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► **To cite this version:**

Bruno Ameduri. Fluoropolymers: A special class of per- and polyfluoroalkyl substances (PFASs) essential for our daily life. *Journal of Fluorine Chemistry*, 2023, 267, pp.110117. <10.1016/j.jfluchem.2023.110117>. <hal-04044498>

**HAL Id: hal-04044498**

**<https://hal.science/hal-04044498v1>**

Submitted on 24 Mar 2023

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# **Fluoropolymers: a Special Class of Per- and polyfluoroalkyl Substances (PFASs) Essential for our Daily Life**

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**Abstract:** Per- or polyfluoroalkyl substances (PFASs) are valuable compounds which fall into two main families: i) the low molar-mass PFASs (including fluorinated telomers, perfluorocarboxylic acids, oligomers and surfactants) which are toxic, persistent, bioaccumulable, mobile and are under severe restricted (several products are already banned) and drastic regulations and ii) polymeric ones. Among the latter, polymers bearing fluorinated side moieties have also undergone such above issues. But, fluoropolymers (FPs) are niche specialty bio-inert and safe polymers, stable thermally, chemically and against UV and aging. They display outstanding properties, their molar masses being able to reach several million  $\text{g}\cdot\text{mol}^{-1}$ . They fulfil the Polymer of Low Concern criteria and are not prone to face such above hazardous issues. These (co)polymers can be used in many High Tech applications (e.g., Energy, electronics, aerospace, automotive and biomedical industries). All these PFASs have recently been under heavy pressure from regulators, press and media. However, the main concerns linked to the industrial productions of FPs have been addressed showing that these high value added polymers must be separated from conventional PFASs and are irreplaceable materials.

Keywords: fluoropolymers, per- and polyfluoroalkyl substances, polymerization aids, regulators, surfactants

## **Introduction**

Fluorine chemistry has been undergoing another severe attack, launched more than one decade ago. Media have been quite active to denounce the bad effects of fluorinated products while many plaintiffs have sued fluorochemical manufacturers. Formerly, the context of chlorofluorocarbons (CFCs) was severe and suitable alternative solutions were found with

hydrogenofluorocarbons (HFCs) [1-2]. Then, another major concern dealt with the fluorosis, a common disorder, characterised by hypomineralisation of tooth enamel caused by ingestion of excessive fluoride during enamel formation [3-4]. The recent years have introduced a new battle against per- or polyfluoroalkyl substances (PFASs) for which the regulations will be stringent in these coming years [5-6]. Various fluorochemicals are already banned and continuous procedure from competent authorities are pursuing their pressure to impose drastic legislations [6-7].

Containing multiple C–F bonds, per- and poly-fluoroalkyl substances (PFAS) represent a group of artificial/ anthropogenic chemicals with unique hydrophobic/lipophobic physicochemical properties and quite different physical, chemical, and biological properties [8-10]. These are valuable compounds, having found many applications in different areas [6,9-12].

From the Organization for Economic Co-operation and Development (OECD) [5], the number of PFAS molecules are >4700 PFASs while it reaches more than 9000 compounds according to the US Environment Protection Agency [13].

### **Applications of PFASs but....**

A peculiar variety of PFASs deals with fluorinated surfactants. These surface active products [14] have been the best ones since their surface tension and surface properties are much better than those of HC surfactants [14-17]. They have been involved in more than 200 applications (e.g., food packaging, paints, firefighting foams, cosmetics, electroplatings, photoacid generators, semiconductors and as polymerization aids in the emulsion radical polymerization of fluorinated alkenes [6, 12, 18]). However, the subject of issues deals with their toxicity, persistence [19-20], bioaccumulation, water solubility and mobility [6, 8, 10-11, 21-22]. Indeed, the modern means of detection have been so powerful that several tenth of ppb [23] (or even hundreds of ppt) can be detected (just to recall that one ppb is a drop of water in an Olympic swimming pool!). As a matter of fact, to the best available Science, the Food and Drug Administration (FDA) has no evidence that PFASs, at levels found in the samplings, induce some human health issues [24].

Actually, persistent organic pollutant (POP) regulation arising from the Stockholm Convention has forbidden perfluorooctane sulfonic acid (PFOS) since 2009, perfluorooctanoic acid (PFOA) since July 2020 and perfluorohexanesulfonic acid (PFHxS) since June 2022 while restrictions on perfluoroalkane carboxylic acids (PFCAs) are coming too [25] while the dossier is still under evaluation [26]. Furthermore, the restrictions imposed

by the POP regulations may also consider substances as themselves or when they act as components of items, or incorporated in formulations above certain thresholds.

Other PFASs are also on the way to be restricted and environmental officials from a number of European countries have already announced a plan to restrict all PFASs by 2030 [26] while more than a dozen of American states have unveiled legislation which includes a ban on PFASs in the coming years. Thus, many efforts have been developed to synthesize alternatives to escape from PFOA and FPOS substituted by other PFAS (except in China where PFOA is still in use as well as others bearing shorter chain lengths as polymerization aids). Industrially produced alternatives are ammonium salt of hexafluoropropylene oxide dimer acid (Gen®X) and 4,8-dioxa-3H-perfluorononanoate (Adona®), developed by Chemours and Dyneon, respectively, but these products also display bioaccumulable, persistent, toxic and mobile (BPTM) issues [10,27].

### How to categorize PFASs?

Other PFASs, which are not necessary surfactants, fall into two main families [28]: i) the low molar-mass PFASs (including fluorinated telomers, perfluorocarboxylic acids and surfactants) which are BPTM.

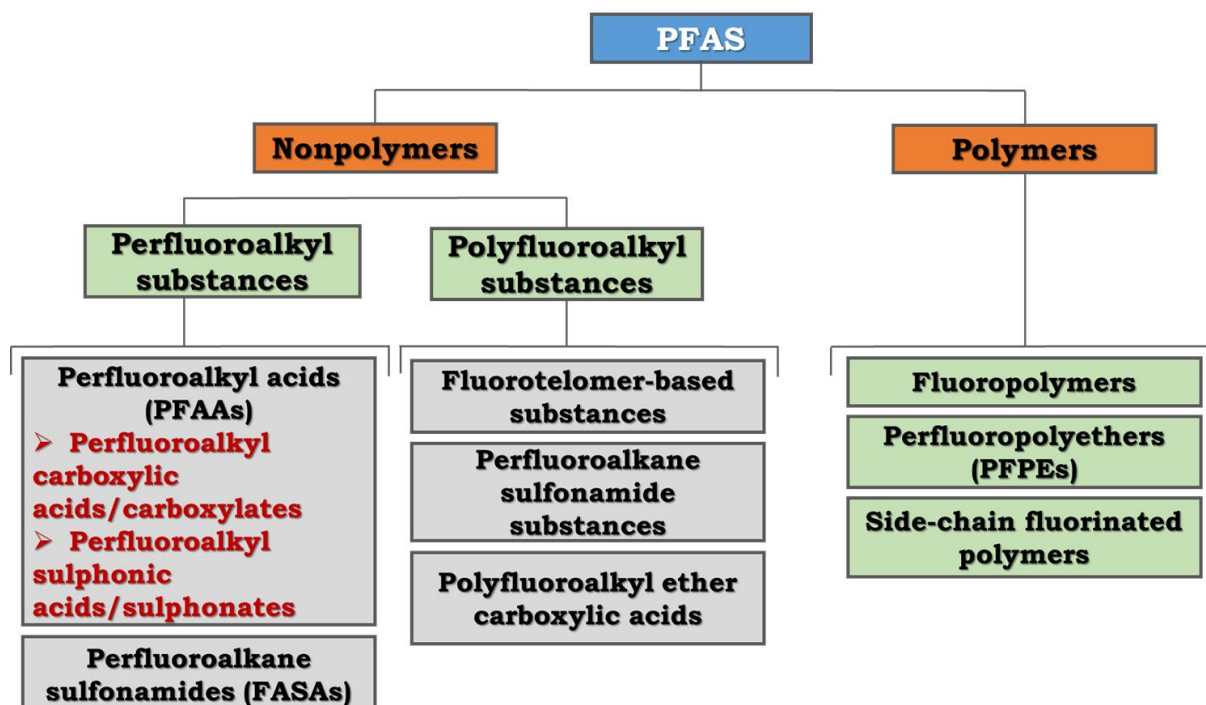


Figure 1: Summary of PFAS categorisations (readapted from ITRC [28].)

The second category encompasses polymeric ones especially polymers bearing fluorinated side chain [as poly(fluoroacrylate)s [29,30], poly(oxetane)s, poly(fluorostyrene)s], perfluoropolyethers (PFPE) [31] and fluoropolymers (FPs).

### **Fluoropolymers**

Among this last family, FPs, containing C-F bonds in the backbone, are niche specialty, bio-inert and safe polymers as well as stable thermally, chemically and against UV and ageing. In addition, they display outstanding properties (high stability against temperature, chemicals, ignition, UV-light irradiation, and other unique characteristics [32-43]), and their molar masses can reach several million  $\text{g}\cdot\text{mol}^{-1}$  [36, 39]. Their relevant combinations of properties are not matched by any of the alternative hydrocarbon polymers, hence making them so unique and valuable.

FPs can be used in many High Tech applications [32-43] (e.g., Energy, electronics aerospace and automotive industries, chemical process industry, transport, telecommunications, food and water treatment, pharmaceutical and medical devices[41-42]). In addition, major actors working on the energetic transition are taking FPs more and more into account (fuel cell membranes, binders for Lithium ion batteries, backsheets for PVs, and electroactive devices such as actuators or artificial muscles as key-examples).

FPs should only be involved in those uses which are required for health, safety, or are critical for the functioning of society and no alternatives are available [12]. Moreover, among all plastics, FPs represent a small tonnage since the production reached 320,300 tons in 2018 (compared to >400 million tons for all polymers), i.e. less than 0.1 % of conventional plastics [39-40,44] while their growth rate is 6-8% [6,40].

Advantageously, most FPs used today fulfill the 13 Polymer of low concern criteria [45-47] considered by OECD: polymer composition, molecular weight, percentage of oligomer, electrical charge, low molecular weight leachables, water/lipid solubility, Reactive Functional Groups, Functional Group Equivalent Weight, particle size, polymer stability, thermal stability, abiotic stability, and biotic stability. Furthermore, they are non-toxic; non-bio accumulative; insoluble in water and thus non-mobile; thermally, chemically and biologically stable; durable; and are not considered as Substances of Very High Concern (SVHC). While they are persistent, the lack of degradability during normal conditions of utilization and at their end of life make such specialty polymers totally inert and not harmful to human health or to the environment. These features also contribute to such PLC criteria.

### **Improvement of FP production in more environmental friendly processes**

Nowadays, FP manufacturers are aware of the potentially negative aspects of PFAS polymerization aids currently used to replace PFOA. The regulatory push from various authorities is helping the industry to swiftly move towards sustainable technologies and many efforts from these producers have been developed according two strategies:

i) to change their process in eliminating fluorinated side-products such as low molar mass-oligomers, residual reactants and other volatiles. By improving abatement techniques and PFAS surfactant recovery in the manufacture of FPs, these processes allowed the reduction of 99% of fluorosurfactant emissions since the 1990s [48] while recent studies have brought it as high as 99.99% [49]. Abundant literature is available in relation to the control and treatment of emitted PFAS from industrial processes [11,18,50] while still many studies are still ongoing [51].

ii) to produce FPs free from fluorinated polymerization aids (FPAs) in high molar masses, without any by-products as well as matching the PLC criteria. Indeed, much progress has been extended in the last decade where several FP manufacturers have modified their production using NFPA (or surfactants). An overview [52] on the technology based on NFPA over the past decade highlights that major innovations and efforts have been taken by the whole industry (such as 3M, AGC, Arkema, Chemours, Daikin, Gujarat Fluorochemicals and Solvay) in filing multiple patents which claim such a NFPA technology. Indeed, in 2022, various FP manufacturers announced the discontinuation of use of FPAs in their manufacturing [52].

Therefore, the legislation process should be focused on the use of FPAs and their emissions. In this regard, a regulatory decision tree can be suggested as below [52] (Figure 2), linked to manufacturing FPs in presence or absence of such surfactants (with process abatement to get a PFAS concentration lower than 25 ppb), essential use criteria (in terms of safety, performance, health), improvement of the process, alternatives.

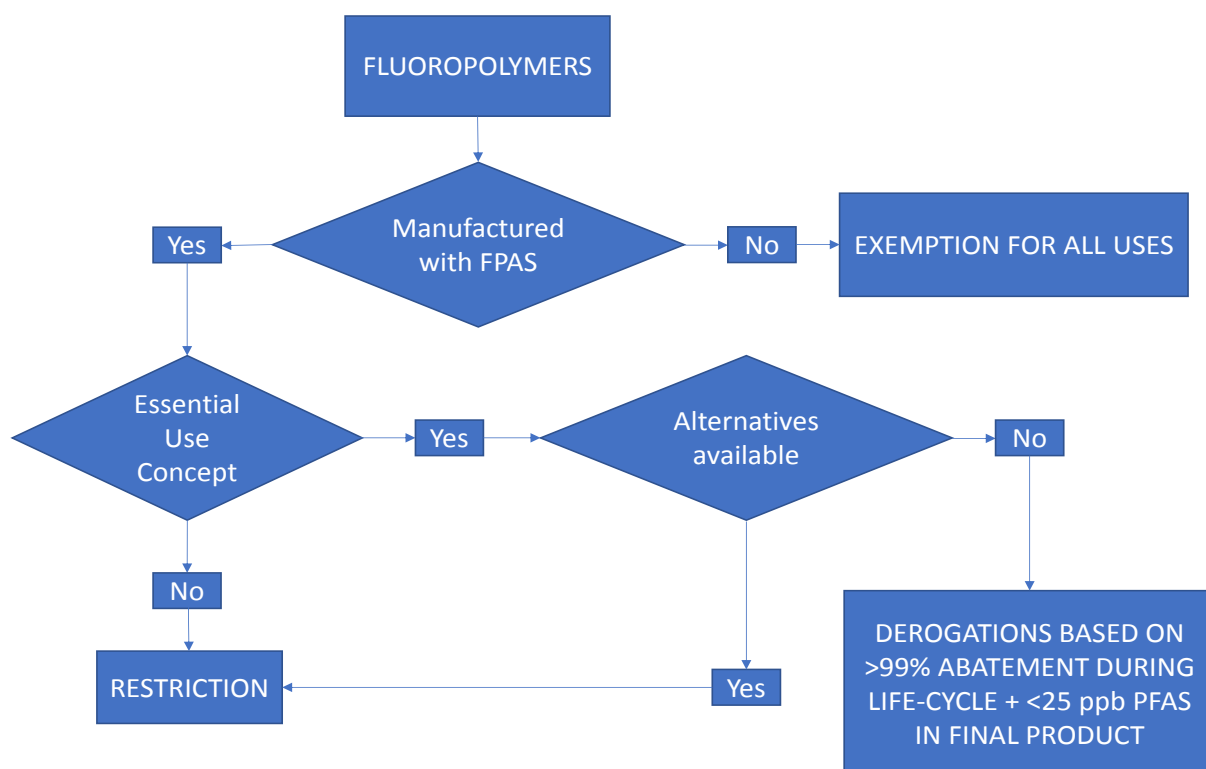


Figure 2: Regulatory decision tree for fluoropolymers (reproduced with permission from [52]).

Hence, FPs produced without any FPAs should be exempted for all uses across all industries including consumer applications as they raise no risk to Environment and to mammal and human health.

### Recycling, Elimination, Reuse, End of Life of FPs

While usages of the FPs have widely spread, waste treatment techniques cannot fulfil their increasing demand. Presently, incineration (83.5%; ca. 20.4 kt) is a way to treat these polymer wastes but, hydrogen fluoride gas, generated during the treatment, seriously damages refractory bricks of incinerators. Moreover, most remaining FP wastes are disposed in landfill (13.1%; 3.1 kt)[44], although unfilled PTFE wastes from manufactures are reused in ram extrusion applications after cleaning [53] and milling or they are decomposed into low molar mass-PTFE by heating, and reused as micro powder blend with other materials [54]. Recycling FPs represents 3.4% (0.8 kt) only. In this tendency, various companies have already found solutions to depolymerize, recycle (“close the loop” even on pilot plant) or reuse FPs [54-56].

Interestingly, Hori’s team [57] reported an effective method to allow complete defluorination of FPs (including PVDF, as well as copolymers of VDF and of TFE, as perfluorosulfonic acid membranes) to  $F^-$  ions, by means of superheated water (or “pressurized” hot water at ca. 100-

300 °C). This water is recognized as environmentally benign in waste management because it enables to transform hazardous chemicals into harmless products or to generate value-added compounds [57]. The procedure may also occur in presence of either potassium permanganate as oxidizing reactant or alkali reagents (NaOH or KOH) [58]. The carbon content was decomposed into CO<sub>2</sub> in the gas phase whereas HCO<sub>3</sub><sup>-</sup> was produced into the reaction solution. In the presence of Ca(OH)<sub>2</sub>, the released fluoride anions led to the formation of "artificial fluorspar" (CaF<sub>2</sub>) as a well-known source of the Fluorine Chemistry process, hence closing the loop as an attractive technology.

## **Conclusions**

PFASs have recently been under heavy pressure from US and EU regulators and increasingly forecast by press and media because of environmental concerns and BPTM issues. Among them, FPs are specialty polymers of high molar-masses, are water insoluble, non-mobile, non-bioaccumulative and non-toxic, and do not release any toxic side products and oligomers under conventional uses. Hence, most FPs (>96% of global production) fulfil the conditions set out by the OECD to meet the definition of Polymers of Low Concern and do not fall into the category of most harmful chemicals. More recently, the main concerns have been addressed, both in using NFPA and controlling >99 % PFAS emissions linked to appropriate abatement technology of the industrial productions of FPs. As a result, FPs produced with NFPA technology has led to a commercial success and further motivates the industry to work harder in this direction. Their applications deal with health, safety, performance and functioning of the society. Furthermore, alternatives to FPs, which should be assessed based on the criteria such as performance, accessibility, resource efficiency, waste generation, cost, availability, stability and lifetime, do not exist. In addition, various solutions are searched for recycling, reuse, circular economy and life cycle analysis. For example, mineralization of FPs generating F<sup>-</sup> ions would contribute toward closing the loop of the Fluorine element. Finally, the Fluoropolymer industry in Europe [59] has evaluated the contribution made by the FP manufacturers in terms of revenue, investment and employments, while many more significant benefits are generated along the value chain via the use of FPs in various critical applications. These high value added polymers must be separated from conventional PFASs and are essential for everyday life.

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## ACKNOWLEDGMENTS

The author thanks coworkers cited in the list of references below and the French Fluorine Network (GIS).

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