

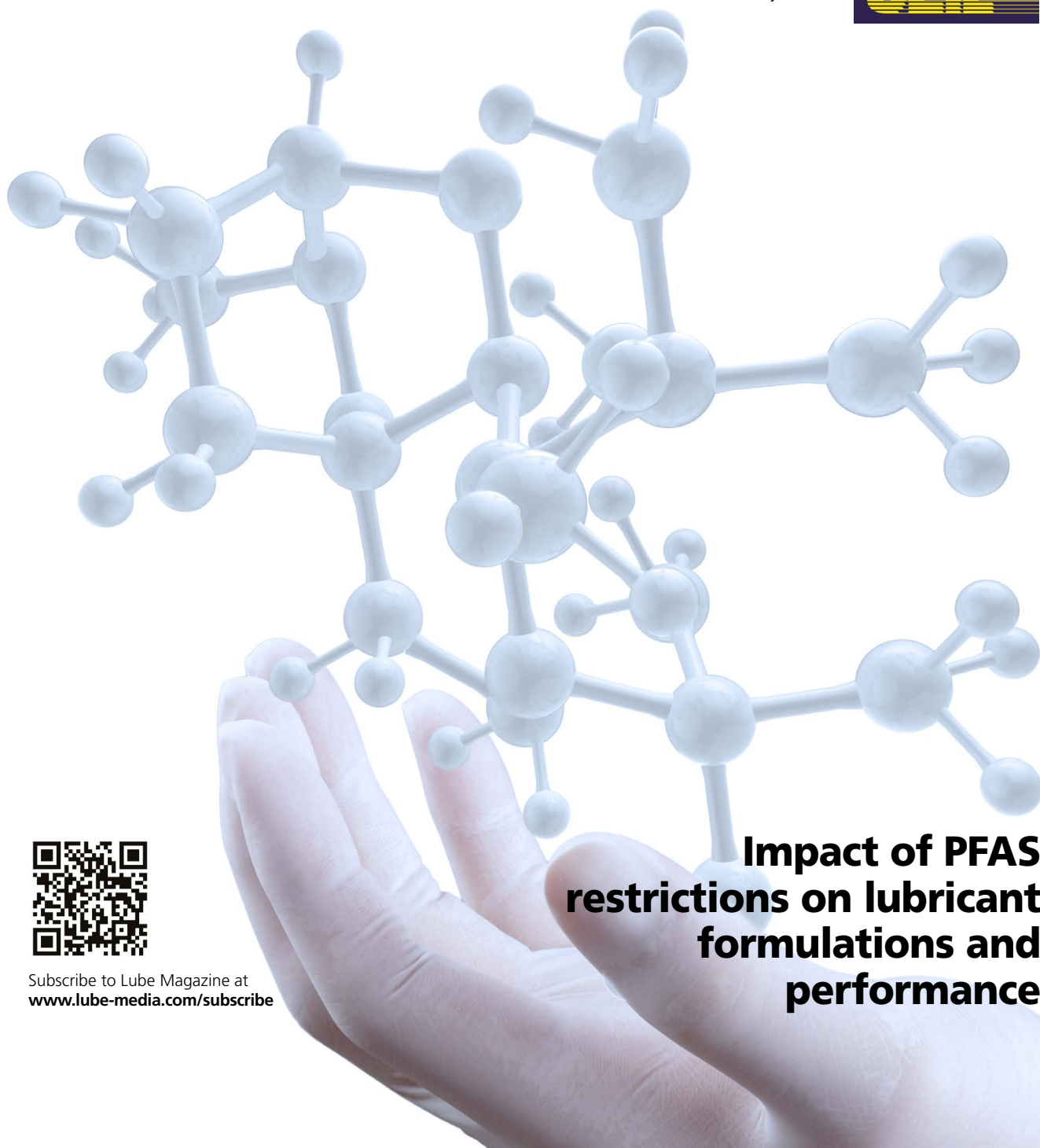
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**Impact of PFAS
restrictions on lubricant
formulations and
performance**

Impact of PFAS restrictions on lubricant formulations and performance

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As tightening global regulations drive the phase-out of many PFAS chemistries, lubricant formulators face a major challenge: maintaining high performance without relying on these long-standing additives. Once valued for their exceptional durability, low friction and chemical

stability, PFAS are now being replaced by a new generation of fluorine-free alternatives designed to balance tribological performance with growing environmental and sustainability demands.



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Introduction

Per- and polyfluoroalkyl substances (PFAS) are often referred to as “forever chemicals” due to their persistence in environments [1]. These substances consist of more than 10,000 synthetic chemicals and are used to provide heat, water, oil, and stain resistance in various applications such as non-stick coating, food packaging, firefighting foam, and aerospace technologies. PFAS have extremely strong and stable C-F bonds, allowing them to be bio-accumulative and resistant to environmental degradation. Their widespread use and propensity for bioaccumulation resulted in extensive environmental contamination and significant concern for human and ecosystem health. This led to accelerated regulatory actions and search for safe and suitable alternatives, as well as advancements in analytical and sampling methods. Certain PFAS such as polytetrafluoroethylene (PTFE), are fluoropolymers with superior lubricity and mechanical properties and

are classified as regulated substances due to their stability and long-term accumulation potential.

The formulation of high-performance lubricants is transitioning from PTFE to complex packages that utilise liquid base oils that are encapsulated within protective shells or adsorbed onto porous carriers. Although PTFE is defined as a low-friction material due to its relatively low friction coefficient, more focus has been targeted towards liquid lubricants because their friction coefficient can be two orders of magnitude lower than solid lubricants. Some liquid base oils that can be used in lubricant formulations are synthetic oils, silicone oils, bio-based and natural oils. Synthetic oils, such as polyalphaolefin (PAO), are high-performance synthetic base oils that are used in microencapsulated systems.

Additive packages are a structural delivery system designed for lubrication on demand. The packages

consist of carriers that can protect the base oil and release it only when the frictional wear occurs. Polymer shell additives are chemicals mixed into plastics or ceramics to further boost performance. Base oils are often packaged in shells that are made of melamine-formaldehyde resins, polysulphone, polyurethane, or polyamides [7]. These shells all ensure that the oil can be released as a solid filler within a coating matrix without leaking. In addition, to improve thermal stability, oils can be encapsulated in hybrid shells using silica particles or adsorbed onto porous materials. Many additive packages are embedded into a polymer matrix that will provide structural integrity for the lubricant coating. Some common examples are epoxy resin, polyimide, and polyamide 6.

Regulatory landscape and phase-out policies

Regulatory efforts have been implemented globally to restrict the production and use of PFAS since the 2000s. The regulatory landscape concerning PFAS is rapidly evolving internationally and nationally. International agreements and regional bodies have established key restrictions on PFAS. The Stockholm Convention on Persistent Organic Pollutants became international law in 2004 and in 2009, the conference decided to list perfluorooctane sulfonic acid (PFOS) and perfluorooctane sulphonyl fluoride in Annex B, requiring parties to ban or restrict their use. In 2020, firefighting foams containing perfluorooctanoic acid (PFOA) were also banned. Perfluorohexanesulfonic acid (PFHxS) at times was used as a replacement for PFOS and was also listed in Annex A, obligating the parties of the treaty to ban the production and use along with PFOA [4]. In 2006, the EU Parliament and Council acknowledged that PFOS is a very persistent, bio-accumulative and toxic chemical.

Figure 1 illustrates the structure of the database through using specific examples of PFAS when used in fluorinated gases and in food contact materials [4]. Alternative assessment frameworks have been developed to avoid substitutions where the alternative can result in unforeseen consequences. The frameworks can be defined as processes for “identifying, comparing, and selecting alternatives to chemicals of concern” based on their hazardous characteristics, performance and economic viability. To efficiently screen and compare a broader range of alternatives, it is encouraged to define the chemical function, end-use functions, and functions as a service of a chemical in a specific use.

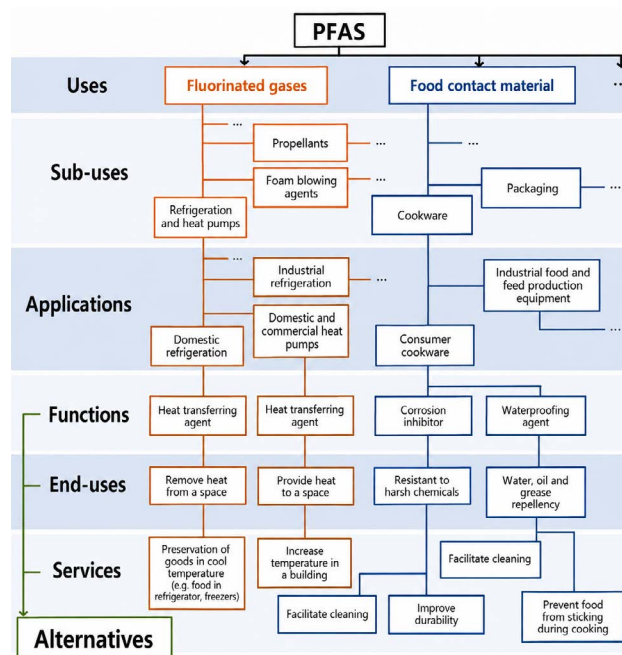


Figure 1: General structure of the database for PFAS [4].

In the European Union (EU), the European Chemicals Agency (ECHA) controls and regulates the various restrictions on chemicals [3]. The European Commission launched the Green Deal, which pushes broader environmental policies and legal alignment with initiatives to regulate PFAS in water. There was also a push to classify PFAS under “essential use” categories to regulate PFAS usage and have better management of the transition to alternatives. Four member states of the European Union (Denmark, Germany, The Netherlands, and Sweden), in addition to Norway submitted a restriction proposal under the Registration, Evaluation, Authorisation, and Restriction of Chemicals (REACH) Regulation to restrict the manufacturing of PFAS. The restriction proposal aligns with the objective of the European Commission with phasing out the use of PFAS in the EU.

In the lubrication section, PFAS are pervasive in high-performance formulations like in PFPE oils. PTFE powders have been widely utilised in self-lubricating polymer composite coatings and greases that are designed for critical mechanical components like shafts, gears, and bushings [7]. PTFE is also increasingly favoured for its use in hostile environments due to its chemical inertness. Fluorinated surfactants act as vital wetting agents, emulsifiers, and stabilisers in manufacturing processes such as metal plating and electronics due to its exceptional stability and surface-active properties [5].

Federal regulatory action in the U.S. has relied on advisories and voluntary agreements but are gradually accelerating toward enforceable standards such as EPA Authority, Significant New Use Rules, and PFOA Stewardship Program [2]. The United States Environmental Protection Agency (EPA) has the authority to regulate PFAS through key statutes such as Toxic Substance Control Act (TSCA), Safe Drinking Water Act (SDWA), and Comprehensive Environmental Response, Compensation and Liability Act (CERCLA). The U.S. EPA released their Lifetime Health Advisory Limits for PFOS and PFOA in 2016. The U.S. saw a rapid increase in the number of states that implemented policies to prevent PFAS pollution after the release. The Significant New Use Rules (SNURs) were published in 2002 and required notification to the EPA prior to manufacturing and importation of 75 PFAS compounds. In 2006, EPA established the PFOA Stewardship Program, allowing for major producers to voluntarily reduce their PFOA emissions by 95% by 2010.

Major manufacturers in clothing, furniture, cosmetics, and other sectors have gradually started phasing out PFAS, indicating that using PFAS is not essential to their business models. In the production and uses of fluoropolymers and in the semiconductor industry, it is more difficult to phase out due to demanding conditions and diverse applications of PFAS. PFAS can be phased out in coatings for strings of musical instruments, in certain food packaging where uncoated paper and paper plates are suitable alternatives, and in lubricants for certain consumer products such as bike chain lubricants. Unfortunately, no alternative has been identified for the chemical industry, such as when used as solvent or for polymer curing, or to produce plastic and rubber to be used as processing aid or mold release agent.

Role of PFAS in lubricant formulations

The role of PFAS is driven by their superior chemical and physical properties which are essential for reducing friction and wear. PFAS and PTFE are widely incorporated into lubricant and low-friction composite formulations due to their unique characteristics. Among various organic polymers, PTFE has one of the lowest surface energy and friction coefficients (COF) of any bulk polymer, with a value of 0.1 [7]. PFAS also have strong thermal and chemical stability, allowing for PFAS to be resistant

to heat, water, oil, chemicals, and stains. Pure PTFE is soft and has a high wear rate, however, it is commonly used as a filler or matrix in composite materials to enhance its performance which would lead to improved lubricity, load transmission, and overall wear resistance.

PFAS are typically used in lubricants, which are applied as self-lubricating polymer composite coatings on mechanical components such as shafts, gears, and bushings. Polymer coatings allow for performance improvement through water/oil release, self-cleaning and healing, and surface corrosion protection. Self-lubricating polymer composite coatings are commonly used in aerospace applications to minimise friction, wear, and conserve energy.

PFAS have been widely utilised due to the diversity of their potential uses, and their ability to deliver multiple functions while using one application. Finding alternatives with these same traits has proved difficult. A friction modifier, also known as lubricity, is a chemical additive that creates a barrier between metal surfaces, reducing friction and wear. PTFE is an example of friction modifier and acts as functional fillers in matrices to enhance energy efficiency. Anti-wear agents can improve the mechanical durability of coatings and prevent debris during frictional wear [7].

Government regulations regarding PFAS are expected to hinder the growth of the low friction coating market, hoping for alternatives to be developed. In general, liquid lubricants such as oil, have a better lubricating effect than solid lubricants. However, liquid lubricants have inherent limitations in their application as coating due to restrictions on replenishment, storage, and maintenance during the wear process of the coating surface [7].

To combat this, a proposed liquid lubricant-filled microcapsule or nanocomposites is brought up. The liquid lubricants are encapsulated or made into a composite adsorbed on a solid material and dispersed as a solid lubricant within the coating layer. When the frictional wear occurs, the capsule or composite would then break, releasing the liquid lubricant, also shown in the SEM image in Figure 2b. The lubricant that flows out of the capsule or composite would form a thin film, providing a lubricating effect and reducing friction wear.

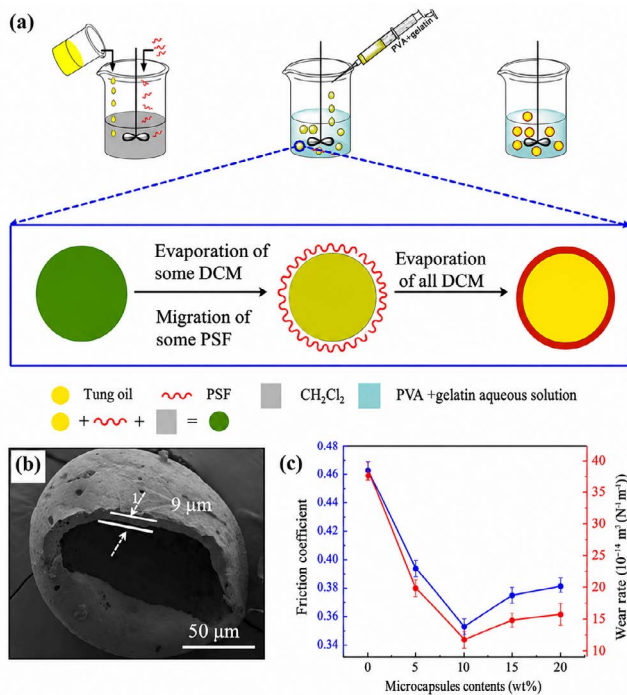


Figure 2: (a) Schematic of microencapsulation method. (b) SEM micrographs of ruptured microcapsule. (c) Frictional coefficients and specific wear rates of self-lubricant coating vs. content of tung oil-loaded microcapsule. [7]

Emerging alternatives and green substitutes

Some alternatives to PTFE for in situ release upon wear include microcapsules containing single-layer polymer shells, pickering emulsion microcapsules that contain inorganic solid shells, microcapsules

containing multilayer polymer shells, and gelation of liquid lubricants. In the study above, the polysulphone shells were filled with tung oil, and another example of single-layer polymer shell would be melamine-formaldehyde resins filled with silicone oil. PSF/SiO₂ hybrid shells filled with lubricant oil can be combined with solvent evaporation techniques that use lubricants as the core material and silica particles as stabilisers for the colloidal system. The silica particles can also regulate the size of the microcapsules. PSF has excellent mechanical qualities SiO₂ has high thermal stability.

Figure 3 (a) Manufacturing process of gelation of liquid lubricants. (b) CLSM images of oil gel capsule in coating layer. (c) Illustration of the mechanism of oil gel microcapsules within coating layer. (d) Linear reciprocating friction test of steel disk samples. [7]

A study conducted by Hyundai Motors and Sahmyook University developed a novel method to encapsulate oils. A net-like fibre structure with a gelator is created, which would effectively trap the oil within cavities. This method uses organogels without plastic shells to prevent the formation of defects by aggregation after the release of internal oils, mitigating against scratches and collisions.

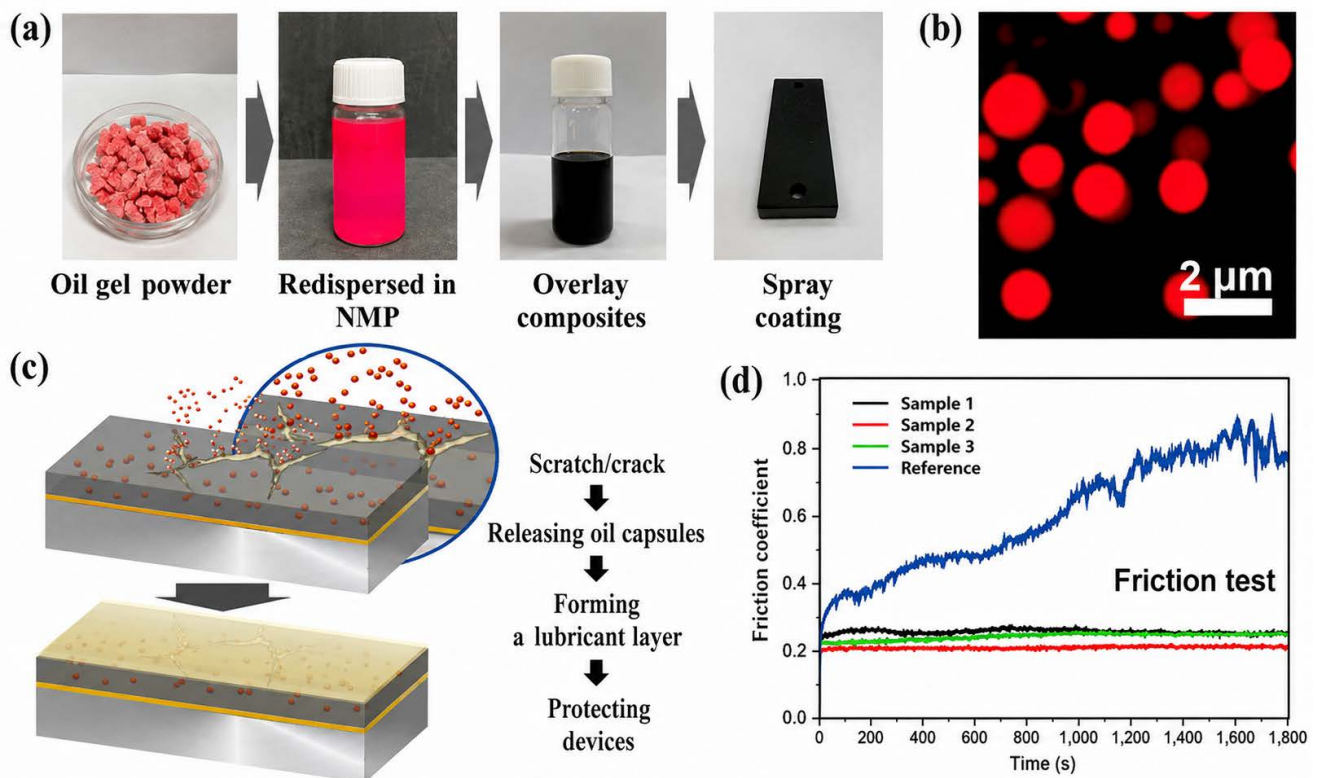


Figure 3: (a) Manufacturing process of gelation of liquid lubricants. (b) CLSM images of oil gel capsule in coating layer. (c) Illustration of the mechanism of oil gel microcapsules within coating layer. (d) Linear reciprocating friction test of steel disk samples. [7]

A temperature-sensitive gel system was then developed to release oil in response to an increase in temperature by scratching and pushing. Figure 3a shows the process of spray coating oil gel capsule powder on a bearing sample and Figure 3b presents the state of the oil-gel capsule within the coating layer using confocal laser scanning microscopy (CLSM).

Technical and industrial challenges

One of the main causes of technical and industrial challenges is developing analytical measurement and sampling methods. PFAS contain numerous substances (around 5,000-10,000) and many of these compounds have unknown structures. This is because PFAS precursors can biotransform in the environment into a mixture of intermediate fluorinated product [8]. In addition, current standardised analytical methods generally focus on targeted analysis of known PFAS. This approach significantly underestimates the total amount of PFAS contamination present in a region.

There is also a lack of consensus or standardisation across federal and state agencies with international organisations regarding PFAS sampling procedures. The reliability and comparability of data is then decreased. Another main challenge is the extraction of PFAS due to the complexity of environmental matrices. Soil, sediment, sludge, and biological tissues can hinder repeatable conversion and complicate the analysis. The manufacturing method of PFAS can yield either branched or linear isomers [6]. Branched isomers may produce lower detection responses in mass spectrometry, leading to inaccurate or low biased measurements, especially if quantification relies only on linear standards (LC/MS/MS).

As mentioned previously, PFAS have strong C-F bonds, causing chemical breakdown of the bond to require extremely high temperatures (often over 1000 °C). Incineration is a common industrial method for degrading PFAS on solids and can result in the formation of hazardous byproducts such as HF gas.

While numerous destruction methods such as reductive processes and low-temperature decarboxylation are promising approaches, they also face practical hurdles for large-scale implementation. Low-temperature decarboxylation requires a large amount of DMSO solvent [5] and advanced reductive processes need to control dissolved oxygen. The risk

of increasing the total dissolved solid (TDS) levels cause it to be impractical to conduct drinking water treatment.

Future research directions

Future research should focus on developing safer and more sustainable alternatives for PFAS without using incompatible substitutions. Researchers should focus on properly characterising the hazard profiles of identified alternatives to ensure that the correct substitution is used. Other areas that researchers should focus on is performance optimisation and identifying missing alternatives. There are 83 applications that no alternatives could be identified. As alternatives are identified, there would be development in the chemical industry and in plastic and rubber production.

The technical limitations in analytical methods and advanced detection should also be addressed. Currently, researchers use targeted methods and there is a lack in non-targeted methods. Future research should focus on developing methods that can overcome the limitations of conventional HPLC-MS/MS methods used to analyze PFAS precursors or isomers.

Conclusions

In conclusion, increasing regulatory restrictions on specific PFAS and PFAS-containing applications are accelerating the search for safer and more sustainable alternatives. This is due to PFAS having extraordinary persistence in the environment and their impact on human and environmental health. One of the most critical challenge in finding alternatives is the compatibility and safeness of the replacement substitutes. PFAS can deliver eight chemical functions in a single coating application, and it is difficult to find an alternative to do the same. Overall, developing advanced analytical methods and collecting more data on the use of PFAS, emission, and its alternatives would allow research on PFAS alternatives to be more efficient.

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