

MICROBIAL DETERIORATION OF PAPER MATERIAL

—A Literature Review

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The microbiodeterioration of paper material like books, archival material, manuscripts both illustrated and written, decorative wall papers, etc. is a serious problem throughout the world in museums, libraries, archives, etc. where these materials are placed. (See, 1917; 1919; Omelianskii, 1925; Bakhatin, 1928; Marcella, 1936; Bryan, 1937; Anchabaze, 1949; Armitage, 1949; Kowalik, 1952, a and b; 1956; 1962; 1963; 1972; 1977; 1980; Belyakova, 1953; Rybakova, 1953; Kathpalia, 1960; Nyuksha, 1961; Gallow, 1963; Deshpande and Mantri, 1966; Hughes, 1968; Flieder, 1969; Lazar and Dumitru, 1972; Fischer, 1977; Lelis, 1980; Miller and Harold, 1981; Nair, 1981; Camargo, 1982; Dhawan and Agrawal, 1986). In India in addition to the above materials, different varieties of miniature paintings were also done on paper which are known as miniature paper paintings (Agrawal, 1984). This problem needs special attention where the climate is hot and humid (Nair, 1972 and Agrawal, 1977).

The basic component of paper is cellulose. In addition to cellulose, it also contains lignin, hemicellulose, pectins, waxes, tanins, proteins, minerals etc. Further more paper may contain resinous materials, fillers, and dyestuffs. Various impurities are added during its productive cycle which forms the basis for microbial nutrition (Kowalik, 1980).

Micro-organisms growing on paper are actinomycetes, bacteria and fungi. These organisms multiplying on paper utilize intentional additives of adventitious impurities and prepare substrate for typical cellulolytic micro-organisms.

Scientists after prolonged observations have mentioned that fungi are exceptionally abun-

dant on the earth, they have a variety of fermentive complexes and high ecological adaptability which give them the leading place in damaging paper material.

It has been experienced by microbiologists that abundance of micro-organisms takes place after the ravages of flood.

Omelianskii (1953) reported after the flood of river Neva in Colombia 1924 where 62,000 books were destroyed, a number of fungal forms were isolated.

In Tuscany, in 1966, over 30 libraries and archives suffered extensive damage by fungi after disastrous flood. In Lisbon, in 1967 many valuable manuscripts belonging to the Calouste Gulbenkian were also damaged (Reed, 1972). Similar damages have been reported in the USA, Canada and Poland libraries, archives and museums, (Fisher, 1977; Cunha, 1977; Martin, 1977; Stanojlowic, 1978).

In India Narayanan (1963 a and b) reported microbial attack on paper after the flood in Poona 1961.

Such disasters involve, rapid movement of flood waters which carry with them materials derived from manifold activities of modern cities, chemicals, oils and different biological agents from sewage which additionally develop rapidly, after contact with the constituents of atmosphere (Reed, 1972).

Microbial spores which are always present in air light on the paper material, and after getting proper humidity (more than 75%) and temperature (25-35°C), start to germinate. Anchabagze (1949) published a report of an investigation of manuscripts at the Georgian State Museum. He enlisted 17 species of fungi and considered air to be the principal cause of the abundant contamination of the material.

Nyuksha (1954 a and b) isolated fungi from books, and at the same time a mycological analysis of the air in the library rooms. Finally the relationship between the mycoflora of paper were studied. Then an investigation of finished paper and new books was carried out and the change in contamination of books was observed as a function of their handling and library processing. On the basis of these studies, the investigator was able to get a clear picture of the constitution, distribution and changes of mycoflora, the manner of their deve-

lopment under different conditions and in different materials during various stages in the existence of the stores.

Nyuksha (1956) explained that many species are known which take no active part in the destruction of paper fiber but are found in large quantities. The particular species referred to was *Mucorals*, *Monilia sitophyla* and many representatives of *Penicillium* variety (asymetrica). These fungi, widely distributed in nature, light on books also in a large number from the surrounding air, so that it is difficult in practice to protect the samples from them as completely as might be desired. Therefore, it was suggested that in the characterisation of book-mycoflora one must not attribute primacy to this group of fungi on the basis of the prevalence but on the ability of the fungi to destroy book material.

In India for the first time aerobiological studies inside the library was conducted by Tilak and Vishwe (1976). They indicated the presence of *Aspergillus*, *Torula*, *Penicillium*, *Trichoderma*, and *Chaetomium* spores.

It is a well known fact that books, and manuscripts, etc. stored under improper atmospheric conditions are likely to become mouldy (Rybakova, 1953). On the basis of his wide studies, 92 different strains of fungi belonging to the 16 genera were reported and a correlation was established between the number of damaged books and the condition of storage. Aleksi (1965) studied the mycoflora of an archival store-room.

Under normal book storage conditions, book bindings are the first to take on moisture from the air while the interior parts do so later. In other words when the RH of the air increases in the room the spores can germinate most quickly on the bindings. It has sometimes been observed that books become mouldy even at a RH of 50-60% which is the recommended RH for keeping paper material. Repeated experiments were also carried out by Thomas and Beckwith (1935) by planting fungi on filter paper to which had been added small amounts of a nutrient solution, it was observed that on various papers of known sizing contents the spore would not germinate, when the RH surrounding the cultures was kept below 75%. It was also noted that with any increase in humidity above 75%, marked acceleration of growth takes place.

Book mould therefore is produced first of all by the type of spores requiring very moderate amounts of moisture or by those which are capable of germinating at relatively low humidity.

They are for the most part adopted to the bindings and in the first stage do not participate in the destruction of cellulose. Somewhat later, other species appear and the penetration of the mould into the depth of the material begins. The book-mycoflora becomes more diversified and fungi capable of destroying cellulose requiring greater humidity than the first occupier of bindings assumes the principal role.

A number of fungal genera have been reported from various paper materials (Sartory, *et al.*, 1934; 1935; Beckwith *et al.*, 1940; Waksman, 1940; Kowalik, 1952 a; Belyakova, 1961; Nyuksha, 1954 b; 1956; Fausta, 1963; Flyate, 1968; Dohlakia and Chhalpar, 1980; Leznicka, 1980; Strzelczyk and Leznicka, 1981; and Lea Nol and Knneth 1983). Some of them are given below:

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|--------------------------|---------------------------|
| 1. <i>Acrothecium</i> | 2. <i>Alternaria</i> |
| 3. <i>Aurobasidium</i> | 4. <i>Aspergillus</i> |
| 5. <i>Botryotrichum</i> | 6. <i>Byossochlamys</i> |
| 7. <i>Cephalosporium</i> | 8. <i>Chaetomium</i> |
| 9. <i>Cladosporium</i> | 10. <i>Curvularia</i> |
| 11. <i>Epicoccus</i> | 12. <i>Eidomella</i> |
| 13. <i>Fusarium</i> | 14. <i>Gliomastix</i> |
| 15. <i>Hormodendrum</i> | 16. <i>Humicola</i> |
| 17. <i>Memmoniella</i> | 18. <i>Monilia</i> |
| 19. <i>Myrothecium</i> | 20. <i>Mucor</i> |
| 21. <i>Neurospora</i> | 22. <i>Paceilomyces</i> |
| 23. <i>Penicillium</i> | 24. <i>Phialophora</i> |
| 25. <i>Phoma</i> | 26. <i>Pestalotia</i> |
| 27. <i>Pelicularia</i> | 28. <i>Pullularia</i> |
| 29. <i>Spicaria</i> | 30. <i>Scopulariopsis</i> |
| 31. <i>Stemphylium</i> | 32. <i>Stachybotrys</i> |
| 33. <i>Trichoderma</i> | 34. <i>Verticillium</i> |
| 35. <i>Ulocladium</i> | |

Joshi (1958), Narayanan (1963 a and b) reported species of *Stachybotrys lobulata* Berk. *Trichocladium opacum* (Crd.) Hughes, *Monotospora cuneiformis* (Rich) Sacc. Tilak and Jadav (1970) isolated *Melanomma marthwadensis* from the decaying brown paper of Maharashtra. Deshpande and Mantri (1966) reported *Cunnighmella* sp. from the rotten filter paper. Nair (1977) conducted studies on fungi growing on old manuscripts and identified *Aspergillus niger*, *A. glaucus*, *A. flavus*, *A. restrictus*, *Alternaria* sp., *Cladosporium* sp., *Curvularia* sp., *Fusarium* sp., *Penicillium* sp., *Trichoderma* sp.

Recently Dhawan and Agrawal (1986) have studied fungal flora of miniature paper paintings and lithographs of State Museum, Lucknow, and have isolated 23 fungal species which were-*Aspergillus chevalieri* (Mangin) Thom and Church, *A. flavus* Link., *A. melleus* Yukawa., *A. nidulans* (Eidam) Winter., *A. niger* van Tieghem., *A. stellatus* Curze. Rend. *A. sydowi* (Bainier and Sertory) Thom and Church., *A. terreus* Thom., *A. ustus* (Bainier) Thom and Church., *A. versicolor* (Vuillemin) Tiraboschi., *A. wenti* Wehmer., *Alternaria alternata* (Fr.) Keissler., *Cephalosporium acremonium* Corda., *Chaetomium globosum* Kunze. ex stend., *Cladosporium cladosporioides* (Fres) de vreis., *Fusarium oxysporum* Schlechtendalid., *Paceilomyces varioti* Bainier., *Penicillium chermesinum* Biourge., *P. chrysogenum* Thom., *P. citrinum* Thom., *P. coryophillum* Dierckz., *P. frequentans* Westling, *Trichoderma viridi* Pers. ex Gray.

Fungi growing on paper produce black, brown, and yellow pigmentation which penetrate the paper (Nyuksha, 1956; Kathpalia, 1960 and Kowalik, 1962; 63 and 80) these stains are difficult to remove by the usual restoration methods.

Nyuksha (1956) studied the pigmentation of different fungi growing on paper and reported that the colour of the pigment may change depending on the condition of the growth and the properties of paper. It was explained that *Gymnoascus sitosus* forms no pigment at all on paper, with colophony. On baryta paper, it produces a fine crimson red colour and on rag paper, with starch sizing, a lemon yellow. In the same way *Penicillium pinophilum* forms a red pigment on rag paper with gelatin sizing and on filter paper a pink shade.

Later on Kowalik (1962, 1963) isolated the following fungi and also observed the pigmentation on paper.

Phoma humicola Gilman and Abbott. grey brown or black spots.

Chaetomella horrida, Oudemans-grey brown spots.

Cephalosporium acremonium, Corda-orange to rose tints according to pH.

Trichoderma viride, Pers. ex. Fr-stained light green.

Aspergillus fumigatus, Fresenius—In the beginning grey, afterwards green, and at last dull brown.

A. Wentii, Wehmer-light rose.

A. niger, Van Tieghem-minute black superficial colonies will appear.

Paecilomyces varioti, Bainier - light yellow brown shade.

Spicaria, Harting - gave lilac spots.

Botryotricum atrogriseum, Van Beyma - steel grey shades.

Sepedonium, Link - Grey spots.

Acrostalgnus cinnabarinus, Corda - stained paper rusty shades.

Eidamella spinosa, Matruchot and Dassonville - developed amaranthine spots.

Mycogone nigra, (Morgan) Jensen - developed green spots.

Stachybotrys atra, Corda - stained and covered paper with black or greenish black growth with greenish grey or brown zones.

Torula, Persoon - caused black spot.

Gliomastix convoluta, (Marchal) Mason-stained paper black.

Gliocladium roseum (Link) Thom. stained paper rose.

Trichocladium, Harz-stained greenish yellow or greenish rusty depending on strain and pH.

Cladosporium herbarum, (Persoon) Link-covered paper by blackish green spots-

Helminthosporium, Link-stained paper greyish white.

Curvularia lunata, (Walker) Boedijn-covered paper by black spots and afterwards developed larger spots.

Trichothecium roseum, Link, developed on paper fluffy colonies, growing well. They formed at the beginning white afterwards pale rose stains.

Acrothecium, Preuss-stained it olive green.

Alternaria, Nees. and *Stemphylium*, Wallroth-stained brown greenish spots.

Epicoccum nigrum, Link-black colonies adhering tightly to the fibers.

Fusarium, Link-discoloured paper giving violet tints, blue spots, pink violet spots and yellowish pink spots.

Nyuksha (1976) demonstrated that during the cohabitation with other fungi *A. flavus*, stimulates pigmentation of other fungi like *Myxotrichum deflexum* on paper suppresses the growth of *Verticillium tenerum*, *Stachybotrys chartarum*, etc.

A Number of actinomycetes strains injurious to old books, manuscripts and archival documents have been interestingly reported by Czerwinska, *et al.* (1953); Kowalik and Sadurska (1956). These also produce variety of colours.

Several bacteria were also isolated from paper material where the relative humidity was quite high (more than 85%). *Pseudomonas*, *Bacillus*, *Micrococcus*, *Saprocytophage*, *Cytophage* and *Cellvibrio* (Firipi and Mazzucchetti, 1963; Berg *et al.*, 1972; Lazar and Dumitru, 1972; Strzelczyk Leznicka, 1981. Oppermann and Wolfson (1961) isolated *Aerobacter*, *Chromobacterium*, *Clostridium*, *Flavobacterium* and *Klebsilla*. They intensely discolour paper.

All these micro-organisms produce different organic acids like oxalic, fumaric, succinic, citric etc., and reduce the pH of the paper. The optimum acidic values of nutrient media for fungi are mostly below pH 7 (Lilly and Barnett, 1953). The upper and lower may be different for different fungi. Bacteria need pH-6.8-8.0, Actinomycetes grow best at pH-5.0-8.0.

The utilization of cellulose for the nutrition of fungi is well known for a long time. Eggins and Pugh (1962) have demonstrated the utilization of cellulose by fungi by using cellulose-agar media.

The complete degradation of cellulose in paper is effected by two groups of celluloses: endoglucanases which hydrolyse internal beta-1,4 glucosidic linkages and exoglucanases which split off mono and disaccharide units from the non reducing end of a cellulose chain. (Berghem and Petterson, 1973; Eriksson and Petterson, 1975; Shikata and Nizisawa, 1975). The intermediates of cellulose degradation have been found to be oligosaccharides with mucose properties (Gascoigne and Gascoigne, 1960; Berg *et al.*, 1972; Rosa and Strzelczyk, 1979).

In the formation of muscilage, fungi and cellulolytic bacteria as well as bacteria forming muscilagenous capsules are responsible (Martin and Dobson, 1945; Gascogine and Gascoigne 1960; Sanborn, 1965; Turner, 1967; Hughes, 1968; Rosa and Strzelczyk, 1979; Strzelczyk and Leznicka, 1981). These muscilagenous substances may be assumed to play a vital role in the consolidation of a book (affected by flood or high moisture) into a block.

Siu and Reese (1953) introduced data concerning the destruction of different forms of cellulose by several type of fungi and remarked that the course of process depends in considerable measure on the properties, purity and composition of cellulose. However, Belaya, *et al.*, (1964), Nyuksha (1964), Zagulyaeva and Flyate (1973) Rosa and Strzelczyk. (1979), explained that the intensity with which paper is degraded by cellulolytic micro-organisms is affected by the presence of natural glues in the paper.

Strzelczyk and Leznicka (1981) studied succession of micro-organisms on the XVIIth and XIXth century paper and they observed that in the XVIIth century paper the first fungi to appear utilized first of all the glutin glue used in the manufacture of paper, e.g.

Verticillium sp. this fungus is known to have poor cellulose degrading ability (Gupta and Heale, 1970). Next fungi to appear on the sample had a full set of cellulolytic enzymes (*Chaetomium*, *Trichoderma* and *Penicillium* sp.). The genera to appear the last were adopted to the utilization of the products of cellulose degradation (*Rhizopus* and *Cladosporium* sp.) However, in case of a 19th century book the fungi isolated were known to have high cellulolytic activity but they showed very poor vigour which may be due to colophony and certain amount of lignin present in the paper.

Basu and Ghose (1962), Cowling and Brown, (1969), Berg *et al.* (1972) demonstrated that delignified short cellulose fibers are degraded more easily than long fibers with a higher lignin content.

In the restoration of paper material, paper is laminated with polyethylene at a temperature of 110°C, it was also found to be attacked by fungal form *A. flavus* (Nyuksha, 1983). *A. flavus* uses inaccessible source of carbon including synthetic and natural complex ethers and carbon chain polymers like polyvinylacetate, polyvinyl chloride, Polyvinyl alcohol, etc. (Greathouse and Wessel 1954 ; Davis, 1963).

Paraffined paper which is also used in restoration of books is easily affected by *A. flavus*. It loses the water repellent property, becomes yellowish (Touson, 1950 ; Nyuksha, 1983) and ultimately degrades paper.

As a result of microbial action on paper there is a considerable reduction in the alpha-cellulose (Pietrykowska Leznicka, 1979) and there is an increase in copper number (Nyuksha, 1956 ; Zagulyaeva, 1968, Strzelczyk and Leznicka, 1981). The weight of the cellulose usually shows only an insignificant decrease but its mechanical strength decreases considerably.

A characteristic brown or rusty spotted discolouration found commonly on old books, manuscripts etc. and even not so old books is referred to as 'foxing'. If we define foxing as the brown coloured spots on paper, in that case any type of brown spots will be termed as foxing. However, that may give rise to several misconceptions. Various studies were conducted to tackle this problem like, X-ray spectral analysis, T.L.C. analysis, Optical photomicrography, observation of spots under U.V. light, etc. (Thomas and Beckwith, 1935 ; Beckwith *et al.*, 1940 ; Ambler and Fenney, 1957 ; Chalk, 1960 ; Carter 1972 ; Baynes Cope

1976; Press 1976; Meynall and Newsman, 1978, 1979; Cain and Miller, 1982; Lea Nol *et al.*, 1983; Hey, 1983).

After going through the literature, it can be concluded that there are two schools of thoughts :—

1. The presence of iron, or similar metals either as concentration of salts or as an actual speck or particle present in the paper.
2. Fungal infection is the main cause of foxing.

The role of fungal activity and iron in the development of foxing is not very clear yet. In our opinion there may be some other reason which have not been so far pointed out. Thus study of foxing needs a continuous and thorough research.

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