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## Applications, Pollution, Toxicity and Bioremediation of Acrylamide

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### ABSTRACT

Acrylamide has many chemical and environmental uses. The main or principle use of acrylamide is in the production of high molecular weight polyacrylamides with up to 99.9% of acrylamide is estimated used to produce polyacrylamide. Acrylamide, in the form of polyacrylamides are used as binders and retention supports for fibres and to retain pigments on paper fibres, to decrease soil erosion, in furrow irrigation systems and in the formulation for the herbicide glyphosate. Acrylamide is a neurotoxic agent and its pollution remains an important global awareness due to its acute toxicity. Due to its toxicity, the maximum permissible limit for acrylamide containing in drinking water of Malaysia is 0.0005 mg/L. Its uses, toxicity, pollution and bioremediation are the subject of this mini review.

### INTRODUCTION

Acrylamide is the foundation for the polymer, polyacrylamide (PAM). The production of commercial polyacrylamide may very well be contaminated with acrylamide, its toxic monomer. Consequently, suggestions have already been focused on the quantity of acrylamide that is contained in polyacrylamide. As an example, a restriction of 500 ppm in polyacrylamide formulations is utilized in agriculture or water treatment method. Some other uses of acrylamide include as sewage-flocculating agent [1], as adhesives, and in tunnels and dams stabilization [2]. Incidentally, one of the identified cases of acrylamide toxicity is due to its application for stabilizing of tunnel in Sweden. In Hallandas village, Sweden, there is a case of cows and fish death caused by the acrylamide pollution resulted by the acrylamide that pumped to the surrounding soil [3].

In our country, Malaysia, large amounts of polyacrylamide are used annually for drinking water treatment. For instance, Kuching Water Board uses approximately 800kgs of polyacrylamide additive (Superfloc and Prestol 2530) yearly in the Sarawak state [4]. In agriculture field, acrylamide is introduced directly into agricultural soil via glyphosate application, where 20-30% polyacrylamide is added in commercial formulations as dispersing agent [5].

Research has shown that under environmental conditions, acrylamide is produced from polyacrylamide degradation and the half-life of acrylamide in soils and water bodies range from weeks to months [6]. In spite of well-known toxicity of acrylamide due to the acrylamide from the degradation of polyacrylamide and its presence as impurity in PAM, there is no investigation for acrylamide in the environment conducted by the Malaysian Department of Environment (DOE) and it is not part of the Interim National Quality Standards for Malaysia which listed maximum permit residual limits for aldrin, lindane, paraquat, chlordane, endosulfan and heptachlor.

There is no acrylamide allowed to be present in drinking water by the US EPA Primary Drinking Water Standards whilst, for the 2003 World Health Organisation (WHO), the allowable residual limits of acrylamide for the drinking water quality are at maximum of 0.10 µg/l [7]. Acrylamide is used as the main resin component in laminated wood and also often use in the plastic and printing production and from such industries, acrylamide can be found abundance as high as 1000 mg/L in the waste water according to Rogacheva and Ignatov [8]. As acrylamide is a neurotoxicant, has some carcinogenic and teratogenic activities its presence in the environment is a hazard and its remediation must be sought [2,9–11].

### Acrylamide

The chemical formula of acrylamide is  $C_3H_5NO$ . Its other names include propenamide, acrylic amide, vinyl amide and 2-propenamide. Acrylamide is a compound which is widely used in industry to make polymeric substance that is utilized in numerous items. In its polymeric form it is called polyacrylamide, and is nontoxic compared to its monomeric form [12]. It has many uses in our everyday life [13]. It exists in either liquid or crystalline forms. The solid pure form is colorless to white. Acrylamide is soluble in dimethyl ether, water, methanol, ethanol, but is insoluble in benzene and heptane. The molecular mass of acrylamide is 71.08. its maximum solubility in water at 30°C is 2155 g/L. Its boiling and melting points are 125°C and 84.5°C, respectively at 1 atmospheric pressure [13]. The specific gravity of acrylamide at 30°C is 1.122 g/cm<sup>3</sup> [14].

### Uses of acrylamide and polyacrylamide

Since the last century, acrylamide and its analogues have been used for many chemical and environmental purposes. The main or principle use of acrylamide is in the production of high molecular weight polyacrylamides which are customized to construct various compounds with different physical and chemical properties to suit the industrial needed. Up to 99.9% of acrylamide is estimated used in the polyacrylamide manufacture in Europe Union. Acrylamide is often utilized in both industrial sectors and medical laboratories since it is extremely reactive and water soluble. In the production of paper and pulp, the polymeric forms i.e. polyacrylamides are utilized as retention supports and as binders for fibres and also to enhance the preservation of pigments on the fibres. Acrylamide is also utilized to lower the erosion of soil, which its usage has been increasing over the years. Polyacrylamides is added onto the irrigation system to avert the deterioration of agricultural furrows, which can reduce as much as 94% of furrow erosion. [15].

In mines, great deal of acrylamide can be used on location to generate polyacrylamide gel which forms the grouting agent needed to seal mineshafts. Polyacrylamides are utilized as flocculants to separate solids from aqueous solutions. Additionally it is utilized in the removal of commercial waste materials and in the purifying of water resources [16]. Polyacrylamide permits a lot more concentrated sludge should they be utilized as dewatering or sludge conditioning materials and is more efficient than inorganic coagulants. The polymers form heavy aggregates when combining with particles and the resultant flocs rapidly settle out of solution as well as leaving a crystal-clear supernatant. The higher the molecular weight of the cationic polyacrylamide used, the more effective the polymer is [17]. In water treatment process acrylamide remains in the supernatant after flocculation with polyacrylamides, as acrylamide has high-water solubility and it tends to be inefficiently absorbed by sludge or sediment and sludge. The degradation of polyacrylamide is minima as the polymer is recalcitrant and at best only 10% will be degraded per year through physicochemical and biological actions [18–24].

There is not any data concerning the use of acrylamide and polyacrylamide obtainable in Malaysia to date; even there exists a range of industries in Malaysia which use polyacrylamide. The industrial sectors which use by far the most polyacrylamide is in the treatment of waste water, paper, and pulp processing. golf courses in Malaysia uses polyacrylamide to strengthen the foundation of the man-made ponds. This is the main route or cause for the toxic contamination of subterranean water which has triggered a number of poisoning and problems of the nervous system. Numerous golfer, caddies, as well as inhabitants have been discovered to experience irritations, skin illnesses as well as

other hypersensitive symptoms [2,9,13,25,26]. In Malaysia, glyphosate in the commercial form of Roundup® is used for the control of a wide range of broad-leaved weeds and grasses in agricultural estate crops such as rubber, oil palm and cocoa [27–37] in the form of the glyphosate (Roundup™) formulation of which polyacrylamide is added as a dispersant and as a stabilizer (25-30% solutions). Estimated local usage gives rise to an estimated 2 million liters of polyacrylamide being dumped to the soil yearly and eventually runoffs into rivers and water bodies.

### Acrylamide as pollutant

Industrially, acrylamide production uses a close system that allows very minimal pollution. This means that most of the pollution attributed to acrylamide comes from its uses in the mining, erosion control, tunneling works and agricultural sectors as herbicide's formulation or in the stabilization of agricultural soils. Due to this, acrylamide presence in the environment continues to be detected and its presence is being reported frequently nowadays [38–49].

The low vapor pressure of acrylamide coupled with its high-water solubility, makes it an uncommon source of air contaminant. Pollution of drinking-water by acrylamide is largely due to its use in the form of polyacrylamide flocculants of which acrylamide forms a significant residue [46]. Legislation has put a maximum acrylamide monomer in polyacrylamide for the use of potable water treatment at 0.05% (w/v) monomer. Acrylamide is stable in tap water for more than two months and will be present in drinking water. The recommended maximum permissible limit for drinking water for acrylamide in Malaysia is 0.0005 mg/L [50].

In the agricultural sector where polyacrylamide is combined with the herbicide glyphosate as additive (25% to 30% solutions) is the source of acrylamide pollutant in agricultural soils and irrigation and drainage systems [18–24]. It has been demonstrated that glyphosate formulation can be more toxic than glyphosate alone. Roundup has been found to be thirty times more toxic to fish than the compound glyphosate itself [51]. The depolymerisation of polyacrylamide to acrylamide, according to Smith *et al.* studies, is strongly affected by light [52].

### Toxicity of acrylamide

Acrylamide is toxic to the nervous system, carcinogen in laboratory animals, and also suspected to be carcinogen in humans [25]. In mouse, it has been shown that acrylamide binds to the DNA and mouse protamine at the spermiogenic stages and is suggested to cause genetic damage [53]. Acrylamide causes peripheral neurotoxicity, prenatal lethality, mutagenicity, clastogenicity, endocrine-related tumors and male reproductive toxicity in rodents, [26]. Yang *et al.* show the potential mutagenicity of acrylamide on *Salmonella* strains TA100 and TA98 [54]. An increase in the incidence of bone marrow's chromosomal aberrations is seen in mice exposed to acrylamide at dose of  $\geq 50$  mg/kg via intraperitoneal injection. Yet, in mice that subjected to acrylamide intraperitoneally at doses of up to 125 mg/kg shows no significant increases in the chromosomal aberrations frequency in the lymphocytes [55].

Acrylamide also affect male rat's reproductive system through causing several histopathological lesions in the seminiferous tubules. Human exposed to acrylamide via breathing the dust or absorption via skin, experience burning feeling or rash upon contact. Human's nervous system is affected which leads to weakness and numbness in the feet and hands, cause heavy sweating, imbalance and slurred tongue [25].

Due to its high-water solubility, acrylamide is absorbed through the lungs, intestinal tract, the placental barrier and the skin. A biomarker for acrylamide exposure is hemoglobin adducts which have been used to estimate the internal dose in the general population exposed to acrylamide through their occupations. At an acrylamide manufacturing plant, 41 workers exhibiting neurotoxicity index that are correlated with the acrylamide biomarker hemoglobin adducts [56]. In a China acrylamide manufacturing factory, hemoglobin adduct levels have been found to increase indicating a very high exposure to acrylamide [57]. In Japan, several cases of acute acrylamide poisoning due to a contamination of the water supply with acrylamide has been reported. Igisu *et al.* [58] reported that in a well-water where the waters are contaminated from a grouting operation 2.5 meters resulted in acrylamide levels as high as 400 mg acrylamide/L. Five people that ingested the contaminated water well experienced acrylamide toxicity such as confusion, memory disturbances, disorientation, hallucinations and truncal ataxia.

Acrylamide enters body via inhalation with contaminated air or consumed with contaminated food or water. Besides that, it also can be absorbed through unbroken skin, lungs and mucous membranes and the gastrointestinal tract. But it will not remain in the body because it will be removed with urine [39,44,45]. The acrylamide action's mechanism is greatly enhanced through its extensive allocation in body fluids and fairly even distribution throughout body organs. Even though acrylamide can be metabolized rapidly and excreted following exposure, the reason why it can possess hazards to workers and human is because of its high reactivity with proteins [59].

### Degradation of polyacrylamide to acrylamide

Specific usages of acrylamide polymers and copolymers include as soil conditioners, flocculants in the dewatering and thickening of sludge, as bone cement binder and in sugar refinery [19,22,23]. It is estimated that in the United States alone a total of 25-50 million kilograms of polymer are utilized yearly. When acrylamide is released to the environment, depending on where the process occurs, acrylamide undergoes several of the degradation and removal processes producing the toxic acrylamide [48].

### Bioremediation of acrylamide

Bioremediation is the use of plants or biologically active agents, microorganisms as a way to break down, sequester and conjugate ecological pollutants [60–70]. Usually the use of plants for this process is called phytoremediation. Certain microorganisms known to have the ability to degrade contaminants in the environment. Advantages of the bioremediation application include the easiness and timing of application and its ability to target specific pollutants. Besides that, bioremediation also can decrease sludge volume and ecological hazard. Even though the bioremediation is considered a new technology presently, the microorganisms have been used for no less than 100 years for the treatment and conversion of waste products.

There are three types of bioremediation predominant in the industry today; first of all is natural attenuation; secondly is biostimulation and the last one is called bioaugmentation. Natural attenuation is the simplest method of bioremediation. For this method, monitoring of the contaminated site for determining the levels of pollutants as a passive way to ensure regulators that the natural removal processes are ongoing. In biostimulation, an alteration to the site in the form of adding nutrients is required in order to boost the local population of the microorganism leading

to an effective remediation of the contaminants. Nutrients that are usually added include nitrogen, phosphorus as well as trace elements. In addition, the adjustment of the soil's pH maybe necessary to make the site's pH suitable for the growth of the bioremediating microorganisms [60–70]. Lastly, bioaugmentation is another bioremediation approach where the absence of native communities due to the overwhelming presence of the hazardous contaminants is augmented or fortified with nonnative microorganisms to kick start the bioremediation process before local microorganisms that are accustomed to the soils conditions take over [60,71–76].

Acrylamide may contaminate groundwater as it is extremely mobile in aqueous environments. Acrylamide has a lower degradation rate in sandy soils than clay soils. Despite this, acrylamide degradation in water is considerably slower than soil [43]. Many species of microorganisms have the ability to degrade acrylamide under dark or light, aerobic or anaerobic conditions. To date numerous acrylamide-degrading microorganisms have been isolated and characterized [4,77–92] and some of the characteristics of these microorganisms is shown in **Table 1**. Many of these microorganisms can degrade aliphatic nitriles and amides and a majority found difficulty in degrading aromatic amides or nitriles.

**Table 1.** Acrylamide-degrading microorganisms.

Microorganism	Optimal conditions for growth	Other substrates	Ref.
<i>Pseudomonas putida</i>	pH 7.0 and 25 °C	Acetonitrile, glutaronitrile, butyronitrile, propionitrile, isobutyronitrile, methacrylonitrile, valeronitrile, succinonitrile, and amides such as butyramide, acetamide, propionamide, isobutyramide, succinamide and methacrylamide	[77]
<i>Cupriavidus oxalaticus</i>	pH 7.0 and 30 °C	Toerant to 60 mM (4,264 mg/L) acrylamide. Other amides such as formamide, acetamide, isobutyramide and urea (aliphatic amide), support growth while there was no activity on benzamide and nicotinamide (aromatic amide)	[92]
<i>Rhodococcus</i> sp.	pH 7.0 and 30 °C	Acrylamide (62.8 mM or 4,463 mg/L) was supplied as the sole growth substrate. Can grow on aliphatic amides such as acetamide, butyramide, propionamide, and isobutyramide	[78]
<i>Klebsiella pneumoniae</i>	pH 7.5 and 28 °C	aliphatic nitriles containing 1 to 5 carbon atoms including acrylonitrile and aromatic nitriles such as benzonitrile as the sole source of nitrogen and propionamide and acetamide as the sole source of both nitrogen and carbon	[93]
<i>Paracoccus</i> sp.	pH 7.0 30 °C	acetonitrile, acrylonitrile, propionitrile, valeronitrile, benzonitrile and the amides dimethylformamide, N,N-dimethylacetamide, N,N-dimethylfulfoxide, dimethylamine, methylamine, methanolamide, ethanolamide, acetamide, propionamide, valeramide, formamide, valeramide and acrylamide supported the growth of the culture with the release of ammonia, while aromatic amides did not support the growth of <i>Paracoccus</i> sp.	[94]

**Table 1.** Acrylamide-degrading microorganisms-continue.

<i>Klebsiella pneumoniae</i> NCTR 1	pH 7.2 and 30 °C	aliphatic amides such as acetamide, butyramide, methacrylamide, propionamide and succinamide) as the sole nitrogen source. Amidase enzyme is a monomer with an apparent molecular weight of 62,000. The temperature and pH for optimal enzyme activity were 65 °C and 7.0, respectively.	[79]
<i>Rhodotorula</i> sp. strain MBH23 KCTC 11960BP	between 27 and 30 °C and between pH 6.0 and 8.0	Tolerant acrylamide concentration is 1500 mg/L. Amides such as nicotinamide, 2-chloroacetamide, methacrylamide, propionamide and acetamide supported growth as nitrogen source. Growth inhibited by mercury, chromium, and cadmium	[95]
<i>Pseudomonas</i> sp. strain DRYJ7	At 15 °C and between pH 7.5 and 8.5.	Antarctic bacterium. Tolerant to 2500 mg/L acrylamide.	[4]
<i>Bacillus cereus</i> strain DRY135	between 25 and 30°C and between pH 6.8 and 7.0	Tolerable acrylamide concentration is 1500 mg/L. Can grow on other amides such as nicotinamide, methacrylamide, propionamide, acetamide and urea. Cannot grow on 2-chloroacetamide.	[4]
<i>Burkholderia</i> sp. strain DR.Y27	between pH 6.0 and 8.0. and at 30°C	Can grow on aliphatic amides such as 2-chloroacetamide, nicotinamide, methacrylamide, propionamide, acetamide and urea	[96]

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