerns of floor polish products containing PFAS at each phase of the product life cycle, using this as our baseline for the health and environmental performance of existing floor polish formulations.



Figure 4. *Phases during which PFAS chemicals enter the environment and lead to human and environmental exposures. Source: Minnesota Pollution Control Agency*²⁵

Production of PFAS

The Environmental Protection Agency (EPA) recognizes that PFAS manufacturing facilities can release PFAS into the air and water, as demonstrated in Figure 4.²⁶ Many cities in the United States have found evidence of PFAS contamination in surface water, the sediment downstream of production facilities, and the effluent from wastewater treatment plants.³ While PFAS pollution during the production phase is one concern, there are also occupational exposure risks to workers in facilities manufacturing PFAS. According to the National Institute for Occupational Safety and Health, occupational exposure to PFAS during production differs from the general public since the exposure sources are less diverse with a relatively stable exposure duration.²⁷ Possible exposure routes include dermal contact of concentrated products or inhalation of PFAS from the workplace.

Application of floor polish with PFAS

Exposure to PFAS during floor polish application is of particular concern for janitorial workers who often apply floor polishes in commercial settings such as schools, hospitals, and retail and grocery stores. During application, there is potential risk for dermal contact and inhalation. A recent study by Zhou et al. looked into actual concentrations of PFAS in the air during and after floor polish application to assess the exposure of janitorial workers. They found estimated levels of PFAS in the air to be one order of magnitude greater during floor polish application than typical indoor air concentrations, suggesting a significant occupational health risk for those who apply floor polish as part of their profession.⁸ We may also interpret an additional risk for those working, visiting, or attending school in locations soon after floor polish is applied, particularly when it comes to sensitive populations such as young children or people visiting hospitals.

Disposal of floor polish containing PFAS

One critical route for PFAS-containing floor polish to end up in the environment is through the disposal of spent product. Using a bucket and a mop is reportedly the most common application method of floor polish, which can lead to unused spent product getting poured down the drain.⁸ Even if there is not any remaining product, just the process of washing off the tools used to apply the polish leads to PFAS going down the drain and entering wastewater treatment plants. Since wastewater treatment plants cannot sufficiently remove PFAS from water, it can then spread into the environment with the treated water. From here, aquatic toxicity is a concern, along with any potential contamination of drinking water. As PFAS can bioaccumulate, any substance that ends up in aquatic species will likely make its way up the food chain. There is also concern surrounding disposal of floor materials containing floor polish that are removed and sent to landfills, which over time could leach into the environment.²⁸

Strategy 1: Rhamnolipids

Inspiration

Biosurfactants are compounds produced by microorganisms to help them advantageously interact with and change the surface of their local environments.²⁹ Although the exact physical and chemical stressors that lead microorganisms to produce biosurfactants are still uncertain, these biosurfactants have garnered widespread interest for their utility.^{29,30} Biosurfactants are already commercially available and used in many industries including agriculture, pharmaceuticals, food, and cosmetics.^{31,32} Several properties that make them attractive for these very different applications include emulsification, cleansing, foaming, and dispersion.^{31,33,34} Additionally, some studies have shown that biosurfactants can degrade under aerobic and anaerobic conditions, which will mitigate their persistence in the environment.^{35,36}

One leading commercial biosurfactant in these various industries is a glycolipid biosurfactant called a rhamnolipid, which we believe holds potential as a levelling and wetting agent in floor polish.³¹ They are produced by the bacterial species *Pseudomonas (P.) aeruginosa* in which three enzymes are responsible for the synthesis of rhamnolipids. Rh1A catalyzes the production of the hydrophobic precursor for the rhamnolipid, while RH1B catalyzes the reaction between the fatty acid and rhamnose molecule to form a mono-rhamnolipid, which will be the main rhamnolipid of our discussion in this section.² Rh1C is a third enzyme that produces di-rhamnolipids by attaching a second glucose molecule. Innovative metabolic engineering research has focused on isolating the enzymes that synthesize rhamnolipids and expressing them in non-pathogenic hosts to improve the safety of rhamnolipid production.³⁷ There has also been substantial research exploring different feedstocks to serve as the substrate for the microbial fermentation (e.g., waste cooking oil, fats, oils, sugars) to create a closed production loop and explore how to manipulate rhamnolipid physicochemical properties.^{35,36}

Technical performance

Rhamnolipids are amphiphilic molecules that have affinities to both water (hydrophilic or oleophobic) and fats (oleophilic or hydrophobic). Rhamnolipids are composed of a sugar molecule (rhamnose) and fatty acid, where the sugar is the hydrophilic part, while the fatty acid chain is generally the hydrophobic end (Figure 5).³⁶ Rhamnolipids act as a surfactant where the hydrophilic end, or the sugar, orients toward the floor polish solution itself –since it is mainly composed of water– and the fatty acid tail orients away from the bulk floor polish solution.



Figure 5. Mono-rhamnolipid molecule where the rhamnose molecule (sugar) is enclosed in the blue box and the fatty acid chain is pointed out as the hydrophobic end. The R1 position is where a second rhamnose molecule can attach to form a di-rhamnolipid, and the R2 position can have fatty acid chains with varying lengths.³⁸

We compare the rhamnolipids performance to our baseline surfactant, SDS, according to the technical metrics described in the approach section (Table 1). We also include the technical metrics for the two most popular fluorosurfactants currently used in floor polish formulations that go by the trade names FS-60 and FS-65. We find rhamnolipids reduce surface tension down to a range of 26 - 29 mN/m, which is comparable to SDS. Notably, the CMC value for rhamnolipids (0.41mM) is much smaller than for SDS (8mM), indicating that we can achieve a relatively low surface tension with a smaller amount of rhamnolipids, while a much higher concentration of SDS is needed to achieve a similar surface tension value. It is also helpful to note that SDS has multiple CMC values but the lowest is 8 mM, and subsequent CMC values only increase from there.³³ Rhamnolipids have strong leveling potential, with surface tension values comparable to the fluorosurfactants (FS-60 and FS-65), and a CMC only two times larger than the fluorosurfactants and 20 times smaller than for SDS.

Technical properties	FS-60	FS-65	Sodium dodecyl sulfate (SDS)	Rhamnolipids		
Water contact angle (°) (Wettability)	M ~51-56° (similar products FS-61 & FS-63)		M ~51-56° (similar products FS-61 & FS-63)		H ~20° at 3.48 mM on PVC ³⁹	M ∼20° at 1.5 mM on PVC
Surface tension (mN/m) (Levelling)	H 19 at 0.05%	H 18 at 0.05%	H 23.8 (tail) and 34.6 (head) ⁴⁰	Н 26-29		
Critical micelle concentration (mM) (Surfactant concentration)	H 0.23 (for Capstone 1157)		M 8-8.5 ³³	H 0.41		
LogKow (octanol water partition coefficient) (Oleophobicity)	M 2.51 (0.276 - 5.99) (for N-EtFOSAA)		H 1.69 (calculated with KOWWIN QSPR) ⁴¹	L 5.77 (4.22 - 7.38)		

Table 1. Technical metrics of three classes of surfactants: fluorosurfactants FS-60 and FS-65, SDS, and our proposed alternative, rhamnolipids

High Efficacy (H)	Moderate Efficacy (M)	Low Efficacy (L)
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Contact angle strongly depends on substrate material. A fair evaluation of wettability potential requires comparing water contact angle measurements across the same substrates. Although floor polish is used on many types of floor material, we tried to find measurements on vinyl, or PVC, since large buildings like stores or schools may use PVC floors and polish them. Based on the information available, we can directly compare wettability potential between SDS and rhamnolipids on PVC. Both surfactants achieve contact angles as low as 20° at similar concentrations, which indicates excellent wettability. However, surfactant concentration also influences the contact angle as seen in Figure 5. Rhamnolipids demonstrate smaller contact angles at almost all concentrations compared to SDS, but the contact angles vary widely from $20^{\circ} - 45^{\circ}$.³⁹ If we use the rhamnolipid's CMC, which is about 260.3 mg/L, the contact angle would increase to about 30-37° (Figure 6). This means that rhamnolipids achieve their best wettability at concentrations larger than its CMC. Rhamnolipids may perform well as both a wetting and levelling agent as indicated by its excellent surface tension and contact angle on PVC; however, testing will be required to find the rhamnolipids concentration that optimizes both functions.



Figure 6. Relationship between contact angle and concentrations of rhamnolipids and SDS³⁹

Finally, the rhamnolipid has a $\log K_{ow}$ of 5.77, which means it is more hydrophobic, suggesting that rhamnolipids favor oil and are not very oleophobic. Thus, if we use oleophobicity to predict a rhamnolipid's ability to assist in recoating, it would not recoat floor polish well and perform more poorly than SDS.

Changing the salt counterion or concentrations to tune surfactant performance

A notable strategy to tune the chemical properties of surfactants is to change the salt counterion or concentration. For example, PFOA without a salt yields an effective surface tension that is less than half of the value of PFOA with a sodium counterion, and PFOS with a potassium salt produces the lowest surface tension compared to other salt counterions (e.g., lithium, ammonium).⁴³ Meanwhile, varying the concentration of lithium chloride mixed with SDS manipulates the resulting contact angle of SDS.⁴⁴ There are even studies exploring how salt concentration impacts a rhamnolipid's ability to lower surface tension.^{42,45} Figure 7 illustrates that for all mixtures of rhamnolipids with different salt (NaCl) concentrations, surface tension decreases as the rhamnolipid concentration increases.⁴² Once the rhamnolipid concentration reaches its CMC, surface tension does not change. In particular, increasing the NaCl concentration up to 40 g/L consistently decreased the overall surface tension value ranges (black, red, and blue curves in Figure 7). Ultimately, this systematic change in sodium chloride concentration demonstrates that rhamnolipid surfactant properties are tunable.



Figure 7. Change in surface tension as a function of rhamnolipid concentration (c mg/L). Different colors correspond with different salt (NaCl) concentrations mixed with rhamnolipids.⁴²

Technical summary

Rhamnolipids may meet the technical performance requirements for floor polishes. Rhamnolipids generate a surface tension that is comparable to SDS and within the range of fluorosurfactants, all at a much smaller concentration than SDS. Rhamnolipids could potentially replace fluorosurfactants as levelling agents in floor polish, particularly because they may also perform well as wetting agents on a PVC surface. However, they are considered rather oleophilic, which suggests poor recoatability potential. We suggest tuning rhamnolipid properties by changing the salt counterion or concentration. While we can try to predict the performance of rhamnolipids as a surfactant in floor polish, its overall effectiveness will best be determined when combined with other elements of a floor polish formulation. We also do not know how well it will perform on different floor substrates. Despite these unknowns, rhamnolipids show potential as a replacement for fluorosurfactants.

Health and environmental performance of rhamnolipids in floor polish

We conducted a life cycle analysis of rhamnolipids in the context of a floor polish product to evaluate its environmental and health outcomes. We then compared the level of concern over rhamnolipids in each stage with the level of concern over PFAS in floor polish. While there is not a vast array of literature concerning the potential health and environmental hazards of rhamnolipids, there is enough evidence to convince us of the expected safety of the compound.

Production

Rhamnolipids demonstrate a lower risk than PFAS compounds in floor polish products across six hazard categories denoted by a low to very low hazard score in our hazard table (Table 2). In particular, there is convincing evidence that most Group I and II endpoints have low concern despite data gaps (Table 2). Patowry *et al.* tested the cytotoxicity of rhamnolipids in mouse cell lines and did not find any cytotoxic effect.⁴⁶ Semkova *et al.* found that rhamnolipids assisted a traditional chemotherapy drug to induce cell damage or death in cancer cells, and posed that rhamnolipids may show promise as a natural anticancer agent.⁴⁷ Overall, rhamnolipids present little concern for production workers who may be exposed to rhamnolipids through dermal contact or inhalation.

While rhamnolipids present a low concern regarding health impacts, their source material, *Pseudomonas (P.) aeruginosa*, presents possible risks during rhamnolipid production. *P. aeruginosa* is an opportunistic infection bacteria that produces a large number of extracellular toxins.⁴⁸ Although there is a potential health risk of occupational exposure to *P. aeruginosa* during production, this risk can be reduced through proper hygiene protocols and personal protective equipment use.⁴⁹ Furthermore, expressing enzymes for rhamnolipid production in non-pathogenic hosts is currently being researched and will also decrease occupational exposure risk.³⁷

Another production concern focuses on the use of hazardous solvents like chloroform to increase the yield of rhamnolipids, which leads to potential employee exposure risks.⁵⁰ However, replacing problematic solvents with greener solvents and optimizing the rhamnolipid extraction and purification process will reduce reliance on and risk of hazardous solvents.^{51,52} Despite human health concerns present during the production of rhamnolipids, clear and growing strategies to address these concerns exist. Additionally, the overall production of rhamnolipids is more environmentally friendly since the process requires less energy than other biosurfactants.⁵³

Application

During floor polish application, there is risk for both acute and chronic exposure. Acute exposure can occur from any accidental spills, while chronic exposure could come from repeated contact with floor polish through either dermal exposure or inhalation. As mentioned above, janitorial staff are particularly at risk, and can be exposed to various PFAS compounds from airborne particles measured during floor polish application.⁸ While safety policies can mitigate PFAS exposure for both janitorial staff and surrounding people (e.g., personal protective equipment, application in empty spaces), we drastically reduce these hazards through using alternative surfactants such as rhamnolipids.⁵⁴ Rhamnolipids display low to very low hazard concern in most of the endpoints considered, including carcinogenicity, mammalian toxicity,

systemic toxicity, respiratory toxicity, and skin irritation. Rhamnolipids do present a higher risk with eye irritation, which is concerning for the application stage, however we suggest the use of personal protective equipment during application to mitigate this risk.

Disposal

As mentioned previously, spent floor polish may enter water systems, which leads to concerns about aquatic toxicity and persistence. PFAS compounds are difficult to remove from the environment, are not fully removed by wastewater treatment, and do not break down easily because of their strong carbon-fluorine bonds. In contrast, we found that rhamnolipids biodegrade under both aerobic and anaerobic conditions and therefore likely would be removed by wastewater treatment plants as they create either aerobic or anaerobic conditions during treatment.^{55,56} This removal presents less of a risk for any unforeseen downstream consequences. Furthermore, rhamnolipids enhance the biodegradation of other pollutants, remove heavy metals, and also help wet soil.^{36,57,58} One remaining area of concern is the potential for rhamnolipids to inhibit algae growth and disrupt soil microbial activity.⁵⁹ There is also minor concern regarding the bioaccumulation potential of rhamnolipids; however, this is a modeled estimate based on the high logK_{ow}, so we would like to see experimental data to confirm the modeled data on bioaccumulation.^{41,60}

The Danish EPA listed rhamnolipids as Category 2 (using GHS criteria) for chronic aquatic toxicity based on modeled results.⁶¹ However, in 2016 Johan *et al.* conducted a full battery of ecotoxicity tests of mono-rhamnolipids, and found low acute toxicity based on fish embryo acute toxicity testing, no mutagenic or estrogenic effects based on the Ames fluctuation assay and in silico modeling, and lower aquatic toxicity than chemical surfactants but slightly higher than other biosurfactants based on immobility testing of daphnids.⁶² Based on the expected concentration of rhamnolipids in cleaning products that could end up in waterways, they conclude that there is minimal toxicity concern.

Rhamnolipids hazard assessment

		N-EtFOSAA	PFBS	Rhamnolipids
	Carcinogen	3	3	5
Group I	Mutagen	LC	LC	4
endpoints	Repro/Dev	DG	2	DG
	Endocrine disruptor	DG	1	DG
	Mammalian Toxicity	4*	2	4
	Systemic Toxicity	DG	2	LC
Group II endpoints	Neurotoxicity	DG	3	DG
	Respiratory Toxicant	DG	PC	4
	Skin irritation	РС	Ι	4
	Eye irritation	DG	2	I
Eco toxicity	Aquatic Toxicity	2*	3	РС
Fata	Persistence	<i>I</i> *	1	5
Ган	Bioaccumulation	PC*	PC	PC

	Table 2.	Rhamnolipid	s hazard assessmer	t with fluoros	urfactants N-EtF	OSAA and PFBS
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1	2	3	4	5	LC	PC	DG
Very Hazardous Hazardous Moderate Low Very Low Low Concern Data g						Data gap	
<i>Italicized = potential score;</i> * = prediction based on similar compound N-EtFOSAA represents a baseline fluorosurfactant in floor polish, and PFBS is its degradant.							

Strategy 2: Amino Acid Surfactants - Sodium Lauroyl Glutamate

Inspiration

Amino acid surfactants are our second strategy for biodegradable commercial surfactants and are currently used in other industries such as cosmetics, cleaning products, and biomedical applications.⁶³ Amino acids form the building blocks for proteins in all living organisms, and the basic structures contain a carboxyl group, amine group, and a distinct variable group (Figure 8).⁶⁴ The variable group grants each amino acid unique chemical properties, such as charge (e.g. anionic, cationic), as demonstrated in Figure 8. Amino acid surfactants form through the condensation of natural amino acids with fatty acids derived from an oleochemical source.¹ We focus on anionic surfactants since their negative charge better allows the molecule to lift and suspend dirt, which is helpful for floor polishes to resist dirt.⁶⁵



Figure 8. General structure of amino acids on the left, where the variable group imparts unique functionality to the amino acid. Aspartic and glutamic acid are anionic amino acids, and glutamic acid is a precursor to sodium lauroyl glutamate.⁶⁴

Sodium lauroyl glutamate (SLG), an anionic amino acid surfactant, is produced via the Schotten-Baumann reaction, which involves the acylation of a free amine of glutamic acid with an acyl chloride.⁶⁶ SLG demonstrates high surface activity, shows low toxicity, and is predicted to have quick biodegradation based on its relatively simple structure (Figure 9).⁶⁷ Although current production methods for amino acid surfactants rely on solvents to increase yield at a lower cost, there is potential for greener production through enzyme catalysis.⁶⁸ Additionally, exploration into more sustainable surfactant production focuses on using renewable and raw feedstock like waste proteins and waste cooking oils to form the fatty acid chain precursor.³⁶

Technical performance

Sodium lauroyl glutamate (SLG) is an amphiphilic molecule with an affinity for both water and fats, where the carboxylate part is hydrophilic, and the fatty acid chain is the hydrophobic (or oleophilic) end (Figure 9). Due to its amphiphilic nature, SLG can perform as a

surfactant where the hydrophilic end will orient toward the floor polish solution that is mainly composed of water, and the fatty acid tail will orient away from the bulk floor polish solution.



Figure 9. Sodium lauroyl glutamate molecule where the hydrophilic component is enclosed in the blue box, and the fatty acid chain is outlined as the hydrophobic end.

For our technical assessment, we use three baseline surfactants to compare with SLG. Our baseline fluorosurfactants, FS-60 and FS-65, are commonly used in floor polish solutions. We also have the non-fluorinated baseline surfactant, sodium dodecyl sulfate (SDS). Comparing the wettability of SLG to the baseline surfactants is difficult since we could not find water contact angle studies with vinyl (PVC) surfaces for all the surfactants. With this limited information, we initially predict that SLG may be a poor wetting agent due to its large contact angle on sericite substrate.⁶⁹ However, we recommend following up on the wettability of SLG on more relevant surfaces given the limited information currently available.

SLG reduces surface tension to less than 30 mN/m, which is effectively reducing the surface tension of water (72 mN/m) by more than 50%.⁷⁰ Although fluorosurfactants achieve the lowest surface tensions, that alone does not consistently predict overall leveling performance.¹⁴ Thus, we determine that SLG has high potential for performance as a levelling agent given that it exceeds our internal metric for surface tension performance and is comparable to SDS. Furthermore, we find that the concentration of SLG needed to achieve the minimum surface tension, measured by critical micelle concentration (CMC), is very low and comparable to the fluorosurfactants' CMC.⁷⁰ Finally, we could not find the logK_{OW} values for sodium lauroyl glutamate and used sodium cocoyl glutamate as a proxy molecule. Sodium cocoyl glutamate's logK_{OW} is much smaller than 5, which means that it is very oleophobic and suggests that it has good recoatability potential.⁷¹

Table 3. Technical comparison of three classes of surfactants: fluorosurfactants FS-60 and FS-65, sodium dodecyl sulfate (SDS), and representative amino acid surfactant sodium lauroyl glutamate (SLG)

Technical properties	FS-60	FS-65	Sodium d	lodecyl	Sodium lauroyl		
Water contact angle (°) (Wettability)	M ~51-56° (similar products FS-61 & FS-63)		M ~51-56° (similar products FS-61 & FS-63)		H ~20° at 3. on PV	48 mM ′C ³⁹	L 138.69° on sericite (mineral) ⁶⁹
Surface tension (mN/m) (Levelling)	H 19 at 0.05%	H H 23.8 (tail) and 34.6 (head) ⁴⁰		l) and ead) ⁴⁰	H <30 ⁷⁰		
Critical micelle concentration (mM) (Surfactant concentration)	H 0.23 (for Capstone 1157)		M 8-8.5	5 ³³	H 0.48 ⁷⁰		
LogKow (octanol water partition coefficient) (Oleophobicity)	M 2.51 (0.276 - 5.99) (for N-EtFOSAA)		H 1.69 (calculated with KOWWIN QSPR) ⁴¹		H 0.597 (for sodium cocoyl glutamate) ⁷¹		

Gemini surfactants

One avenue to tune the physicochemical properties of SLG and other amino acid surfactants is to link two identical surfactant molecules together with a spacer chain, as shown in Figure 10.⁶³ Amino acid gemini surfactants appear to not be widely used in commercial applications yet, and a gemini surfactant with SLG base units has not yet been synthetically explored. However, there is growing research on amino acid gemini surfactants since they offer unique benefits to traditional gemini surfactants including preparation from renewable resources, possibly low ecotoxicity and cytotoxicity, and molecular tunability.⁷²



Figure 10. Example of a cationic gemini surfactant composed of two surfactants from arginine sources. The alkylene diamine spacer chain is enclosed by a blue box and can have varying carbon atom length.⁷²

Gemini surfactants exhibit critical micelle concentrations up to two orders of magnitude lower than their single surfactant counterparts.⁷³ This means that a smaller concentration of a gemini surfactant is needed to achieve its lowest surface tension, which can result in production cost savings. However, their impact on absolute surface tension values are variable and dependent on the base surfactant molecule.^{73,74} While gemini surfactants more efficiently reduce surface tension, their overall surface tension values at critical micelle concentrations still range from 30 - 34 mN/m, which is not that different from our proposed SLG surfactant.^{72,75,76} Although gemini surfactants cannot achieve a surface tension as low as a fluorosurfactant, that alone is not predictive of the final performance of amino acid gemini surfactants in floor polish. The gemini surfactant form still influences surface activity compared to its monomer form.

Other components of the gemini surfactant can further tune surfactant properties. Some gemini surfactants with a short chain exhibit high viscosity at a low concentration, which may be undesirable for floor polish applications and a property to note for caution.⁷³ A study on the length of the hydrophobic fatty acid chain illustrated that increasing the carbon length from 6 to 14 carbons corresponds with a decrease in the critical micelle concentration to achieve their lowest surface tension values (Figure 11).⁷⁵



Figure 11. Relationship between surface tension and anionic gemini surfactant concentration with different fatty acid chain lengths (corresponding with different markers: upside down triangles = 6 carbons and right-side up triangles = 14 carbons)⁷⁵

While there are some concerns that the increased chemical stability of gemini surfactants may result in poorer biodegradation, there are a growing number of studies that explore building biodegradation into these molecules. Gemini surfactants that are sourced from arginine show better biodegradation and lower aquatic toxicity than bis quaternary ammonium halide surfactants and monomeric counterparts.^{63,72} Additionally, synthetically incorporating a spacer chain that is easily breakable reduces persistence.⁷⁷

Overall, gemini surfactants have garnered significant attention due to drastic changes in their surfactant properties by combining two single surfactant units together. Most gemini surfactants that have been investigated are cationic due to easier synthetic manipulation, so exploration of anionic gemini surfactants have potential for growth.^{75,77} In general, most gemini surfactants have been studied for their micelle formation to apply toward cleaning applications (e.g. home, pharmaceuticals, oil recovery), and there is opportunity for further studies on other surfactant properties that may be more relevant for floor polish applications.

Technical summary

Sodium lauroyl glutamate (SLG) is a representative anionic amino acid surfactant that can potentially replace fluorosurfactants in floor polish. SLG produces a surface tension comparable to SDS at a very small surfactant concentration. SLG shows high oleophobicity indicating good recoatability potential. Although SLG initially appears to not be a strong wetting agent, more wettability studies on relevant floor surfaces will clarify SLG's potential.

Single amino acid surfactants can also be combined to form gemini surfactants. They have very low critical micelle concentrations to achieve their surface tensions. Investigating further use of this class of surfactants may help amino acid surfactants with their final performance in floor polish formulations. While we focused on SLG for our technical comparison, there is a large variety of amino acid surfactants available. The abundance provides opportunities to test different amino acid surfactants but also means that it can be harder to find or conduct studies to provide fair comparisons between different surfactant choices.

Health and environmental performance of sodium lauroyl glutamate in floor polish

Similar to our assessment of rhamnolipids, we considered the health and environmental impacts of using sodium lauroyl glutamate (SLG) in floor polish during each life cycle stage of the product, from production to use to disposal. SLG seems to be safer than PFAS at each stage of the lifecycle, which is consistent with its listing on the EPA's Safer Chemical Ingredients List.⁷⁸

Production

During production, relevant exposure pathways for production workers include inhalation and dermal exposure. When it comes to the health hazards during production, we are concerned by SLG's possibly high skin and eye irritation potential as identified by the European Chemicals Agency (ECHA), particularly in the higher doses expected by workers in production facilities.⁷¹ Although we expect to see some degree of irritation with surfactants because they reduce the surface tension of and disrupt the cell's lipid membrane, we suggest protocols for personal protective equipment to minimize skin and eye irritation.⁷⁹ Further identification of relevant concentrations will also determine safety protocols, as there are some conflicting studies and statements supporting SLG's low irritation potential.^{70,80–82} Group 1 endpoints shown in Table 4 are important to consider when it comes to the health of production workers since they often experience chronic exposure in varying levels to the substances they produce. There is potential concern over reproductive and developmental toxicity of SLG, because the Danish EPA Advisory List determined that SLG is suspected of damaging the fertility of an unborn child.⁸³ However, this evaluation was based on a modeled result, so more experimental studies on SLG's impact on reproductive and developmental toxicity will be helpful.⁸³ Additionally, SLG exhibits low concern as a mutagen since it did not show reactivity to the Ames test.⁶⁶ This is important because production workers can be exposed to higher amounts of the material than consumers over long periods of time, so any signs pointing to carcinogenic potential would be concerning.

Application

We previously established that janitorial workers who apply floor polish regularly carry the largest health burden. Routes of exposure during floor polish application include dermal contact and inhalation. Skin and eye irritation are a potential concern for the person applying the floor polish; however, these hazard endpoints are less concerning since production workers are more likely to encounter the high concentrations that would induce severe skin and eye irritation. SLG is used safely in cosmetics, so small concentrations likely do not lead to severe irritation.

There is concern over both acute effects from any accidental exposure during the use phase, as well as chronic, low level exposure. SLG appears to not have significant concerns for Group I endpoints that can be associated with chronic exposure, as seen in Table 4. However, there are also substantial data gaps regarding the long-term human health impacts from exposure to SLG. Further research that clearly defines and evaluates the safety or hazards of SLG will be valuable.

Disposal

Aquatic toxicity, persistence, and bioaccumulation are important to consider given the possibility of floor polish materials getting washed down the drain. SLG provides a substantial improvement over PFAS compounds because SLG readily biodegrades and poses a low concern regarding aquatic toxicity.⁸⁴ We would, however, feel more confident with additional publicly available peer-reviewed studies.

Sodium lauroyl glutamate hazard assessment

Table 4. Comparative hazard assessment between current PFAS used in floor polish and it's degradant and sodium lauroyl glutamate

		N-EtFOSAA	PFBS	Sodium lauroyl glutamate
	Carcinogen	3	3	DG
Group I	Mutagen	LC	LC	LC
endpoints	Repro/Dev	DG	2	PC
	Endocrine disruptor	DG	1	DG
	Mammalian Toxicity	4*	2	5
	Systemic Toxicity	DG	2	LC
Group II	Neurotoxicity	DG	3	DG
endpoints	Respiratory Toxicant	DG	PC	LC
	Skin irritation	PC	1	2
	Eye irritation	DG	2	2
Eco toxicity	Aquatic Toxicity	2*	3	LC
Fato	Persistence	1*	1	5
Fate	Bioaccumulation	PC*	PC	DG

1	2	3	4	5	LC	PC	DG
Very Hazardous	Hazardous	Moderate	Low	Very Low	Low Concern	Potential Concern	Data gap
<i>Italicized = potential score;</i> * = prediction based on similar compound N-EtFOSAA represents a baseline fluorosurfactant in floor polish, and PFBS is its degradant.							

Additional Strategies Considered

Alkyl polyglucosides

Alkyl polyglucosides are non-ionic surfactants. Similar to rhamnolipids and amino acid surfactants, alkyl polyglucosides have a hydrophobic fatty acid chain that can be sourced from raw materials like vegetable or waste oil and starch.⁸⁵ Like the rhamnolipid, the alkyl polyglucoside's hydrophilic component is a sugar molecule (glucose, Figure 12).



Figure 12. *Alkyl polyglucoside structure where the bracket contains the glucose molecule and the fatty acid chain is* $(CH_2)_n CH_3$.

Alkyl polyglucoside surfactants are used in a wide variety of applications such as cleaning, cosmetics, and agriculture due to their wettability, emulsification performance, and biodegradation.⁸⁶ Alkyl polyglucosides also present a low toxicity concern as they are on the EPA's Safer Chemical Ingredient List.⁸⁷ Commercial alkyl polyglucosides, such as the Dow Triton CG-425, reduces surface tension down to 28.8 mN/m at a critical micelle concentration of 61 ppm, which is about nine times higher than fluorosurfactant concentrations in floor polish solutions.⁸⁸ We strongly considered alkyl polyglucosides as an alternative strategy; however, they did not perform well in floor polish solutions according to our industry consultant.¹²

Siloxanes

Siloxanes, also known as silicones, are utilized in multiple industries as a PFAS replacement due to their effective performance as an oil and water repellent and as a surfactant. They have even been considered in floor polish formulations and performed fairly well.¹² According to a patent for use of siloxanes in floor polish, ionic silicone surfactants have good levelling and wetting properties and maintain resistance to soiling, which is an important element of floor polish.⁸⁹ Siloxane contains a silicon and oxygen molecule and is capable of forming multiple different polymers that all have differing properties.⁹⁰

However, while some scientists argue that they are safe, siloxanes have been recently scrutinized for their health and environmental hazard potential. In 2018, three siloxane monomers were added to the EU's List of Substances of Very High Concern due to their persistence in the environment, bioaccumulation, and toxicity.⁹¹ Some siloxanes have been associated with endocrine disruption and reproductive effects, which is concerning.⁹² There is a possibility that only certain siloxanes have a higher human hazard potential depending on the

degree of polymerization and the chemical structure.⁹² Unfortunately, the argument for siloxane's strong technical performance and the tradeoff between structural stability and environmental persistence is reminiscent of previous justifications of PFAS use, which is why we did not pursue siloxanes further.⁹³

Waxy plant leaves

In addition to biosurfactants and synthetic surfactants, we also explored naturally occurring substances from plants that demonstrated desirable properties for floor polish. In our initial bio-inspiration search, tropical plant leaves caught our attention for their glossy and waxy surfaces and water-repellent properties. We investigated several plant leaves and explored their usability for floor polish applications. Ultimately, we did not propose plant leaves or their chemical constituents as a direct PFAS alternative due to challenges in production, scalability, and the fact that it does not replace the exact function of PFAS in floor polish, which is how we ultimately decided to limit our project scope.

The first floor waxes came from carnauba wax and are still commercially used. Fatty acid esters (80-85%) and fatty alcohols (10 - 16%) compose the majority of the chemical mixture of carnauba wax.⁹⁴ It stands out from other natural waxes due to its high melting point (78-85°C), glossy finish, and strong abrasive resistance.⁹⁵⁻⁹⁷ However, the carnauba palm only grows in Brazil, so we had concerns about sustainable harvesting. Additionally, the application of floor polish products with carnauba wax is labor intensive.

We investigated banana leaves as another natural wax source. They demonstrate properties comparable to carnauba wax, especially a high melting point.⁹⁸ Thus, commercializing banana leaves could increase the amount of natural wax used in products. However, the properties may vary with banana leaf genotype due to different leaf wax compositions.



Figure 13. (*a*) Image of lotus leaf and (b) the hierarchical micro/nanostructures composed of papillose epidermal cells responsible for the lotus leaf's extreme wetting surface⁹⁹

Lotus leaves are extremely hydrophobic and exhibit extreme wetting capabilities due to papillose epidermal cells enveloped in hydrophobic epicuticular wax.⁹⁹ The epidermal cells are composed of randomly oriented rough structures observed at the nano and micro-scales (Figure

13). These structures provide air-pockets and result in a rough interface between a water droplet and the leaf surface so that water runs off very easily.⁹⁹ The lotus leaf architecture serves as an engineering inspiration to implement nano-textured surfaces such as in the floor substrates themselves or with the application of the floor polish solution.

Beyond fluorosurfactant substitution

While it is necessary to replace PFAS in floor polish, there are other potentially hazardous ingredients in many floor polish formulations. These range from VOCs to harmful preservatives to styrene copolymers.⁸⁹ Using products with those chemicals could lead to additional occupational, consumer, and environmental exposure risks. Exposure to other ingredients in floor polish is associated with health outcomes such as cancer, brain damage, birth defects and asthma. Therefore, the issue of finding safe and eco-friendly chemicals in floor polish products should not be limited to PFAS substitutes only.

Furthermore, we have considered whether floor polish use can be eliminated in some settings. While floor polish makes floors more durable, we can consider floor material that is sustainable, durable, and does not require polishing. Syracuse University recently installed a number of spaces on their campus with rubber flooring that is easy to clean, visually appealing, and does not require floor polish.¹⁰⁰ Other schools, tired of the yearly process of stripping and repolishing floors, are also looking into alternative floor materials.⁵⁴

Recommendations

Technical comparison of rhamnolipid and SLG surfactants

We found that rhamnolipids and sodium lauroyl glutamate (SLG) both have technical potential to replace fluorosurfactants in floor polish formulations. Both alternatives can reduce the surface tension of water by more than 50% at a concentration comparable to fluorosurfactants. Rhamnolipids display strong wettability performance with a low contact angle on PVC. It will be helpful to further study the wettability potential of both rhamnolipids and SLG on a larger variety of surfaces that are relevant for floor polish applications. We predict that SLG may have better recoatability potential than rhamnolipids, but the best way to evaluate recoatability will be to test the performance of these alternative surfactants when mixed into a floor polish solution.

We outlined two strategies for tuning the properties of rhamnolipids and amino acid surfactants: changing the salt counterion or concentration and using gemini surfactant forms. Another strategy that may be useful for tuning surfactant properties is to mix different surfactants together. Synergistic interactions can occur in surfactant mixtures, which often lead to smaller surface tensions and critical micelle concentrations than single component counterparts for rhamnolipids, amino acids, and gemini surfactants.^{101–105} For example, surface tension is drastically lowered to below 1 mN/m when combining rhamnolipids with a synthetic non-fluorinated surfactant.⁶⁷ Combining surfactants also tunes other surface activity such as micelle formation.^{102,104} Overall, a possible strategy for overcoming technical deficits with one surfactant may be to add another surfactant.

Hazard assessment comparison of rhamnolipid and SLG surfactants

Rhamnolipids and SLG demonstrate low hazard concerns across many endpoints, especially compared to PFAS compounds. Most importantly, both alternatives degrade and do not persist. SLG possibly exhibits low concern for aquatic toxicity, while rhamnolipids may have slightly higher potential to pose a hazard to aquatic life. However, both assessments were based on limited studies and modeled data, so we advocate for more studies on the aquatic toxicity of these surfactants. In particular, this information will reconcile our understanding for the ecotoxicity of rhamnolipids, which are commonly employed to break down pollutants in waterways.

For the human health endpoints, rhamnolipids clearly pose minimal concern across Group 1 and 2 endpoints, while SLG possibly presents low concern but requires more studies. The main concerns for the production of rhamnolipids center on the use of a bacterial host and hazardous solvents. However, there are clear strategies and alternatives (e.g. safety protocols, green solvents) that reduce occupational exposure risk during production. Skin and eye irritation remain as the endpoints of concern for people in both the production and use stages of the surfactants. Some degree of irritation is expected with surfactants since they disrupt the cell's lipid membrane. Distinguishing the concentrations required for irritation and floor polish use will be helpful in establishing safety procedures and personal protective equipment for the production and application of floor polish containing these surfactants.

Additional research on the carcinogenicity, reproductive and developmental toxicity, endocrine disruption, and neurotoxicity will provide more information on data gaps and increase our confidence in score assignments. We encourage studies to provide more detailed information on the exposure routes to better evaluate hazards for surfactants and implement safety measures.

 Table 5. Final summary of our technical, health and environmental assessment of our two

 proposed solutions, rhamnolipids and sodium lauroyl glutamate

	Technical	Human Health	Environmental
Rhamnolipids	 Strong levelling and wetting potential More available studies Broad commercial applications 	 Low concern across Group I and II human health endpoints Data gaps in literature 	 More studies on environmental outcomes demonstrating safety Biodegrades and degrades other pollutants Aquatic toxicity
Sodium Lauroyl Glutamate	 Strong potential for levelling and recoatability Not used as widely in industry 	 On EPA's List of Safer Chemical Ingredients Data gaps in literature 	 Appears safe and is able to biodegrade Insufficient studies

Outlook

Although both rhamnolipids and SLG are technically comparable, rhamnolipids are commercially used in a larger variety of industries than amino acid surfactants, so they may be a good first choice to investigate as an alternative surfactant in floor polish. Furthermore, we found a larger number of studies that evaluate the surfactant properties and hazard outcomes of rhamnolipids. While amino acid surfactants are growing in commercial applications, finding comprehensive studies and information that are specific to an amino acid surfactant can be challenging given the vast diversity of possible amino acid surfactants.

Beyond our initial exploration into these alternative surfactants, we anticipate that a few questions regarding the performance of rhamnolipids will be clarified when mixed into floor polish solution and tested. First, the necessary concentration for rhamnolipids will need to be determined. Ideally, a low concentration reduces production costs and minimizes modifications of the final floor polish properties.¹² Additionally, lab tests will need to evaluate the effectiveness of floor polish solutions with rhamnolipids on different floor materials. Finally, a longer-term direction could entail pursuing one of the surfactant optimization strategies we outlined earlier, such as salt counterion, gemini surfactants, or mixing surfactant systems together.

Appendices

Appendix A: Project Team Members

Yuning Xu is a second year MPH student in the Environmental Health Sciences program with a concentration in industrial hygiene. She has experience working on a project related to PFAS policy in waste water while applying the p-sufficient approach.

Tessa Wardle is a second year MS student in the Environmental Health Sciences program of UC Berkeley's School of Public Health. She has a background in assessing the health consequences of toxic chemicals, and is familiar with the current crisis of widespread PFAS contamination. In the past she has worked on conducting an extensive literature review of nitrate and arsenic effects on birth outcomes, examining industrial facilities chemical releases, and understanding the health implications of exposure to multiple different pesticides.

Jenna Tan is a fifth year PhD chemistry student whose research focuses on the crystallization of organic molecules for semiconductor applications. She will use her chemistry background to help understand and evaluate the molecular properties of PFAS and the functional alternatives.

		N-EtFOSAA	PFBS	PFOS	Rhamnolipid	Sodium lauroyl glutamate	
	CAS #	67584-51-4	375-7 3-5	1763-23- 1	4348-76-9	29923-31-7	
Group I endpoints	Carcinogen	3	3	2	5	DG	
	Mutagen	LC	LC	3	4	LC	
	Repro/Dev	DG	2	1	DG	PC	
	Endocrine disruptor	DG	1	1	DG	DG	
	Mammalian Toxicity	4*	2	4	4	5	
	Systemic Toxicity	DG	2	1	LC	LC	
Group II	Neurotoxicity	DG	3	1	DG	DG	
enapoints	Respiratory Toxicant	DG	PC	PC	4	LC	
	Skin irritation	PC	1	LC	4	2	
	Eye irritation	DG	2	2	1	2	
Eco toxicity	AquaticEco toxicityToxicity		3	2	PC	LC	
Fate	Persistence	1*	1	1	5	5	
	Bioaccumulati on	PC*	PC	1	PC	DG	

Appendix B: Complete comparative hazard table

1	2	3	4	5	LC	PC	DG			
Very Hazardous	Hazardous	Moderate	Low	Very Low	Low Concern	Potential Concern	Data gap			
<i>Italicized = potential score;</i> * = prediction based on similar compound N-EtFOSAA represents a baseline fluorosurfactant in floor polish, and PFBS is its degradant.										

Appendix C: Hazard assessment references

We acknowledge the <u>PFAS-Tox Database</u> as an extremely helpful resource to find relevant toxicological studies for PFAS compounds.

N-EtFOSAA

- 1. Shearer, J. J. *et al.* Serum Concentrations of Per- and Polyfluoroalkyl Substances and Risk of Renal Cell Carcinoma. *J Natl Cancer Inst* **113**, 580–587 (2020).
- 2. Cohn, B. A. *et al.* In utero exposure to poly– and perfluoroalkyl substances (PFASs) and subsequent breast cancer. *Reproductive Toxicology* **92**, 112–119 (2020).
- 3. Product-Chemical Profile for Food Packaging with PFASs: Discussion Draft. 184.
- 4. Preston, E. V. *et al.* Maternal Plasma per- and Polyfluoroalkyl Substance Concentrations in Early Pregnancy and Maternal and Neonatal Thyroid Function in a Prospective Birth Cohort: Project Viva (USA). *Environ Health Perspect* **126**, 027013 (2018).
- 5. Cardenas, A. *et al.* Plasma Concentrations of Per- and Polyfluoroalkyl Substances at Baseline and Associations with Glycemic Indicators and Diabetes Incidence among High-Risk Adults in the Diabetes Prevention Program Trial. *Environ Health Perspect* **125**, 107001 (2017).
- 6. Zhang, C. *et al.* A Prospective Study of Pre-pregnancy Serum Concentrations of Perfluorochemicals and the Risk of Gestational Diabetes. *Fertil Steril* **103**, 184–189 (2015).
- 7. Xiao, C. *et al.* Associations of Exposure to Perfluoroalkyl Substances With Thyroid Hormone Concentrations and Birth Size. *J Clin Endocrinol Metab* **105**, 735–745 (2019).
- 8. Preston, E. V. *et al.* Associations of Per- and Polyfluoroalkyl Substances (PFAS) With Glucose Tolerance During Pregnancy in Project Viva. *J Clin Endocrinol Metab* **105**, e2864–e2876 (2020).
- Berk, M. *et al.* Pop, heavy metal and the blues: secondary analysis of persistent organic pollutants (POP), heavy metals and depressive symptoms in the NHANES National Epidemiological Survey. *BMJ Open* 4, e005142 (2014).
- Lyall, K. *et al.* Prenatal Maternal Serum Concentrations of Per- and Polyfluoroalkyl Substances in Association with Autism Spectrum Disorder and Intellectual Disability. *Environ Health Perspect* 126, 017001 (2018).
- 11. Oh, J. *et al.* Prenatal exposure to per- and polyfluoroalkyl substances in association with autism spectrum disorder in the MARBLES study. *Environment International* **147**, 106328 (2021).
- 12. Shin, H.-M., Bennett, D. H., Calafat, A. M., Tancredi, D. & Hertz-Picciotto, I. Modeled prenatal exposure to per- and polyfluoroalkyl substances in association with child autism spectrum disorder: a case-control study. *Environ Res* **186**, 109514 (2020).
- Shiue, I. Arsenic, heavy metals, phthalates, pesticides, hydrocarbons and polyfluorinated compounds but not parabens or phenols are associated with adult remembering condition: US NHANES, 2011–2012. *Environ Sci Pollut Res* 22, 6381–6386 (2015).
- Shiue, I. Urinary heavy metals, phthalates and polyaromatic hydrocarbons independent of health events are associated with adult depression: USA NHANES, 2011–2012. *Environ Sci Pollut Res* 22, 17095–17103 (2015).
- 15. Shiue, I. Urinary arsenic, pesticides, heavy metals, phthalates, polyaromatic hydrocarbons, and polyfluoroalkyl compounds are associated with sleep troubles in adults: USA NHANES, 2005–2006. *Environ Sci Pollut Res Int* **24**, 3108–3116 (2017).
- 16. Short-chain Polyfluoroalkyl Substances (PFAS).

- US EPA, O. KABAM Version 1.0 User's Guide and Technical Documentation Appendix F -Description of Equations Used to Calculate the BCF, BAF, BMF, and BSAF Values. <u>https://www.epa.gov/pesticide-science-and-assessing-pesticide-risks/kabam-version-10-users-guide-and-technical-3</u> (2015).
- Burkhard, L. P. Evaluation of Published Bioconcentration Factor (BCF) and Bioaccumulation Factor (BAF) Data for Per- and Polyfluoroalkyl Substances Across Aquatic Species. *Environmental Toxicology and Chemistry* 40, 1530–1543 (2021).
- O'Brien, T. M., Carlson, R. M., Oliveira, P. J. & Wallace, K. B. Esterification Prevents Induction of the Mitochondrial Permeability Transition by N-Acetyl Perfluorooctane Sulfonamides. *Chem. Res. Toxicol.* 19, 1305–1312 (2006).
- 20. PTX1737.pdf.
- 21. Glycine, N-ethyl-N-[(nonafluorobutyl)sulfonyl]-, potassium salt. Pharos https://pharosproject.net/chemicals/2032659#hazards-panel.

PFBS

- Lin, H.-W., Feng, H.-X., Chen, L., Yuan, X.-J. & Tan, Z. Maternal exposure to environmental endocrine disruptors during pregnancy is associated with pediatric germ cell tumors. *Nagoya J Med Sci* 82, 323–333 (2020).
- Eriksen, K. T. *et al.* Genotoxic potential of the perfluorinated chemicals PFOA, PFOS, PFBS, PFNA and PFHxA in human HepG2 cells. *Mutation Research/Genetic Toxicology and Environmental Mutagenesis* 700, 39–43 (2010).
- Brase, R. A., Mullin, E. J. & Spink, D. C. Legacy and Emerging Per- and Polyfluoroalkyl Substances: Analytical Techniques, Environmental Fate, and Health Effects. *Int J Mol Sci* 22, 995 (2021).
- 4. Toxicological Profile for Perfluoroalkyls, Agency for Toxic Substances and Disease Registry CDC
- 5. Gaballah, S. *et al.* Evaluation of Developmental Toxicity, Developmental Neurotoxicity, and Tissue Dose in Zebrafish Exposed to GenX and Other PFAS. *Environ Health Perspect* **128**, 047005 (2020).
- Ulhaq, M., Örn, S., Carlsson, G., Morrison, D. A. & Norrgren, L. Locomotor behavior in zebrafish (Danio rerio) larvae exposed to perfluoroalkyl acids. *Aquatic Toxicology* 144–145, 332–340 (2013).
- Liu, M. *et al.* Dietary administration of probiotic Lactobacillus rhamnosus modulates the neurological toxicities of perfluorobutanesulfonate in zebrafish. *Environ Pollut* 265, 114832 (2020).
- Slotkin, T. A., MacKillop, E. A., Melnick, R. L., Thayer, K. A. & Seidler, F. J. Developmental Neurotoxicity of Perfluorinated Chemicals Modeled in Vitro. *Environ Health Perspect* 116, 716–722 (2008).
- 9. Zhang, Q. *et al.* Effects of perfluorooctane sulfonate and its alternatives on long-term potentiation in the hippocampus CA1 region of adult rats in vivo. *Toxicol Res (Camb)* **5**, 539–546 (2016).
- Lieder, P. H., Chang, S.-C., York, R. G. & Butenhoff, J. L. Toxicological evaluation of potassium perfluorobutanesulfonate in a 90-day oral gavage study with Sprague–Dawley rats. *Toxicology* 255, 45–52 (2009).

- Newsted, J. L., Beach, S. A., Gallagher, S. P. & Giesy, J. P. Acute and Chronic Effects of Perfluorobutane Sulfonate (PFBS) on the Mallard and Northern Bobwhite Quail. *Arch Environ Contam Toxicol* 54, 535–545 (2008).
- 12. Qin, X.-D. *et al.* Association of perfluoroalkyl substances exposure with impaired lung function in children. *Environmental Research* **155**, 15–21 (2017).
- Burkhard, L. P. Evaluation of Published Bioconcentration Factor (BCF) and Bioaccumulation Factor (BAF) Data for Per- and Polyfluoroalkyl Substances Across Aquatic Species. *Environmental Toxicology and Chemistry* 40, 1530–1543 (2021).
- 14. Perfluorobutanesulfonic acid. Pharos https://pharosproject.net/chemicals/2012913#hazards-panel.

PFOS

- 1. Product-Chemical Profile for Food Packaging with PFASs: Discussion Draft. 184.
- Registration Dossier ECHA. https://echa.europa.eu/sl/registration-dossier/-/registered-dossier/11409/7/5/2.
- 3. Perfluoroalkylated substances: PFOA, PFOS and PFOSA.
- 4. Harris, M. H. et al. Prenatal and childhood exposure to per- and polyfluoroalkyl substances (PFASs) and child cognition. Environ Int 115, 358–369 (2018).
- 5. Zhang, Q. et al. Effects of perfluorooctane sulfonate and its alternatives on long-term potentiation in the hippocampus CA1 region of adult rats in vivo. Toxicol Res (Camb) 5, 539–546 (2016).
- 6. Fuentes, S., Vicens, P., Colomina, M. T. & Domingo, J. L. Behavioral effects in adult mice exposed to perfluorooctane sulfonate (PFOS). Toxicology 242, 123–129 (2007).
- Johansson, N., Fredriksson, A. & Eriksson, P. Neonatal exposure to perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) causes neurobehavioural defects in adult mice. NeuroToxicology 29, 160–169 (2008).
- Sørli, J. B. et al. Per- and polyfluoroalkyl substances (PFASs) modify lung surfactant function and pro-inflammatory responses in human bronchial epithelial cells. Toxicol In Vitro 62, 104656 (2020).
- 9. Lau, C. et al. Perfluoroalkyl Acids: A Review of Monitoring and Toxicological Findings. Toxicological Sciences 99, 366–394 (2007).
- 10. PERFLUOROOCTANESULFONIC ACID (PFOS, C-8). Pharos https://pharosproject.net/chemicals/2011510#hazards-panel.

Rhamnolipids

- 1. Semkova, S. *et al.* Rhamnolipid Biosurfactants—Possible Natural Anticancer Agents and Autophagy Inhibitors. *Separations* **8**, 92 (2021).
- Patowary, K., Das, M., Patowary, R., Kalita, M. C. & Deka, S. Recycling of Bakery Waste as an Alternative Carbon Source for Rhamnolipid Biosurfactant Production. *Journal of Surfactants and Detergents* 22, 373–384 (2019).
- 3. Johann, S. *et al.* Mechanism-specific and whole-organism ecotoxicity of mono-rhamnolipids. *Science of The Total Environment* **548–549**, 155–163 (2016).
- Muthaliar, A., Sevakumaran, V. & Bhubalan, K. PRODUCTION AND TOXICITY EVALUATION OF RHAMNOLIPIDS PRODUCED BY Pseudomonas STRAINS ON L6 AND HepG2 CELLS. *Malaysian Applied Biology* 48, 149–156 (2019).

- 5. Stipcevic, T., Knight, C. P. & Kippin, T. E. Stimulation of adult neural stem cells with a novel glycolipid biosurfactant. *Acta Neurol Belg* **113**, 501–506 (2013).
- 6. US EPA Office of Pesticide Programs. BIOPESTICIDES REGISTRATION ACTION DOCUMENT: RHAMNOLIPID BIOSURFACTANT.
- 7. 2-O-Rhamnopyranosyl-rhamnopyranosyl-3-hydroxyldecanoyl-3-hydroxydecanoate. Pharos <u>https://pharosproject.net/chemicals/2194944#hazards-panel</u>.

Sodium Lauroyl Glutamate (SLG)

- 1. Burnett, C. L. *et al.* Safety Assessment of Amino Acid Alkyl Amides as Used in Cosmetics. *Int J Toxicol* **36**, 17S-56S (2017).
- Kyadarkunte, A., Patole, M. & Pokharkar, V. In Vitro Cytotoxicity and Phototoxicity Assessment of Acylglutamate Surfactants Using a Human Keratinocyte Cell Line. *Cosmetics* 1, 159–170 (2014).
- 3. Burnett, C. L. *et al.* Safety Assessment of Amino Acid Alkyl Amides as Used in Cosmetics. *Int J Toxicol* **36**, 17S-56S (2017).
- 4. Mansouri, K. *et al.* CERAPP: Collaborative Estrogen Receptor Activity Prediction Project. *Environmental Health Perspectives* **124**, 1023–1033 (2016).
- Registration Dossier ECHA. <u>https://echa.europa.eu/sl/registration-dossier/-/registered-dossier/11409/5/3/1</u>.
- 6. Sodium lauroyl glutamate. Pharos https://pharosproject.net/chemicals/2077300#hazards-panel.

References

- ATSDR. PFAS chemicals overview. https://www.atsdr.cdc.gov/pfas/health-effects/overview.html (2021).
- Kato, K., Wong, L.-Y., Jia, L. T., Kuklenyik, Z. & Calafat, A. M. Trends in exposure to polyfluoroalkyl chemicals in the U.S. Population: 1999-2008. *Environ Sci Technol* 45, 8037–8045 (2011).
- US EPA. Technical Fact Sheet Perfluorooctane Sulfonate (PFOS) and Perfluorooctanoic Acid (PFOA). (2017).
- US EPA, O. Reducing PFAS in Drinking Water with Treatment Technologies. https://www.epa.gov/sciencematters/reducing-pfas-drinking-water-treatment-technologies (2018).
- Venkatesan, A. K. & Halden, R. U. National inventory of perfluoroalkyl substances in archived U.S. biosolids from the 2001 EPA National Sewage Sludge Survey. *Journal of Hazardous Materials* 252–253, 413–418 (2013).
- Maine Discovers New Toxic PFAS Contamination in Drinking Water. *Defend Our Health* https://defendourhealth.org/news/maine-discovers-new-toxic-pfas-contamination-in-drinking-water/.
- LD 1503, HP 1113, Text and Status, 130th Legislature, First Special Session. https://legislature.maine.gov/legis/bills/display_ps.asp?LD=1503&snum=130.
- 8. Zhou, J. *et al.* Per- and polyfluoroalkyl substances (PFASs) in airborne particulate matter (PM2.0) emitted during floor waxing: A pilot study. *Atmospheric Environment* **268**, 118845 (2022).
- Glycine, N-ethyl-N-[(nonafluorobutyl)sulfonyl]-, potassium salt. *Pharos* https://pharosproject.net/chemicals/2032659#hazards-panel.
- 10. Zeng, Z. *et al.* Assessing the human health risks of perfluorooctane sulfonate by in vivo and in vitro studies. *Environment International* **126**, 598–610 (2019).
- 11.Perfluorobutanesulfonic acid. *Pharos* https://pharosproject.net/chemicals/2012913#hazards-panel.
- 12. Savaglio, C. Fundamentals of Floor Care with Carmine Savaglio. (2021).
- 13. Chemours Fluorochemicals. CapstoneTM Fluorosurfactants: For high value-in-use applications

that require maximum performance. (2017).

- 14. Savaglio, C. Conversation with Carmine. (2021).
- 15. Lauren, S. What is Wettability. *What is Wettability*

https://www.biolinscientific.com/blog/what-is-wettability (2019).

- 16. Jarray, A., Wijshoff, H., A. Luiken, J. & Otter, W. K. den. Systematic approach for wettability prediction using molecular dynamics simulations. *Soft Matter* **16**, 4299–4310 (2020).
- 17. Korpela, J. What is surface tension?

https://www.biolinscientific.com/blog/what-is-surface-tension.

- 18. Surface Tension. Chemistry LibreTexts
 - https://chem.libretexts.org/Bookshelves/Physical_and_Theoretical_Chemistry_Textbook_Maps/Supple mental_Modules_(Physical_and_Theoretical_Chemistry)/Physical_Properties_of_Matter/States_of_M atter/Properties_of_Liquids/Surface_Tension (2013).
- Laurén, S. Surface tension of water Why is it so high?
 https://www.biolinscientific.com/blog/surface-tension-of-water-why-is-it-so-high.
- 20. Wetting. Wikipedia (2021).
- 21. Critical micelle concentration (CMC) and surfactant concentration.

https://www.kruss-scientific.com/en-US/know-how/glossary/critical-micelle-concentration-cmc-and-su rfactant-concentration.

- 22. Critical micelle concentration. *Wikipedia* (2021).
- 23. Octanol-water partition coefficient. *Wikipedia* (2021).
- Cumming, H. & Rücker, C. Octanol–Water Partition Coefficient Measurement by a Simple 1H NMR Method. ACS Omega 2, 6244–6249 (2017).
- 25. Minnesota Pollution Control Agency. Protecting communities and families from PFAS contamination. *Minnesota Pollution Control Agency*

https://www.pca.state.mn.us/about-mpca/protecting-communities-and-families-pfas-contamination (2021).

26. US EPA, O. Research on Per- and Polyfluoroalkyl Substances (PFAS).

https://www.epa.gov/chemical-research/research-and-polyfluoroalkyl-substances-pfas (2014).

- 27. NIOSH. PFAS. US CDC https://www.cdc.gov/niosh/topics/pfas/default.html (2021).
- 28. Carignan, S. PFAS Found in Landfills; No Clear Path on What to Do About It. *Bloomberg Law News*

https://news.bloomberglaw.com/environment-and-energy/pfas-found-in-landfills-no-clear-path-on-wha t-to-do-about-it (2020).

- Ocampo, G. Role of biosurfactants in nature and biotechnological applications. *Journal of Bacteriology & Mycology: Open Access* Volume 2, (2016).
- 30. Biosurfactants. (Springer Science+Business Media ; Landes Bioscience, 2010).
- Sekhon Randhawa, K. K. & Rahman, P. K. S. M. Rhamnolipid biosurfactants—past, present, and future scenario of global market. *Frontiers in Microbiology* 5, 454 (2014).
- Rawat, G., Dhasmana, A. & Kumar, V. Biosurfactants: the next generation biomolecules for diverse applications. *Environmental Sustainability* 3, 353–369 (2020).
- Ruiz-Morales, Y. & Romero-Martínez, A. Coarse-Grain Molecular Dynamics Simulations To Investigate the Bulk Viscosity and Critical Micelle Concentration of the Ionic Surfactant Sodium Dodecyl Sulfate (SDS) in Aqueous Solution. *J. Phys. Chem. B* 122, 3931–3943 (2018).
- Müller, M. M. *et al.* Rhamnolipids—Next generation surfactants? *Journal of Biotechnology* 162, 366–380 (2012).
- 35. European Commission. Directorate General for Research and Innovation., University of Bologna,
 & Fraunhofer ISI,. *Top 20 innovative bio-based products: task 3 of "Study on Support to R&I Policy in the Area of Bio based Products and Services"*. (Publications Office, 2019).
- 36. Liepins, J. *et al.* Glycolipid Biosurfactant Production from Waste Cooking Oils by Yeast: Review of Substrates, Producers and Products. *Fermentation* **7**, 136 (2021).
- Chong, H. & Li, Q. Microbial production of rhamnolipids: opportunities, challenges and strategies. *Microb Cell Fact* 16, 137 (2017).

- Drakontis, C. E. & Amin, S. Biosurfactants: Formulations, properties, and applications. *Current Opinion in Colloid & Interface Science* 48, 77–90 (2020).
- Costa, S. G. V. A. O. *et al.* Wettability of Aqueous Rhamnolipids Solutions Produced by Pseudomonas aeruginosa LBI. *Journal of Surfactants and Detergents* 12, 125–130 (2009).
- Dissolution Behaviour of Sulphonamides into Sodium Dodecyl Sulfate Micelles: A Thermodynamic Approach | Elsevier Enhanced Reader.

https://reader.elsevier.com/reader/sd/pii/S002235491549968X?token=C9CD6AD57FD00E1685269E8 23FB55435E99375354285395E6BDE8FF0121C2791CD10680EB4C21B196C3EB436ABE1E080&o riginRegion=us-east-1&originCreation=20211117190001 doi:10.1021/js9502251.

- Hodges, G. *et al.* A comparison of log Kow (n-octanol–water partition coefficient) values for non-ionic, anionic, cationic and amphoteric surfactants determined using predictions and experimental methods. *Environmental Sciences Europe* **31**, 1 (2019).
- 42. Wu, L. *et al.* Comparative studies on the surface/interface properties and aggregation behavior of mono-rhamnolipid and di-rhamnolipid. *Colloids and Surfaces B: Biointerfaces* **181**, 593–601 (2019).
- Costanza, J., Arshadi, M., Abriola, L. M. & Pennell, K. D. Accumulation of PFOA and PFOS at the Air–Water Interface. *Environ. Sci. Technol. Lett.* 6, 487–491 (2019).
- Krustev, R., Müller, H. J. & Toca-Herrera, J. L. The thickness and contact angle of sodium dodecyl sulfate foam films depending on the concentration of LiCl. *Colloid & Polymer Science* 276, 518–523 (1998).
- Helvaci, S. S., Peker, S. & Ozdemir, G. Effect of electrolytes on the surface behavior of rhamnolipids R1 and R2. *Colloids Surf B Biointerfaces* 35, 225–233 (2004).
- Patowary, K., Das, M., Patowary, R., Kalita, M. C. & Deka, S. Recycling of Bakery Waste as an Alternative Carbon Source for Rhamnolipid Biosurfactant Production. *Journal of Surfactants and Detergents* 22, 373–384 (2019).
- Semkova, S. *et al.* Rhamnolipid Biosurfactants—Possible Natural Anticancer Agents and Autophagy Inhibitors. *Separations* 8, 92 (2021).

- Diggle, S. P. & Whiteley, M. Microbe Profile: Pseudomonas aeruginosa: opportunistic pathogen and lab rat. *Microbiology (Reading)* 166, 30–33 (2020).
- 49. Pseudomonas Infection | Michigan Medicine. https://www.uofmhealth.org/health-library/abj6977.
- 50. Dobler, L. *et al.* Environmentally friendly rhamnolipid production for petroleum remediation. *Chemosphere* **252**, 126349 (2020).
- 51. Eslami, P., Hajfarajollah, H. & Bazsefidpar, S. Recent advancements in the production of rhamnolipid biosurfactants by Pseudomonas aeruginosa. *RSC Advances* **10**, 34014–34032 (2020).
- Byrne, F. P. *et al.* Tools and techniques for solvent selection: green solvent selection guides.
 Sustain Chem Process 4, 7 (2016).
- Tiso, T. *et al.* Integration of Genetic and Process Engineering for Optimized Rhamnolipid
 Production Using Pseudomonas putida. *Frontiers in Bioengineering and Biotechnology* 8, 976 (2020).
- 54. Lombardo, T. Overview of a Maine School District's Experiences with Floor Wax. (2021).
- 55. Difference Between Aerobic and Anaerobic Wastewater Treatment. *Compare the Difference Between Similar Terms*

https://www.differencebetween.com/difference-between-aerobic-and-vs-anaerobic-wastewater-treatme nt/.

- Mohan, P. K., Nakhla, G. & Yanful, Ernest. K. Biokinetics of biodegradation of surfactants under aerobic, anoxic and anaerobic conditions. *Water Research* 40, 533–540 (2006).
- Kumarasamy, R. *et al.* Screening of Rhamnolipid Produced by Marine Bacterium for Heavy Metal Removal in Mangrove Soil. 1, 29–36 (2019).
- Chrzanowski, Ł. *et al.* Biodegradation of rhamnolipids in liquid cultures: Effect of biosurfactant dissipation on diesel fuel/B20 blend biodegradation efficiency and bacterial community composition. *Bioresource Technology* 111, 328–335 (2012).
- 59. Wei, Z. *et al.* Potential use of biochar and rhamnolipid biosurfactant for remediation of crude oil-contaminated coastal wetland soil: Ecotoxicity assessment. *Chemosphere* **253**, 126617 (2020).
- 60. n-Octanol/Water Partition Coefficient (Kow/logKow).

https://www.chemsafetypro.com/Topics/CRA/n_Octanol_Water_Partition_Coefficient_Kow.html.

- 61. 2-O-Rhamnopyranosyl-rhamnopyranosyl-3-hydroxyldecanoyl-3-hydroxydecanoate. *Pharos* https://pharosproject.net/chemicals/2194944#hazards-panel.
- 62. Johann, S. *et al.* Mechanism-specific and whole-organism ecotoxicity of mono-rhamnolipids. *Science of The Total Environment* **548–549**, 155–163 (2016).
- 63. Morán, M. C. et al. "Green" amino acid-based surfactants. Green Chem. 6, 233–240 (2004).
- 64. Amino Acids | BioNinja.

https://ib.bioninja.com.au/standard-level/topic-2-molecular-biology/24-proteins/amino-acids.html.

- 65. Understanding Surfactants: A Guide. *International Products Corporation* https://www.ipcol.com/blog/an-easy-guide-to-understanding-surfactants/ (2018).
- Burnett, C. L. *et al.* Safety Assessment of Amino Acid Alkyl Amides as Used in Cosmetics. *Int J Toxicol* 36, 178-568 (2017).
- 67. Pinheiro, L. & Faustino, C. *Amino Acid-Based Surfactants for Biomedical Applications*. *Application and Characterization of Surfactants* (IntechOpen, 2017). doi:10.5772/67977.
- 68. Scott, E., Peter, F. & Sanders, J. Biomass in the manufacture of industrial products—the use of proteins and amino acids. *Appl Microbiol Biotechnol* **75**, 751–762 (2007).
- 69. EastHill Corporation (www.ieasthill.com). http://www.ieasthill.com/.
- 70. SODIUM LAUROYL GLUTAMATE Personal & Hair Care AAKO. *AAKO. Chemical Excellence*. https://www.aako.nl/products/personal-and-hair-care/sodium-lauroyl-glutamate/.
- 71. Registration Dossier ECHA.

https://echa.europa.eu/de/registration-dossier/-/registered-dossier/11409/4/1.

- Pérez, L., Pinazo, A., Pons, R. & Infante, Mr. Gemini surfactants from natural amino acids.
 Advances in Colloid and Interface Science 205, 134–155 (2014).
- Pinazo, A., Pons, R., Pérez, L. & Infante, M. R. Amino Acids as Raw Material for Biocompatible Surfactants. *Ind. Eng. Chem. Res.* 50, 4805–4817 (2011).
- 74. Pal, N., Hoteit, H. & Mandal, A. Structural aspects, mechanisms and emerging prospects of

Gemini surfactant-based alternative Enhanced Oil Recovery technology: A review. *Journal of Molecular Liquids* **339**, 116811 (2021).

- 75. Yoshimura, T. & Esumi, K. Synthesis and surface properties of anionic gemini surfactants with amide groups. *Journal of Colloid and Interface Science* **276**, 231–238 (2004).
- 76. Sodium Salts of Bis(1-dodecenylsuccinamic Acids): A Simple Route to Anionic Gemini Surfactants | Elsevier Enhanced Reader. https://reader.elsevier.com/reader/sd/pii/S0021979701975496?token=84DE9494FD3B223FE068CBA

D1B996580204DF70AF365A546D5FE8628D14548619AEDF6B9A1E733E6823AD63D9E3C1CAC & originRegion=us-east-1&originCreation=20211214003357 doi:10.1006/jcis.2001.7549.

- 77. Akram, M., Ansari, F., Bhat, I. A., & Kabir-ud-Din. Probing interaction of bovine serum albumin (BSA) with the biodegradable version of cationic gemini surfactants. *Journal of Molecular Liquids* 276, 519–528 (2019).
- 78. System of Registries | US EPA.

https://sor.epa.gov/sor_internet/registry/substreg/searchandretrieve/substancesearch/search.do?synId=1 241605&displaySynonym=.

- 79. Parsi, K. Interaction of detergent sclerosants with cell membranes. *Phlebology* 30, 306–315 (2015).
- Burnett, C. L. *et al.* Safety Assessment of Amino Acid Alkyl Amides as Used in Cosmetics. *Int J Toxicol* 36, 178-568 (2017).
- Pureact GLT by Innospec Performance Chemicals Personal Care & Cosmetics. https://www.ulprospector.com/en/na/PersonalCare/Detail/2626/1003656/Pureact-GLT.
- Sodium Lauroyl Glutamate (Huzhou Ouli Biotechnology) Huzhou Ouli Biotechnology datasheet.

https://cosmetics.specialchem.com/product/i-huzhou-ouli-biotechnology-sodium-lauroyl-glutamate-hu zhou-ouli-biotechnology.

83. Sodium lauroyl glutamate. *Pharos* https://pharosproject.net/chemicals/2077300#hazards-panel.

- ECHA. Registration Dossier L-Glutamic acid, N-coco acyl derivs., monosodium salts. https://echa.europa.eu/sl/registration-dossier/-/registered-dossier/11409/5/3/1.
- Li, G. *et al.* Alkyl polyglycoside: a green and efficient surfactant for enhancing heavy oil recovery at high-temperature and high-salinity condition. *J Petrol Explor Prod Technol* 9, 2671–2680 (2019).
- Yan, L. Alkyl polyglucoside applications in industry. *Coating Additives Manufacturer* | *IRO Coating Additive Ltd* https://www.irocoatingadditive.com/alkyl-polyglucoside-applications-industry/ (2017).
- 87. EWG. Alkyl Polyglucoside (C10-16).

http://www.ewg.org/guides/substances/151986-AlkylPolyglucosideC1016.

- 88. Dow Consumer & Industrial Solutions. TRITONTM CG-425 Alkyl Polyglucoside Surfactant.
- Yarmey, S., Paiva, A., Liu, A. & Baran, J. Use of a Silicone Surfactant in Polishing Compositions. 21.
- 90. Wong, I. Y. & Wong, D. Chapter 104 Special Adjuncts to Treatment. in *Retina (Fifth Edition)*(eds. Ryan, S. J. et al.) 1735–1783 (W.B. Saunders, 2013). doi:10.1016/B978-1-4557-0737-9.00104-1.
- Lovell, T. Downstream users raise concerns over siloxanes SVHC decision. *Chemical Watch* https://chemicalwatch.com/68292/downstream-users-raise-concerns-over-siloxanes-svhc-decision (2018).
- 92. Mojsiewicz-Pieńkowska, K., Jamrógiewicz, M., Szymkowska, K. & Krenczkowska, D. Direct Human Contact with Siloxanes (Silicones) – Safety or Risk Part 1. Characteristics of Siloxanes (Silicones). *Front Pharmacol* 7, 132 (2016).
- 93. Lassen, C., Hansen, C. L., Mikkelsen, S. H. & Maag, J. Siloxanes Consumption, Toxicity and Alternatives. 111

https://www2.mst.dk/udgiv/publications/2005/87-7614-756-8/pdf/87-7614-757-6.pdf.

94. Ph. D., B. S., B. A., P. and M., Facebook, F. & Twitter, T. What's Carnauba Wax? *ThoughtCo* https://www.thoughtco.com/what-is-carnauba-wax-607371.

- 95. The queen of waxes P&B Products. https://pbproducts.com.br/the-queen-of-the-waxes/.
- 96. Carnauba Wax an overview | ScienceDirect Topics.

https://www.sciencedirect.com/topics/biochemistry-genetics-and-molecular-biology/carnauba-wax.

97. The Evolution Of Floor Finishes. *CleanLink*

http://www.cleanlink.com/cleanlinkminute/details/The-Evolution-Of-Floor-Finishes--36566.

- 98. Charumanee, S., Yotsawimonwat, S., Sirisa-ard, P. & Pholsongkram, K. Characterization and chemical composition of epicuticular wax from banana leaves grown in Northern Thailand. 8 (2017).
- Shin, S. *et al.* Bio-Inspired Extreme Wetting Surfaces for Biomedical Applications. *Materials* (*Basel*) 9, 116 (2016).
- Winter's Worst No Match for Premium Rubber Flooring at Syracuse University -.
 Spaces4Learning https://spaces4learning.com/articles/2017/07/28/rubber-flooring.aspx (2017).
- Nguyen, T. T., Youssef, N. H., McInerney, M. J. & Sabatini, D. A. Rhamnolipid biosurfactant mixtures for environmental remediation. *Water Research* 42, 1735–1743 (2008).
- Chen, M. *et al.* Influence of Calcium Ions on Rhamnolipid and Rhamnolipid/Anionic Surfactant Adsorption and Self-Assembly. *Langmuir* 29, 3912–3923 (2013).
- 103. Zhang, W., Gao, Z., Zhu, H. & Zhang, Q. Mixed micellization of cationic/anionic amino acid surfactants: Synergistic effect of sodium lauroyl glutamate and alkyl tri-methyl ammonium chloride. *Journal of Dispersion Science and Technology* **0**, 1–13 (2021).
- 104. Ji, X., Tian, M. & Wang, Y. Temperature-Induced Aggregate Transitions in Mixtures of Cationic Ammonium Gemini Surfactant with Anionic Glutamic Acid Surfactant in Aqueous Solution. *Langmuir* **32**, 972–981 (2016).
- 105. Sharma, K. S., Rodgers, C., Palepu, R. M. & Rakshit, A. K. Studies of mixed surfactant solutions of cationic dimeric (gemini) surfactant with nonionic surfactant C12E6 in aqueous medium. *Journal of Colloid and Interface Science* 268, 482–488 (2003).